

# Optimizing the Operation of Direct-Flow Hollow Fibre Filtration



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A thesis submitted for the degree of  
*Doctor of Philosophy*

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## **Statement of Originality**

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## Abstract

In current water industry, direct-flow hollow fibre filtration is increasingly used as an intermediate or complete water purification technique. Compare to classical crossflow filtration, direct-flow filtration avoids high pressure drop that incurs high energy cost in the feed channel and permits the use of smaller diameter fibres which increases the filtration area per unit volume. Such filtration is achieved by efficiently capping one end of the crossflow device and it is usually operated in dead-end mode with regular intermittent backwash. Direct-flow filtration therefore forms the dominant method for producing potable water from ground or surface water. This thesis focuses on the operational considerations and design aspects of the direct-flow filtration membrane modules, specifically those of the pressure driven inside feed format. From a comprehensive literature survey of direct-flow filtration it was established that there was a general lack of information on direct-flow filtration behaviours. In particular there were no tools to simulate the hydrodynamic efficiencies within these devices. This study therefore set out to develop various direct-flow filtration models which one can also apply in various industrial processes. With a dimensionless mathematical model for the fluid flow in an axisymmetric direct-flow pipe, simple investigations on direct-flow filtration was achieved. Then two realistic hollow fibre models which included finite wall thicknesses and fouling effects were developed. These models were used to investigate various influences upon the axial dependence of the transmembrane pressure and the cumulative filtration volume under non-fouling and fouling

conditions. The current fouling model is limited to chemically inert, incompressible, suspended solids. Geometrical parameters including fibre radius, fibre wall thickness, fibre length, module void fraction and imposed average flux were varied over a broad range values encompassing the values applied in today's water industry. Various scenarios of membrane fouling and cleaning process in which fibres were subject to identical hydrodynamic conditions have been investigated. The study has proposed two important metrics and a strategy for optimizing them to promote the best filtration performance: the module productivity parameter, and the filtration uniformity factor. Overall, optimized design ranges and operating regimes of the direct-flow hollow fibre filtration devices were summarised, and the modelling results accord with commercial products as used in industrial practice that has evolved over the last 20 years and show guidance for future designs.

To My Parents

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# Contents

|                                                                   |          |
|-------------------------------------------------------------------|----------|
| <b>Chapter 1 Introduction .....</b>                               | <b>1</b> |
| 1.1 Motivation and objectives .....                               | 1        |
| 1.2 Contribution.....                                             | 2        |
| 1.3 Thesis outline.....                                           | 3        |
| <b>Chapter 2 Overview of Membrane Filtration .....</b>            | <b>6</b> |
| 2.1 History of membrane filtration.....                           | 7        |
| 2.2 Membrane processes and applications .....                     | 9        |
| 2.2.1 Membrane based gas separation .....                         | 9        |
| 2.2.2 Pressure-driven membrane processes .....                    | 10       |
| 2.2.3 Membrane bioreactors (MBRs).....                            | 12       |
| 2.2.4 Membrane contactors.....                                    | 13       |
| 2.2.5 Ion exchange membrane processes: electrodialysis .....      | 13       |
| 2.2.6 Others.....                                                 | 14       |
| 2.3 Membrane applications in water industry .....                 | 15       |
| 2.3.1 Desalination.....                                           | 15       |
| 2.3.2 Wastewater treatment and reuse .....                        | 16       |
| 2.3.3 Drinking water treatment.....                               | 17       |
| 2.4 Membrane materials, modules and filtration fundamentals ..... | 18       |
| 2.4.1 Membrane materials.....                                     | 18       |
| 2.4.2 Membrane modules .....                                      | 21       |
| 2.4.3 Membrane filtration fundamentals.....                       | 23       |

|                                                                    |                                                     |           |
|--------------------------------------------------------------------|-----------------------------------------------------|-----------|
| 2.5                                                                | Membrane fouling and cleaning.....                  | 26        |
| 2.5.1                                                              | Membrane fouling.....                               | 26        |
| 2.5.2                                                              | Fouling mechanisms .....                            | 28        |
| 2.5.3                                                              | Cleaning processes .....                            | 29        |
| 2.6                                                                | Membrane flow configurations .....                  | 31        |
| 2.6.1                                                              | Dead-end filtration .....                           | 32        |
| 2.6.2                                                              | Crossflow filtration .....                          | 32        |
| 2.6.3                                                              | Direct-flow filtration.....                         | 33        |
| 2.7                                                                | Conclusion.....                                     | 35        |
| <b>Chapter 3 Literature Review of Direct-Flow Filtration .....</b> |                                                     | <b>36</b> |
| 3.1                                                                | Direct-flow filtration fundamentals.....            | 37        |
| 3.1.1                                                              | Module formats.....                                 | 37        |
| 3.1.2                                                              | Operating modes.....                                | 42        |
| 3.1.3                                                              | Membrane materials and module designs.....          | 46        |
| 3.2                                                                | Fouling and cleaning in direct-flow filtration..... | 48        |
| 3.2.1                                                              | Fouling in direct-flow filtration.....              | 48        |
| 3.2.2                                                              | Cleaning processes in direct-flow filtration.....   | 48        |
| 3.2.2.1                                                            | Physical cleaning .....                             | 49        |
| 3.2.2.2                                                            | Chemical cleaning .....                             | 51        |
| 3.3                                                                | Module optimization .....                           | 52        |
| 3.4                                                                | 2D direct-flow channel.....                         | 57        |
| 3.5                                                                | Conclusion.....                                     | 65        |
| <b>Chapter 4 Simple Direct-Flow Filtration Devices .....</b>       |                                                     | <b>66</b> |

|                                                                    |                                                             |           |
|--------------------------------------------------------------------|-------------------------------------------------------------|-----------|
| 4.1                                                                | Introduction .....                                          | 67        |
| 4.2                                                                | Model development: direct-flow pipe.....                    | 67        |
| 4.3                                                                | Discussion of results and optimization .....                | 71        |
| 4.3.1                                                              | Pressure and TMP profiles .....                             | 72        |
| 4.3.2                                                              | Cumulative fluid volume .....                               | 76        |
| 4.3.3                                                              | Filtration uniformity and productivity .....                | 78        |
| 4.4                                                                | Conclusion.....                                             | 84        |
| <b>Chapter 5 Direct-Flow Hollow Fibre Filtration Modules .....</b> |                                                             | <b>86</b> |
| 5.1                                                                | Introduction .....                                          | 87        |
| 5.2                                                                | Filtration parameters.....                                  | 88        |
| 5.3                                                                | Model development .....                                     | 89        |
| 5.3.1                                                              | Setup .....                                                 | 89        |
| 5.3.2                                                              | Lubrication equations.....                                  | 92        |
| 5.4                                                                | Results and discussion .....                                | 96        |
| 5.4.1                                                              | Influence of fibre spacing on axial pressure profiles ..... | 96        |
| 5.4.2                                                              | Selection of design parameters.....                         | 100       |
| 5.4.2.1                                                            | Influence of void fraction.....                             | 101       |
| 5.4.2.2                                                            | Influence of fibre inner radius .....                       | 103       |
| 5.4.2.3                                                            | Influence of fibre wall thickness .....                     | 105       |
| 5.4.2.4                                                            | Influence of fibre length .....                             | 107       |
| 5.4.2.5                                                            | Influence of imposed average flux.....                      | 111       |
| 5.4.3                                                              | Filtration performance .....                                | 113       |
| 5.4.4                                                              | Taguchi method.....                                         | 116       |
| 5.5                                                                | Conclusion.....                                             | 119       |

## **Chapter 6 Time Evolution of Fouling Development in Direct-Flow Filtration..... 120**

|       |                                                                               |     |
|-------|-------------------------------------------------------------------------------|-----|
| 6.1   | Introduction .....                                                            | 121 |
| 6.2   | Model development .....                                                       | 121 |
| 6.2.1 | Setup .....                                                                   | 121 |
| 6.2.2 | Cake resistance of fouling development .....                                  | 125 |
| 6.3   | Filtration parameters.....                                                    | 126 |
| 6.4   | Results and discussion .....                                                  | 127 |
| 6.4.1 | Influence of backwash cleaning .....                                          | 127 |
| 6.4.2 | Influence of imposed average flux .....                                       | 132 |
| 6.4.3 | Influence of fraction of flux and crossflow effects .....                     | 136 |
| 6.4.4 | Influence of backwash cleaning frequency .....                                | 138 |
| 6.4.5 | Influence of fibre geometry (void fraction, fibre length, fibre radius) ..... | 140 |
| 6.5   | Conclusion.....                                                               | 143 |

## **Chapter 7 Concluding Remarks and Future Work..... 144**

|     |                          |     |
|-----|--------------------------|-----|
| 7.1 | Concluding Remarks ..... | 144 |
| 7.2 | Future Work.....         | 146 |

## **Appendix-A Direct-Flow Filtration in PDO format ..... 148**

|     |                                         |     |
|-----|-----------------------------------------|-----|
| A.1 | Introduction.....                       | 149 |
| A.2 | PDO model development .....             | 150 |
| A.3 | Flow in PDO direct-flow filtration..... | 154 |
| A.4 | Practical PDO model development.....    | 157 |

## **Appendix-B Supplementary of Productivity Parameter ( $\beta$ ) ..... 158**

**Appendix-C Publications and Presentations ..... 160**

    A.    Publications..... 160

    B.    Conference Presentations ..... 161

**Nomenclature ..... 162**

**Abbreviations ..... 167**

**Bibliography ..... 169**

# List of Figures

|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |    |
|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| Figure 2.1: Membrane pore size and particle ranges of the main membrane filtration processes (MF, NF, UF, RO). .....                                                                                                                                                                                                                                                                                                                                                                                | 10 |
| Figure 2.2: Particle fouling mechanisms in membrane filtration. ....                                                                                                                                                                                                                                                                                                                                                                                                                                | 28 |
| Figure 2.3: Schematics of (a) dead-end, (b) crossflow and (c) direct-flow filtration. Please note in particular for (b) and (c) aspect ratio (length/radius width) is not to scale. ....                                                                                                                                                                                                                                                                                                            | 31 |
| Figure 3.1: Process flows of the two formats as seen in cross-section of the hollow fibre modules. ...                                                                                                                                                                                                                                                                                                                                                                                              | 39 |
| Figure 3.2: Schematic of the flow within a single direct-flow 2D channel. Solid lines indicate impermeable walls, blue dashed lines indicate permeable walls, and the dotted line indicates symmetry in the flow. Flow enters from the bottom and passes through the permeable walls. Beyond the capped end, the fluid continues to an outlet following the black arrows. The inside of the membrane-walled channel is referred as Region 1, while that outside is Region 2 (Herterich, 2015). .... | 59 |
| Figure 4.1: Schematic of the flow within a single 3D direct-flow axisymmetric pipe. Red dashed lines indicate the permeable walls, the black dashed lines indicate the ‘symmetry’ between two pipes, and the hashed areas indicate non-permeable areas. Flow enters from the lumen side and passes through the permeable walls. Beyond the capped-end, the fluid continues to an outlet following the blue arrows. ....                                                                             | 68 |
| Figure 4.2: Variation of fluid pressure in the lumen side, $p$ , and in the shell side, $q$ , under the condition of permeability $k = 1$ and the flux $Q = 1$ , with dimensionless module spacing parameters (a) $\ell = 1$ and (b) $\ell = 10$ . ....                                                                                                                                                                                                                                             | 73 |
| Figure 4.3: Profiles of transmembrane pressure difference (TMP), in a capped-end axisymmetric pipe. Variation with (a) module spacing parameter, $\gamma$ (with $Q = 1, k = 1$ ); (b) flux, $Q$ (with $\gamma = 1, k = 1$ ); and (c) permeability, $k$ , (with $\gamma = 1, Q = 1$ ). ....                                                                                                                                                                                                          | 74 |

|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |    |
|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| Figure 4.4: The cumulative fluid volume transported through the fibre walls, $V_c(\%)$ , versus axial distance, $z$ , in (a) a 2D channel (as shown in Section 3.4) and (b) an axisymmetric pipe for various values of module spacing parameters ( $\ell$ and $\gamma$ respectively). In both cases, the varying quantity assumes the values 0.1, 0.5, 1, 1.5 and 5, respectively at $Q = 1$ and $k = 1$ . (Optimal performance would correspond to a linearly increasing cumulative flux, as shown by the straight black line.) ..... | 77 |
| Figure 4.5: The normalized module productivity, $\alpha$ , versus different values of dimensionless module spacing parameters (a) $\ell$ in a 2D channel (as shown in Section 3.4) and (b) $\gamma$ in an axisymmetric pipe respectively. In both cases, $Q = 1$ and $k = 1$ . .....                                                                                                                                                                                                                                                   | 79 |
| Figure 4.6: Illustration of the filtration uniformity parameter definition according to the cumulative fluid volume line (The $V_{actual}$ line can be below the diagonal and also be 'S' shaped, as shown later in Figure 4.8). .....                                                                                                                                                                                                                                                                                                 | 80 |
| Figure 4.7: The measure of the uniformity of the overall filtration, $\varphi$ , versus different values of module spacing parameters (a) $\ell$ in a 2D channel (as shown in Section 3.4) and (b) $\gamma$ in an axisymmetric pipe respectively. In both cases, the filtration is setup at $Q = 1$ and $k = 1$ . .....                                                                                                                                                                                                                | 82 |
| Figure 4.8: (a) The cumulative fluid volume transported through the fibre walls (%) versus axial distance, $z$ , in an axisymmetric pipe with different values of permeability $k$ ; (b) The measure of the uniformity of the overall filtration, $\varphi$ , in a pipe versus permeability $k$ . In (a), the varying quantity assumes the values 0.1, 0.5, 1, 1.5, 5, and 10, respectively (the black dashed line depicts the optimal filtration case). In both cases, the filtration is setup at $Q = 1$ and $\gamma = 1$ . .....    | 83 |
| Figure 5.1: Schematic of capped-ended hollow fibre membrane module. Please note aspect ratio (length/fibre diameter) is not to scale. ....                                                                                                                                                                                                                                                                                                                                                                                             | 90 |
| Figure 5.2: Schematic of a single fibre as used in model development (Section 5.3) (a) cross sectional view; (b) overall side view with flow direction indicated. The aspect ratio (length/fibre diameter) is not to scale. ....                                                                                                                                                                                                                                                                                                       | 91 |

Figure 5.3: Variation of fluid pressure in the lumen side,  $p$ , and in the shell side,  $q$ . Fixed parameters:

$a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70\text{ L m h}$  and  $km = 300\text{ L m h/bar}$  with spacing parameters (a)  $\gamma = 0.5$ , (b)  $\gamma = 1$  and (c)  $\gamma = 10$ . .....97

Figure 5.4: The effect of spacing parameter,  $\gamma$ , upon the transmembrane pressure difference (TMP)

profiles versus axial distance,  $z$  at different spacing parameter  $\gamma$ . Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70\text{ L m h}$  and  $km = 300\text{ L m h/bar}$ . .....98

Figure 5.5: The effect of spacing parameter,  $\gamma$ , upon the cumulative volume transported through the

fibre walls,  $V_c$  (%), versus axial distance,  $z$ . Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ , and  $J_{av} = 70\text{ L m h}$  with permeances (a)  $km = 300\text{ L m h/bar}$ , (b)  $km = 1150\text{ L m h/bar}$ , (c)  $km = 11500\text{ L m h/bar}$ , and (d)  $km = 115000\text{ L m h/bar}$ . .....99

Figure 5.6: The effect of void fraction,  $\epsilon$ , upon (a) the transmembrane pressure difference (TMP)

profiles and (b) the cumulative volume transported through the fibre walls,  $V_c$  (%), versus axial distance,  $z$ . Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70\text{ L m h}$  and  $km = 300\text{ L m h/bar}$ . .....102

Figure 5.7: The effect of inner radius,  $a$ , upon (a) the transmembrane pressure difference (TMP) profiles

and (b) the cumulative volume transported through the fibre walls,  $V_c$  (%), versus axial distance,  $z$ . Fixed parameters:  $\epsilon = 0.45$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70\text{ L m h}$  and  $km = 300\text{ L m h/bar}$ . .....104

Figure 5.8: The effect of wall thickness,  $\Gamma$ , upon (a) the transmembrane pressure difference (TMP)

profiles and (b) the cumulative volume transported through the fibre walls,  $V_c$  (%), versus axial distance,  $z$ . Fixed parameters:  $a = 0.45\text{mm}$ ,  $\epsilon = 0.45$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70\text{ L m h}$  and  $km = 300\text{ L m h/bar}$ . .....106

Figure 5.9: The effect of fibre length,  $L$ , upon (a) the transmembrane pressure difference (TMP) profiles

and (b) the cumulative volume transported through the fibre walls,  $V_c$  (%), versus percentage of the fibre length at different fibre length,  $L$  (%). Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $\epsilon = 0.45$ ,  $J_{av} = 70\text{ L m h}$  and  $km = 300\text{ L m h/bar}$ . .....108

Figure 5.10: The effect of fibre length,  $L$ , upon the transmembrane pressure difference (TMP) profiles versus percentage of the fibre length at different fibre length,  $L$  (%). Fixed parameters:  $\Gamma = 0.25\text{mm}$ ,  $J_{av} = 70\text{ L m h}$  and  $km = 300\text{ L m h/bar}$ . (a)  $a = 0.35\text{mm}$ ,  $\epsilon = 0.4$ , (b)  $a = 0.35\text{mm}$ ,  $\epsilon = 0.5$ , (c)  $a = 0.4\text{mm}$ ,  $\epsilon = 0.4$ , (d)  $a = 0.4\text{mm}$ ,  $\epsilon = 0.5$ , (e)  $a = 0.45\text{mm}$ ,  $\epsilon = 0.4$ , (f)  $a = 0.45\text{mm}$ ,  $\epsilon = 0.5$ . .....110

Figure 5.11: The effect of imposed average flux,  $J_{av}$ , upon the transmembrane pressure difference (TMP) profiles versus percentage of the fibre length at different fibre length,  $L$  (%). Except where indicated standard parameters were as follows:  $a = 0.3\text{ mm}$ ,  $\epsilon = 0.35$ ,  $\Gamma = 0.25\text{ mm}$ ,  $L = 1.6\text{ m}$  and  $km = 300\text{ L m h/bar}$ . .....112

Figure 5.12: The normalized module productivity,  $\beta$ , and the uniformity of the filtration behaviour,  $\varphi$ , of the filtration process versus the design control parameters. (a) void fraction,  $\epsilon$ , (b) inner radius,  $a$ , (c) fibre length,  $L$ , and (d) imposed average flux,  $J_{av}$ , respectively. Standard parameters:  $a = 0.4\text{ mm}$ ,  $\epsilon = 0.45$ ,  $\Gamma = 0.25\text{ mm}$ ,  $L = 1.6\text{ m}$ ,  $J_{av} = 70\text{ L m h}$  and  $km = 300\text{ L m h/bar}$ . .....113

Figure 6.1: 3D Schematic flow regimes (n sections) of direct-flow filtration in a single hollow fibre membrane in PDI filtration mode. ....122

Figure 6.2: (a) The normalized module productivity,  $\beta$ , and (b) the uniformity of the filtration behaviour,  $\varphi$ , of the filtration process versus time,  $t$ , under fouling conditions with or without backwash cleaning. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65\text{ mm}$ ,  $a = 35b$ ,  $L = 1.6\text{ m}$ ,  $J_{av} = 70\text{ L m h}$ ,  $km = 300\text{ L m h/bar}$ , and  $tc = 30\text{ mins}$ . .....129

Figure 6.3: (a) The overall resistance,  $R$ , and (b) the pump driving pressure,  $Pp$ , of the filtration process versus time,  $t$ , under fouling conditions with or without backwash cleaning. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65\text{ mm}$ ,  $a = 35b$ ,  $L = 1.6\text{ m}$ ,  $J_{av} = 70\text{ L m h}$ ,  $km = 300\text{ L m h/bar}$ , and  $tc = 30\text{ mins}$ . .....131

Figure 6.4: (a) The normalized module productivity,  $\beta$ , and (b) the uniformity of the filtration behaviour,  $\varphi$ , of the filtration process versus time,  $t$ , under fouling conditions with backwash cleaning

|                                                                                                                                                                                                                                                                                                                                                                                                                             |     |
|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| incorporated. Standard parameters: $\epsilon = 0.4$ , $b = 0.65$ mm, $a = 35b$ , $L = 1.6$ m, $km = 300$ L m h/bar, and $tc = 30$ mins. ....                                                                                                                                                                                                                                                                                | 134 |
| Figure 6.5: (a) The normalized module productivity, $\beta$ , and (b) the uniformity of the filtration behaviour, $\varphi$ , of the filtration process versus processed fluid feed, $Vh$ , at various imposed average values under fouling conditions with backwash cleaning incorporated. Standard parameters: $\epsilon = 0.4$ , $b = 0.65$ mm, $a = 35b$ , $L = 1.6$ m, $km = 300$ L m h/bar, and $tc = 30$ mins. ....  | 135 |
| Figure 6.6: The normalized module productivity, $\beta$ , and the uniformity of the filtration behaviour, $\varphi$ , of the filtration process versus imposed average flux, $Jav$ , under fouling conditions with backwash cleaning incorporated. Standard parameters: $\epsilon = 0.4$ , $b = 0.65$ mm, $a = 35b$ , $L = 1.6$ m, $km = 300$ L m h/bar, $tc = 30$ mins, and $Vh = 32$ m <sup>3</sup> /m <sup>2</sup> ..... | 136 |
| Figure 6.7: (a) The normalized module productivity, $\beta$ , and (b) the uniformity of the filtration behaviour, $\varphi$ , of the filtration process versus time at various $rf$ ratios. Standard parameters: $\epsilon = 0.4$ , $b = 0.65$ mm, $a = 35b$ , $L = 1.6$ m, $Jav = 70$ L m h, $km = 300$ L m h/bar, and $tc = 30$ mins. ....                                                                                | 137 |
| Figure 6.8: (a) The normalized module productivity, $\beta$ , and (b) the uniformity of the filtration behaviour, $\varphi$ , of the filtration process versus time at various backwash cleaning frequencies. Standard parameters: $\epsilon = 0.4$ , $b = 0.65$ mm, $a = 35b$ , $L = 1.6$ m, $Jav = 70$ L m h, and $km = 300$ L m h/bar. ....                                                                              | 139 |
| Figure 6.9: (a) (c) (e) The normalized module productivity, $\beta$ , and (b) (d) (f) the uniformity of the filtration behaviour, $\varphi$ , of the filtration process versus time at various void fraction, fibre diameter and fibre lengths. Standard parameters: $\epsilon = 0.4$ , $b = 0.65$ mm, $a = 35b$ , $L = 1.6$ m, $Jav = 70$ L m h, $km = 300$ L m h/bar, and $tc = 30$ mins. ....                            | 141 |
| Figure A.1: Schematic of process flows of the two formats of direct-flow filtration illustrated for a single fibre. (a) Pressure driven inside feed (PDI) format; (b) Pressure driven outside feed (PDO) format.....                                                                                                                                                                                                        | 149 |

|                                                                                                                                                                                                                                                                                                                                                                                                |     |
|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| Figure A.2: Variation of PDO mode fluid pressure in the lumen side, $p$ , and in the shell side, $q$ . Fixed parameters: $a = 0.4\text{mm}$ , $\Gamma = 0.25\text{mm}$ , $L = 1.5\text{m}$ , $J_{av} = 70\text{ Lmh}$ and $km = 300\text{ Lmh/bar}$ with spacing parameters (a) $\gamma = 0.5$ , (b) $\gamma = 1$ and (c) $\gamma = 10$ . .....                                                | 154 |
| Figure A.3: The effect of void fraction, $\epsilon$ , upon (a) the transmembrane pressure difference (TMP) profiles and (b) the cumulative volume transported through the fibre walls, $Q_c$ (%), versus axial distance, $z$ in PDO mode. Fixed parameters: $a = 0.4\text{mm}$ , $\Gamma = 0.25\text{mm}$ , $L = 1.5\text{m}$ , $J_{av} = 70\text{ Lmh}$ and $km = 300\text{ Lmh/bar}$ . ..... | 156 |
| Figure A.4: 3D Schematic flow regimes (n sections) of direct-flow filtration in a single hollow fibre membrane in PDO filtration mode. ....                                                                                                                                                                                                                                                    | 157 |

# List of Tables

|                                                                                                                            |     |
|----------------------------------------------------------------------------------------------------------------------------|-----|
| Table 2.1: Summary of flow situations in three filtration modes .....                                                      | 34  |
| Table 3.1: Operating characteristics and limitations in inside and outside formats .....                                   | 41  |
| Table 3.2: Overview of filtration variations at constant flux and constant pressure operations .....                       | 44  |
| Table 3.3: Overview of optimal hollow fibre module design for PDI modules. ....                                            | 53  |
| Table 5.1: Summary of table of modelling results in this chapter. ....                                                     | 115 |
| Table 5.2: Control parameters and their industrially relevant range.....                                                   | 116 |
| Table 5.3: Control parameters and their levels used in this study.....                                                     | 117 |
| Table 5.4: L <sub>16</sub> (4 <sup>5</sup> ) Orthogonal Array of Taguchi research control parameters.....                  | 117 |
| Table 5.5: Control parameters and their linear regression of $\beta$ and $\varphi$ developed from Taguchi method.<br>..... | 118 |
| Table 6.1: Summary of investigation results in this thesis. ....                                                           | 142 |

# Chapter 1

## Introduction

### 1.1 Motivation and objectives

With the increase in world population and the extensive usage of industrial technology over past decades, there remains an unmet demand for drinking water and wastewater treatment (Aldhous, 2003). Membrane technologies can be widely applied in many parts of the water industries spanning over wastewater treatment, drinking water production, brackish water and seawater desalination. Thus membranes will continue to play an increasingly influential role in mitigating the world water crisis (Hoek and Tarabara, 2013). Studies on membrane-based filtration processes will therefore be a long-lasting topic in the field of water research.

In general, membrane filtration processes can be operated in three filtration formats: dead-end, crossflow, and direct-flow (Noble and Stern, 1995, Culfaz et al., 2011). Although the membrane industry was driven through the last century by crossflow filtration, the modern water membrane market is based on the use of direct-flow filtration (Pearce, 2011). Direct-flow filtration combines a filtration cycle operated in dead-end mode with a regular intermittent backwash. At an appropriate frequency the latter makes membrane fouling mainly reversible. As the development of the membrane market for direct-flow filtration is still at a relative young age, there still exists the need for research to understand and thereby provide guidelines for the operation and design of direct-flow filtration processes. Such guidelines need to be fully cognisant of the fouling issues. Therefore the work presented in this thesis focuses on understanding direct-flow filtration

processes, especially on how to optimize the filtration performance under various operating conditions and module designs.

## 1.2 Contribution

This thesis has developed various direct-flow filtration models that have elaborated our understanding of direct-flow filtration and which can also be applied in future membrane filtration studies. The main contributions include:

- Simple direct-flow filtration model for an axisymmetric system operated in the inside feed format under constant flux non-fouling conditions (Chapter 4).
- Realistic direct-flow filtration for hollow fibre membranes with inside feed operated under constant flux non-fouling conditions (Chapter 5).
- Practical direct-flow filtration at constant flux for hollow fibre membranes with inside feed that gives rise to fouling phenomenon (Chapter 6).
- Realistic direct-flow filtration for hollow fibre membranes with outside feed under constant flux with and without being subject to fouling conditions (Appendix-A).

This thesis has introduced two novel parameters by which to assess membrane filtration performances. Firstly, a uniformity parameter which is taken as a measure for a system to withstand irreversible fouling and secondly a productivity parameter. The value of the latter is self-evident and the basis of the former is that greater uniformity it is a proxy for a reduced tendency to irreversibly foul; greater uniformity will minimise high local fluxes and thus

minimise the chance of exceeding the threshold flux which would trigger irreversible fouling. These two parameters can be used as supportive measures in membrane module design. Herein design ranges for various fibre module geometries are given. The guidelines accord with the current industrial practice that has evolved over the last 20 years. Changes in membrane materials will give rise to new permeabilities which in turn will generate new optimal design ranges. The models developed herein will contribute to and provide a guidance for future membrane module designs especially those operated in direct-flow mode.

## **1.3 Thesis outline**

This thesis is outlined as follows:

Chapter 1 gives a brief introduction to the background of this research and outlines the aim and structure of the whole thesis.

Chapter 2 provides an overview of membrane filtration including the history of the technology development of the essential membrane processes. It introduces the basic principles and rules of membrane applications in drinking water treatment. General review and discussion on membrane fouling and cleaning approaches are also presented in Chapter 2. This Chapter shows the importance of membrane technologies and how direct-flow filtration has arisen within the membrane water treatment market.

Having given the overall background of the research field, Chapter 3 reviews specific background knowledge on direct-flow filtration particularly that concerned with design and operating problems. The chapter introduces different module formats, filtration operating modes,

membrane materials, module designs and cleaning methods in direct-flow filtration. Specifically, Chapter 3 summarises the research results from related direct-flow filtration studies in the existing literature.

Chapter 4 develops a simple direct-flow filtration model for an axisymmetric 3D system based on a capped permeable tube to describe the essential filtration behaviours of a direct-flow system. The model ignores the wall thickness effect so that a simple introductory investigation of direct-flow filtration can be achieved. The model developed in Chapter 4 will be presented in dimensionless format so that the results can be compared with an existing literature study for a 2D system.

Chapter 5 develops realistic direct-flow filtration models aiming to investigate the operational considerations and design aspects of the filtration. Geometrical parameters including fibre radius, fibre wall thickness, fibre length, module void fraction and imposed average flux effects will be studied over a broad range values encompassing the values applied in current membrane market. The chapter aims to provide a guidance on the optimized design ranges for direct-flow filtration modules.

Chapter 6 develops practical direct-flow filtration models aiming to investigate the filtration performance under fouling conditions. Direct-flow filtration performance is studied with particle deposition and backwash cleaning processes. The geometrical parameters studied in chapter 4 and 5 will be re-evaluated in chapter 6 so that fouling effects can be incorporated. Especially, the effect of imposed average flux on the filtration performance will be emphasised in this chapter.

Chapter 7 concludes the research and summarizes the implications for possible future works related to this subject.

Appendix-A develops the equations of an additional practical direct-flow filtration system, i.e. pressure driven outside feed. The contents provided will be useful for readers who want to extend the output of Chapter 5 and 6 and to compare the optimized design ranges of outside feed to those of inside feed. The practical models developed in both Chapter 6 (inside feed) and Appendix-A (outside feed) can also be used for direct-flow filtration under constant pressure operations. This thesis provides developed models for future works on direct-flow filtration with different feed formats (inside and outside feed) and under different operating conditions (constant flux and constant pressure).

## **Chapter 2**

# **Overview of Membrane Filtration**

This chapter gives an overview of membrane science and technology with an emphasis upon membrane applications in water treatment. Firstly it provides an overview of membrane technology development related to water treatment, gas separation, membrane reactors (MBRs), membrane contactors and electrochemical processes. It then particularly introduces the basic principles and rules of membrane applications in drinking water treatment, followed by the discussion of challenges inherent in membrane fouling and cleaning.

## **2.1 History of membrane filtration**

Membrane separation technologies play a central role in various industry sectors and scientific disciplines including indoor air filtration, food and beverage industries, drinking water purification, domestic and industrial waste water treatment, biotechnology, and pharmaceutical sciences (Pearce, 2011, Hoek and Tarabara, 2013, Ni Mhurchu and Foley, 2006). Back in 1748, the first observations of membrane phenomenon, osmosis, were made by French scientist Abbe Nollet (Nollet, 1784, Wang et al., 2008, Singh, 2014). Nollet demonstrated the concept of semi-permeability through observations on the permeation of water through pig bladders and his original paper was reproduced in 1995 (Nollet, 1995). Through the 19<sup>th</sup> and early 20<sup>th</sup> centuries, membranes had no industrial or commercial uses, and ultrafiltration (UF) and microfiltration (MF) membranes were produced primarily for laboratory separations to understand the phenomena involved in interfacial and ideal liquid and gas systems (Baker, 2004). In the early 1900s, Bechhold devised a new technique to prepare nitrocellulose membranes of graded pore sizes (Bechhold, 1907). This technique was gradually improved by other researchers (Zsigmondy and Bachmann, 1918, Ferry, 1936, Elford, 1937) and after the early 1930s microporous collodion membranes became commercially available (Baker, 2012). In the next 20 years, this early microfiltration membrane technology was then gradually expanded to other polymers, notably cellulose acetate (Baker, 2012).

In the second half of the 20<sup>th</sup> century, the membrane industry underwent dramatic evolution with its units displacing some traditional separation processes, capitalizing on the higher separation efficiency, lower use of energy, smaller footprint, and modular design of membrane systems (Doshi et al., 1977, Zondervan and Roffel, 2008, Vera et al., 2015). Historically, commercial ion exchange membranes were developed in 1940s (Murphy, 1943, Kressman, 1950, Juda and

McRae, 1950), electro-dialysis systems were developed in 1950s (Mason and Kirkham, 1959), cellulose acetate films were first used to desalinate water in 1959 (Reid and Breton, 1959), sea salt production facilities were first employed in 1961 (Woodcock and Spencer, 1961), integrally skinned asymmetric cellulose acetate membranes were developed for seawater desalination in 1963 (Loeb and Sourirajan, 1963), thin-film composite membranes were developed in 1975 (Cadotte, 1975), the first reverse osmosis (RO) seawater desalination facilities were innovated in 1979 and opened in 1982 (Muirhead et al., 1982), and nanofiltration (NF) was developed as a new low energy purification tool for the removal of colour, hazardous organic contaminants and hardness from drinking water sources in the second half of the 1980s (Watson and Hornburg, 1989). In the late 1980s, low-pressure, submerged air-sparged membranes were developed (Yamamoto et al., 1989). Later in the mid-1990s, economical microfiltration/ultrafiltration systems for the treatment of municipal water sources and for the application of membrane bioreactors in sewage treatment plants has developed (Baker, 2004). Since then, membrane applications in the treatment of municipal water has become one of the most rapidly growing membrane technologies. Membrane applications in biological areas also become competitive in comparison with conventional biological treatment in terms of cost and product quality (Baker, 2012).

The revolution of membrane industry over the last century laid the foundation for modern membrane science and technology development in membrane-based gas separation, pressure driven membrane processes, membrane bioreactors (MBRs), and membrane contactors. These membrane applications ultimately play an increasingly influential role in mitigating the world water crisis and addressing the global challenge for energy and power supply.

## **2.2 Membrane processes and applications**

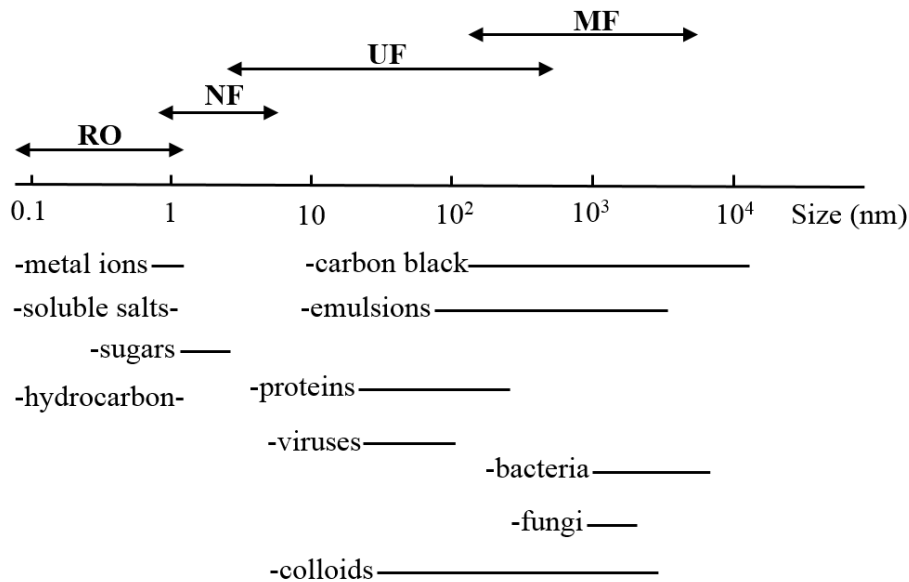
### **2.2.1 Membrane based gas separation**

The earliest large-scale application of membrane-based gas separations was originally applied in the hydrogen recovery process from ammonia purge gases in the 1970s (Ockwig and Nenoff, 2007, Bernardo et al., 2009, Bernardo and Clarizia, 2013). Since then, membrane technologies have driven much simpler, more efficient gas separation devices in the industry while rapidly shifting medium and large-scale membrane-based gas separation processes to on-site gas productions (Baker et al., 1991, Baker, 2000, Baker, 2004). The present day membrane based gas separation processes can be broadly applied to treat flue gas, syngas, and other off-gases for emissions control and carbon dioxide capture (Hoek and Tarabara, 2013).

During membrane-based gas separations, specific gas molecules permeate more selectively from a mixture of gases through the membrane. Since the processes are usually driven by the pressure differences between the two sides of the membrane without phase change, membrane-based gas separation consumes less energy than other gas separation process (Nunes and Peinemann, 2001). A typical membrane gas separation system includes just a blower, a membrane module, and a vacuum pump or a compressor (Baker, 2004). Due to strong advantages, membrane-based gas separation processes have grown fast in some parts of the gas separation market in the past three decades, examples include hydrogen separation in industry, nitrogen production for inert gas applications, oxygen enrichment in medical treatment, and large-scale industrial processes such as natural gas purifications (Hoek and Tarabara, 2013).

## 2.2.2 Pressure-driven membrane processes

Since the 1980s, pressure-driven membrane processes have been widely applied in the water treatment market following in the steps of rapid developments in polymer science (Zondervan and Roffel, 2008, Vera et al., 2015). These membrane separations based on hydrostatic pressure driving forces can be classified in four main processes: MF, UF, NF and RO (Scott and Hughes, 2012). The distinction between these processes depends on the size, shape, and charge of the particles and dissolved components (partially) retained by the membrane, which range from suspended particles in MF to small molecules and ions in RO (Van der Bruggen et al., 2003). The overall membrane pore size distribution, and its membrane selectivity with respect to particles retained, for the main types of pressure-driven membrane filtration processes (MF, UF, NF, and RO) is illustrated in Figure 2.1.



**Figure 2.1:** Membrane pore size and particle ranges of the main membrane filtration processes (MF, NF, UF, RO).

MF membranes have the largest pores, 0.1-10  $\mu\text{m}$ , and the highest permeability which permits a sufficient flux even at a low pressure (Van der Bruggen et al., 2003, Baker, 2004). MF were historically applied in various micro range applications, including the retention and concentration of microorganisms, cellular fragments, fine precipitates and other particles (Le, 1987, Gatenholm et al., 1988, Patel et al., 1987, Hoffmann et al., 1987, Lafforgue et al., 1987, Le et al., 1984, Bentham et al., 1988). UF membranes have smaller pores, 2-100 nm, and the permeability is considerably lower than in MF, and consequentially require higher pressure during in the operations (Van der Bruggen et al., 2003, Foley, 2013). UF can be applied to purify larger molecules like proteins either in solution or bound to affinity polymers, and smaller molecules such as residual substrates and metabolites (Bautista et al., 1986, Larsson and Mattiasson, 1984, Mattiasson and Ling, 1986, Nakao et al., 1986). NF membranes have even smaller pore sizes, typically around 1 nm, which are used in the removal of relatively small organics (Van der Bruggen et al., 2003). RO membranes are usually dense membranes without predefined pores, which results in solution-diffusion mechanism and slower permeations (Van der Bruggen et al., 2003, Santos et al., 1991). During water treatment processes, MF and UF were usually applied for pre-treatment, NF is used for softening and removal of dissolved metals and organic molecules, and RO is for desalination. In addition to the desalination process, RO is also used for ultrapure water purification, i.e. the electrical driven process of brackish water become more competitive with RO treatment (Hoek and Tarabara, 2013). RO applications in the high technology sector for ultrapure water production will be discussed in detail in Section 2.3.

### **2.2.3 Membrane bioreactors (MBRs)**

In the late 1960s, the first membrane bioreactor system was installed by Dorr-Oliver Inc. combining a conventional activated sludge process with flat sheet ultrafiltration plate and frame membrane (Bemberis et al., 1971). In the 1970s and 1980s, most of the MBR systems were used for industry water treatment and its progress was only considerable in Japan (Radjenović et al., 2008). With the development of submerged membranes, the application of MBRs in the treatment of municipal wastewater has increased (Yamamoto et al., 1989) and the MBR market has experienced accelerated growth since 1990s (Judd, 2010). The commercial significance of the MBRs technology is consideration with a predicted market of US\$ 3.44 billion in 2018 (Basile et al., 2015).

MBRs technology combines biological-activated sludge process and membrane filtration in a system, whereas conventional treatment cannot cope with either its efficiency or effluent quality in wastewater treatment applications (Radjenović et al., 2008, Judd, 2016). Conventional treatment of municipal wastewater usually includes sedimentation of solids, aerobic degradation of organic matter, and secondary sedimentation to remove biomass. Unlike the three-stage process, MBRs can replace the two physical separation process by the involvement of a membrane so that the biomass can be physically separated from the treated water (Judd, 2008). The use of MBRs for wastewater treatment applications has a number of advantages: higher effluent quality, small footprint, lower sludge production and most importantly obviates a further tertiary disinfection process (Hoek and Tarabara, 2013).

## **2.2.4 Membrane contactors**

Membrane contactors are devices for gas/liquid or liquid/liquid mass transfer, wherein an interface between the phases is created without dispersion of one phase into the other (Wiesler, 1996, Gabelman and Hwang, 1999, Roizard, 2016). The key difference with respect to other membrane processes is that the membrane in membrane contactors is used only as an interface and has no selective property towards specific species (Hoek and Tarabara, 2013). With the combination of a microporous and generally hydrophobic membrane (rarely hydrophilic membranes), membrane contactors provide better performance both in efficiency and capacity than conventional dispersed phase contactors (Gabelman and Hwang, 1999).

Membrane distillation, as one of the membrane contactors processes, has gained significant attraction both in industry and academic research in recent years (Drioli et al., 2015). During the thermally-driven separation process of membrane distillation, only vapour molecules can pass through the porous hydrophobic membrane. This provides lower operating temperatures, lower hydrostatic pressure, and simply operations in comparison to conventional processes (Alkhudhiri et al., 2012). Developing applications of membrane distillation in present industry consist of seawater desalination, wastewater treatment, separation of volatile compounds, concentration of non-volatile compounds and processing of dairy fluids (Wang and Chung, 2015).

## **2.2.5 Ion exchange membrane processes: electrodialysis**

Electrodialysis is another widely applied membrane process. Charged ion selective membranes are used to separate ions from solutions using the driving force of electrical potential difference

(Baker, 2012). As early as 1890s, ion exchange membranes were described but had only poor mechanical and chemical properties (Ostwald, 1890, Donnan, 1911). Then other researchers developed selective anion- and cation- exchange membranes and multicell arrangement of electrodes to remove ions from solutions (Manegold and Kalauch, 1939, Meyer and Straus, 1940). Later in the early 1970s, electrodialysis polarity reversal system was designed so that the membrane electrodes automatically reversed and this gave higher efficiency (Siwak, 1993). Since then, electrodialysis has gradually developed in industrial scale for desalination of brackish groundwater and the production of boiler feed water (Seko et al., 1983, Association, 1995). Today electrodialysis is now a mature technology in the electrolysis process markets and its application in industrial process streams, food (cheese) processing, and wastewater treatment systems will continue to develop in the next decades (Baker, 2012).

## **2.2.6 Others**

In biotechnology and medicine industry, membranes have been used in drug delivery control to ensure slow release of pharmaceuticals from reservoir into patient bloodstream since 1970s (Sears, 1979). In the food and beverage industry, membranes are incorporated in the process of producing pure water, pure fluid streams, soft drinks and meat packaging materials (Hoek and Tarabara, 2013). In industrial dehydration of organic solvents, membranes are used in pervaporation processes to generate vapour pressure differences (Baker, 2012). Membrane-based processes in batteries, fuel cells, and osmotic energy recovery also contribute to our modern science and technology development (Ni Mhurchu and Foley, 2006).

## **2.3 Membrane applications in water industry**

### **2.3.1 Desalination**

With the increase of population and increasing industrialisation, the global water usage is around 9 trillion cubic meters in 2017 and is expected to an annual increase at approximately 60 billion cubic meters (Herndon, 2017). However, the global water reserve reveals only 2.5% of earth's total stock of water is fresh water and the rest is saline (Le and Nunes, 2016). As a result, only 0.007% of the global water in the earth is applicable for direct use (Afgan, 2008, Le and Nunes, 2016). Membrane technologies, which can be applied to produce potable water from alternative water sources such as seawater, brackish water, and wastewater to drinking water standards, therefore play an influential role in mitigating the world water crisis.

Among all the emerging membrane technologies, desalination play an important role in water sustainability. Besides seawater desalination there are applications in produced water treatment from oil and gas industries, desalted water for agriculture, desalination in mining industry, heavy metal removal from saline wastewater (Le and Nunes, 2016). Since 2000, massive effort has been put into seawater desalination plants in more than 100 countries to address the world water crisis (Mickley, 2012, Leven, 2013). Saudi Arabia paves the way for potable water supply by obtaining roughly 24 million cubic meters desalination water a day from the Persian Gulf, Qatar and Kuwait obtain 100% of their water supply for domestic and industrial by desalination, and Singapore has a second large seawater RO desalination plant in construction to meet nearly 30% of its potable water usage (Ghaffour, 2009, Hoek and Tarabara, 2013, Sun, 2016, Le and Nunes, 2016, Herndon, 2017). With a record of 19,372 (2017) versus 18,983 (2016) desalination plants in operation or in construction throughout the world producing potable water (Tonner, 2016,

IDA, 2017), membrane technology impact billion lives worldwide (Mickley, 2012, Leven, 2013). The desalination plants are mostly located in the Middle East or North Africa, using their petroleum resources to offset limited water resources (Schenkeveld et al., 2004). The world's largest seawater RO desalination plant is in Ashkelon, Israel, achieves a production of 624,000 m<sup>3</sup> water every day (Fritzmann et al., 2007).

### **2.3.2 Wastewater treatment and reuse**

In wastewater treatment processes, perspective membrane applications have been combined within the traditional biological wastewater treatment process since 1960s (Hoek and Tarabara, 2013). For municipal wastewater, the combination of MBRs with either a MF or UF membrane in the conventional treatment process can remove the biodegradable compounds and limited chemicals efficiently. For industrial wastewater, conventional water treatment technologies or MBR alone normally cannot remove all of the toxic chemicals. Especially, heavy metals, rare earth elements and trace organic contaminants require specific membrane processes. For the removal and recovery of heavy metals and rare earth elements, UF, NF and RO all have proven their competitiveness since they all have low energy and high selectivity advantages (Kurniawan et al., 2006, Abhanga et al., 2013). UF separates macromolecules (5 – 20 nm) at moderate transmembrane pressure conditions, RO recovers more than 99% for Cu (II), Ni (II), Zn (II) and As (V) at very high pressure operations, and RO provides an intermediate treatment process between UF and RO intermediate pressure operations (Le and Nunes, 2016). For trace organic contaminant, the standalone processes used in wastewater treatment all have certain advantages

and disadvantages, therefore a combination of MBRs with other membrane processes such as NF, RO and forward osmosis (FO) is usually recommended (Le and Nunes, 2016).

### **2.3.3 Drinking water treatment**

Although the relative contribution of various water sources including desalination has increased from 0.5% to around 1%, world water supply is still dominated by surface water sources (63.2%) and ground water sources (36.3%) (Pearce, 2011). In the present day the major water sources are effectively non-renewable and will diminish as a proportion of total abstraction over the coming decades but will remain important and will probably require greater treatment. This thesis mainly concerns membrane-based water treatment of surface water and ground water for drinking water supply and thus it is worth underlying fundamentals of membrane technology and development in the drinking water industry.

As discussed in Section 2.2.2, UF and MF membranes have been widely used in the water treatment market since the 1980s (Vera et al., 2015, Zondervan and Roffel, 2008). The early stage membranes tended to be UF with a fine or medium pore size. These were for either process water filtration or ultra-pure water production. In the second half of the 1980s, smaller pore-size NF emerged as a separate pressure-driven membrane process as a new low energy purification tool for the removal of colour, hazardous organic contaminants, and hardness from drinking water sources (Watson and Hornburg, 1989). In the 1990s, coarser UF and MF membranes were developed for drinking water, and these same membranes were then utilized in RO pre-treatment applications for wastewater and seawater duties (Pearce, 2016). The present day drinking water

membrane industry is dominated by coarse UF with a pore size rating of around 0.02  $\mu\text{m}$  or fine MF with a pore size rating of 0.04 – 0.1  $\mu\text{m}$  (Pearce, 2011).

The expansion of the membrane market has been driven by legislation for drinking water use which has led to a rapid uptake in membrane systems. The early products were expensive which tempted system designers to use high fluxes. Fouling tends to increase exponentially with flux (Yoon, 2015), so many early plants faced challenging operational problems resulting from an excessive development of transmembrane pressure (TMP). Furthermore, some of the early products were weaker than today's commercial offerings which led to integrity issues and occasional plant failures (Gijssbertsen-Abrahamse et al., 2006). The revolution has been influenced mainly by the development of membrane material and module designs in recent years. With increasing selectivity of separating components and growing membrane surface area per unit volume, membranes have improved providing better permeability and strength. Also, system design has become more conservative with a tendency to select a more moderate design flux. Nevertheless, the lessons from those early experiences are still relevant since some applications pose serious fouling challenges.

## **2.4 Membrane materials, modules and filtration fundamentals**

### **2.4.1 Membrane materials**

Membrane filtration developments have been driven by two main factors: the industry capability to produce high permeability, high selectivity, sufficient mechanical stability and defect-free

membranes on a large scale, and secondly, the ability to fabricate compact, high-surface-area, economical membrane modules (Van der Bruggen et al., 2003, Baker, 2012). Therefore membrane materials and modules are both important in the evolution of membrane technology.

Traditional membrane materials are organic polymers to ensure membrane hydrophilicity and hydrophobicity (Mulder, 2012). Membrane hydrophilicity refers to the ability to sustain stable and high fluxes while inhibiting fouling due to organic compounds, while hydrophobic membranes have little/no tendency to adsorb water due to low surface tension (Hoek and Tarabara, 2013, Abdelrasoul et al., 2013). Typically, hydrophilic membranes are identified with the presence of active groups which forms hydrogen-bonds with water, while hydrophobic membranes have the opposite interaction with water; water tends to bead on their surfaces (Abdelrasoul et al., 2013). Fouling tends to be enhanced with hydrophobic membranes but there are newly developed hydrophilic/hydrophobic composite membranes which have proved to present with less fouling (Khayet et al., 2005). Additionally, polymer blends are usually used in membrane fabrication to increase membrane hydrophilicity so that fouling can be inhibited (Kreuer, 2001, Zhao et al., 2007). Specifically, hydrophobic materials including polytetrafluoroethylene (PTFE), poly(vinylidene fluoride) (PVDF), polypropylene (PP), polyethylene (PE), and hydrophilic materials including cellulose esters, polycarbonate (PC), polysulfone/poly(ether sulfone) (PSF/PES), polyimide/poly(ether imide) (PI/PEI), aliphatic polyamide (PA), and polyetheretherketone (PEEK) are usually used in MF membranes (Van der Bruggen et al., 2003).

UF membranes must be prepared by phase inversion which are also characterized by their hydrophilic and hydrophobic properties (Wienk et al., 1996). Especially, strength and durability are essential in UF processes to withstand mechanical breakdown during chemical cleanings

(Hoek and Tarabara, 2013). However, it is hard for UF membranes to achieve both high fouling resistance and high durability (Hoek and Tarabara, 2013). Among all the materials, PVDF membranes have been proved to perform long term stability (5-10 years) (Matsuura, 1993). Other applicable UF membrane materials are PSF/ PES/ sulfonated polysulfone (SPSF), polyacrylonitrile and related block-copolymers, cellulose acetate, PI/PEI (Boom et al., 1992, Wienk et al., 1996). NF membrane materials are aromatic polyamide, PSF/PES/SPSF, cellulose acetate, and poly(piperazine amide), while RO membranes materials are cellulose triacetate, aromatic polyamide, interfacial polymerization of polyamide and poly(ether urea) (Van der Bruggen et al., 2003).

In recent decades, ceramic membranes have been commercially developed for both UF and MF membranes (Coronas and Santamaria, 1999). Compare to polymer membranes, ceramic membranes can provide superior chemical, thermal, and mechanical stability, and relatively high porosity and narrow pore size distribution (Oh et al., 2007a, Hofs et al., 2011). Relatively high hydrophilicity in ceramic materials promotes high fluxes at low pressure operations thus giving long term stability (Van der Bruggen et al., 2003). The high mechanical stability of ceramic membranes is specially favoured for high pressure backwash processes (Lerch et al., 2005, Meyn et al., 2008, Shirasaki et al., 2008). In terms of membrane permeability and fouling behaviours, ceramic membranes also have better performances due to higher volume/area ratio, for the same format, than polymer materials (Hofs et al., 2011). However, the production cost is high for ceramic membrane fabrication processes and ceramic materials are more prone to breakage than polymeric materials (Hofs et al., 2011, Amin et al., 2016).

## 2.4.2 Membrane modules

Along with the advances in membrane materials, the development of polymers also benefits membrane module innovation. Membrane modularization is aiming to pack large area of active membranes into a relatively small volume (high packing density) as well as maintain the system hydraulics to minimize fouling effects (Hoek and Tarabara, 2013). In the current membrane industry, there are various membrane module configurations: tubular, plate-and-frame, spiral wound, hollow fibre, and pleated Cartridges. The membrane module selection varies from process to process, where hollow fibre membrane (HFM) modules are designed for MF and UF systems, spiral wound modules are employed in NF and RO systems, and plate-and-frame modules are applied in UF and RO systems (Allgeier et al., 2005).

In general, tubular modules are applied to tolerate high concentration of suspended solids in the feed so that the membranes are minimally susceptible to blockage (Hoek and Tarabara, 2013). Tubular modules are easy to clean but limitation arise due to high cost, low packing density, and the replacement of membranes may require the entire module assembly to be replaced (Langerak et al., 1993, Balster, 2015, Hoek and Tarabara, 2013). Similar limitations exist in plate-and-frame modules. Plate-and-frame modules have slightly higher packing density than tubular modules, but the capital cost is still very high and the modules require longer time for mechanical cleaning and maintenance (Marriott, 2001). Especially, plate-and-frame modules are easy to be damaged during the loading and reloading of membrane plates (Hoek and Tarabara, 2013). Although the earliest RO membrane applications are tubular and plate-and-frame modules, spiral wound modules became an alternative option in the late 1960s (Westmoreland, 1968, Bray, 1968, Kremen, 1977). In spiral wound modules, the packing density are greatly increased, the membrane area per unit volume are much higher, and the capital and installation costs are lower

than other systems. However, mechanical cleaning cannot be operated in spiral wound modules and uncleaned fouling tends to be permanent, which in turn, causes shorter membrane lifetime (Baker, 2004, Hoek and Tarabara, 2013). Pleated cartridge modules are unsuitable for treating high concentration of suspended solids and are usually used in MF applications to treat relatively clean fluids, e.g. deionized water (Porter, 1989).

Out of all the membrane modules, HFM modules are mostly favoured in industrial applications (Ismail and Matsuura, 2016) and is the configuration of greatest interest in this thesis. Up to 80% of commercial gas separation (Drioli and Barbieri, 2011) and nearly all membrane products in the mainstream filtration duties in surface water treatment use membranes in the hollow fibre format since this has been found to provide the ideal combination of packing density, hydrodynamic efficiency, and integrity (de Ven et al., 2008, Pearce, 2011). A HFM module is a bundle of small porous hollow fibres packed into a shell cylinder which assembles the configuration of a shell and tube heat exchanger (Rogers and Long, 1997). The feed flow through a HFM can either be on the lumen side or the outside of the fibres, which will be discussed in detail in Section 3.1. Although HFM require filtration to remove large particles which could block the feed channels of each fibre, the modules themselves are much more resistant to fouling than spiral wound modules (Hoek and Tarabara, 2013). Unlike other configurations, HFM modules can maintain positive flow across the entire active surface of the membrane, which therefore eliminates stagnant areas where fouling and scaling tend to manifest (Katsoufidou et al., 2010).

In recent years, HFM modules became popular for both UF and MF applications because the following advantages: higher membrane area per unit volume to ensure higher productivity (Drioli and Barbieri, 2011, Li, 2008); higher packing density to provide more effective separation

(Ismail and Matsuura, 2016, Kim and Lee, 2011, Bessiere et al., 2008); self-mechanical support to permit backflushing in liquid separation (Li, 2008); much more resistant to fouling and scaling (Hoek and Tarabara, 2013); cheapest manufacturing costs (Drioli and Barbieri, 2011, Bessiere et al., 2008); and good flexibility and easy handling during module fabrication, operation and maintenance (Li, 2008).

### 2.4.3 Membrane filtration fundamentals

As early as in the 1970s, the solution-diffusion model for transport across dense membranes was adopted to describe the phenomenon in membrane filtrations (Hoek and Tarabara, 2013). Based on Darcy's Law (Eq. 2.1), permeate flux  $J$  depends on the membrane resistance (influenced by microstructure parameters of membranes and fouling layer accumulations), osmotic pressure difference ( $\Delta\Pi$ ), viscosity of the permeate ( $\mu$ ), and pressure difference across membrane ( $\Delta P$ ). For an unfouled membrane

$$J = \frac{\Delta P - \Delta\Pi}{\mu \cdot R_m}, \quad (2.1)$$

where  $J$  is the permeate flux, and  $R_m$  is the resistance of the clean membrane. The osmotic pressure difference,  $\Delta\Pi$ , is generally close to  $\Pi_m$ , the osmotic pressure at the interface between membrane and suspension, because the permeate is often of low osmotic pressure. The osmotic pressure is well known as the limiting phenomenon during RO processes such as the desalination of seawater (Bessiere et al., 2005), but to a smaller extent in most UF processes. Normally, the osmotic pressure is intensified by concentration polarisation. Concentration polarization,  $CP$ , is

linked to the flux,  $J$ , and the mass transfer coefficient,  $k_f$ , of the clean membrane system, via the Film model (Chong et al., 2008):

$$CP = \frac{C_m}{C_b} = \exp\left(\frac{J}{k_f}\right). \quad (2.2)$$

More recently it has been realized that when osmotic pressure has an influence upon the driving force the presence of a cake layer creates an unstirred region in which there is a gradient of osmotic pressure. In RO this phenomenon has been called Cake Enhanced Osmotic Pressure (CEOP) (Hoek et al., 2002, Hoek and Elimelech, 2003, Chong et al., 2008). Though not important for the direct-flow processes which are the subject of research in this thesis, the cake layer on the surface of a membrane will intensify the degree of concentration. As fouling and hence  $R_f$  changes with time the value of  $J/k_f$  changes and therefore  $\Delta\Pi$  changes; the osmotic pressure at the membrane surface is a function of time as well as the flux.

$$J = \frac{\Delta P - \Delta\Pi}{\mu(R_m + R_f)}. \quad (2.3)$$

When there is a cake layer present, the back transport is hindered (Hoek and Elimelech, 2003, Chong et al., 2008) so that:

$$CP = \exp\left(\frac{J}{k_{eff}}\right), \quad (2.4)$$

$$\frac{1}{k_{eff}} = \frac{1}{k_f} + \frac{1}{k_{fc}}, \quad (2.5)$$

where  $k_{eff}$  is the effective mass transfer coefficient that combines the mass transfer in the unfouled section of the membrane; and  $k_{fc}$  is the mass transfer in the cake layer which is a function of the cake layer thickness and the solute diffusion in the cake layer.

One should note that Eq. 2.1 can only be applied to membrane filtration systems when the feed stream is a clean fluid (i.e. no foulants). In the presence of fouling, membrane resistance during the filtration process can be varied due to particle accumulation on the membrane surface and/or in the membrane pores, which both will affect the flux and the TMP in the system. The modelling of membrane filtration in the presence of fouling is given by Eq. 2.6 (Field and Pearce, 2011).

$$J = \frac{\Delta P - \Delta \Pi}{\mu \cdot (R_m + R_{ads} + R_{rev} + R_{irrev})}, \quad (2.6)$$

where  $R_m$  still represents the hydraulic resistance of the clean membrane,  $R_{ads}$  is the additional resistance due to surface or pore adsorption that occurs independently of flux,  $R_{rev}$  is the increased resistance added to the system due to reversible component that can be removed via physical means (e.g. backwash or perhaps just a simple switch away from the feed and back to pure solvent), and  $R_{irrev}$  is the resistance added by irreversible component (i.e. deposited material that can only be removed by chemical cleaning operations). If no distinction is being made between the three fouling terms ( $R_{ads}$ ,  $R_{rev}$ ,  $R_{irrev}$ ),  $R_f$  represents the sum. The variation in the membrane resistance represents different fouling mechanisms and requires specific cleaning processes, which will be discussed in Section 2.5.3.

Since the stable performance and fouling issues confer operational and maintenance cost, and especially affect the lifetime of membrane, understanding the challenges of membrane fouling is one of the objectives of this thesis.

## **2.5 Membrane fouling and cleaning**

### **2.5.1 Membrane fouling**

Although membrane applications are favoured over many other separation and treatment options, the major limitation of membrane fouling arises in a broad range of applications (Belfort et al., 1994, Decarolis et al., 2001, Liu et al., 2007, Chen and Columbia, 2011, Gijiu et al., 2012, Ochando-Pulido et al., 2014, Zhou et al., 2015), especially in the liquid filtration processes (Stoller et al., 2013). It not only shortens the life time of the membrane module but also severely curtails the practical productivity of the filtration process (Yan-jun et al., 2000, Zhu et al., 2015, Park et al., 2017). Other issues caused by membrane fouling, e.g. reduced separation efficiency and increased energy consumption are also reported in the literature (Di et al., 2017). As a result, commercial membrane products are normally designed with an excessive oversized capacity, which adds an additional (up to 35%) capital and operating costs to the system (Stoller et al., 2013).

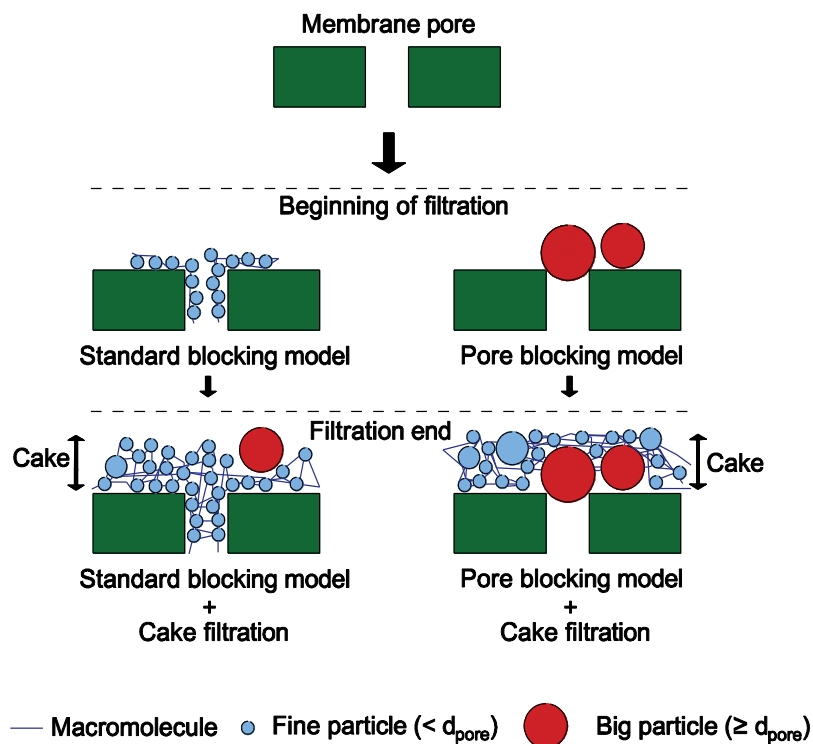
Membrane fouling happens when particles deposit on the membrane surface and/or inside its porous structure due to interparticle and particle-membrane interactions (Di et al., 2017). The fouling characteristics depend on both feed stream properties (particle size, shape and nature) and the operation of the membrane systems. According to the type of fouling materials, there are four types of membrane fouling categories: particulates fouling, inorganic fouling (scaling), organic fouling, and micro-biological fouling (Liu et al., 2001, Pearce, 2011). Particulates fouling can either be caused by inorganic or organic components which bind or block the membrane surface. Inorganic fouling is usually caused by dissolved components and colloids which tend to precipitate onto the membrane surface due to a pH change (scaling), or due to oxidation (Pearce,

2011). This type of fouling is major concern for RO and NF operations, while in MF and UF systems it is of little concern (Liu et al., 2001). Organic fouling happens when the process feed contains relatively high natural organic matters which attach to the membrane surface due to adsorption. This type of fouling was reported as a significant factor contributing to flux decline in various operations (Mallevialle et al., 1989, Lahoussine-Turcaud et al., 1990). Microbiological fouling refers to the type of fouling caused by the formation of biofilms on membrane surfaces due to algae, bacteria, and other vegetative matters (Pearce, 2011).

With time various materials either due to physical or chemical binding will gradually accumulate on the membrane surface and thus affect the filtration efficiency. The accumulation of fouling will result in flux decline in constant pressure operation and pressure increase in constant flux filtration (Sun et al., 2008). However, fouling should be distinguished with the phenomenon of concentration polarization. Concentration polarisation is a natural consequence of the semi-permeability and selectivity of membrane products (Shi et al., 2014), which occur in conjunction with irreversible fouling of the membrane as well as reversible gel layer formation. It normally results in an accumulation layer of rejected solutes and/or particles adjacent to the membrane surface, often referred to as gel formation. The main difference is that the gel layer is adjacent to the membrane surface (still in the solution), while the fouling layer is more closely bound to and/or inside the surface (Yan-jun et al., 2000, Aimar et al., 1991). More importantly, concentration polarization is reversible since the interactive forces between the macromolecules in the gel is relatively low whereas fouling may cause irreversible loss of the membrane permeability.

## 2.5.2 Fouling mechanisms

Particle removal technologies are prone to fouling because of the very challenge that they are trying to address. A simplified representation of the two main physical fouling mechanisms is shown in Figure 2.2. The role of particle size may differentially influence membrane fouling in various filtration operations (Tarabara et al., 2002). If the pore or aperture openings of the filtration media are larger than the particles the particles may enter the throat of the pore and block it or severely restrict the flow through it. Additionally, a high concentration of particles of different sizes congregating near the pore openings may cause a cake layer to build-up. As the cake layer develops, it effectively becomes the filtration medium since it prevents particles approaching the surface.



**Figure 2.2:** Particle fouling mechanisms in membrane filtration.

A third type of fouling is due to the adsorption of molecules onto the membrane surface which may bind other foulants or restrict pore openings. Adsorptive fouling normally has to be removed by some form of chemical cleaning.

If feed flow is maintained constant (i.e. constant flux filtration), then the consequence of fouling (and incomplete cleaning) is that the differential pressure rises from cycle to cycle, either due to the restriction in the number of pores available, or due to the additional resistance of the developing cake layer, or a combination of both. Often this requires the combination of both physical and chemical cleaning to enhance the filtration efficiency. The next section describes various methods for fouling control in a general view, and a more detailed discussion of cleaning specifically related to direct-flow filtration in this thesis will be in Section 3.2.

### **2.5.3 Cleaning processes**

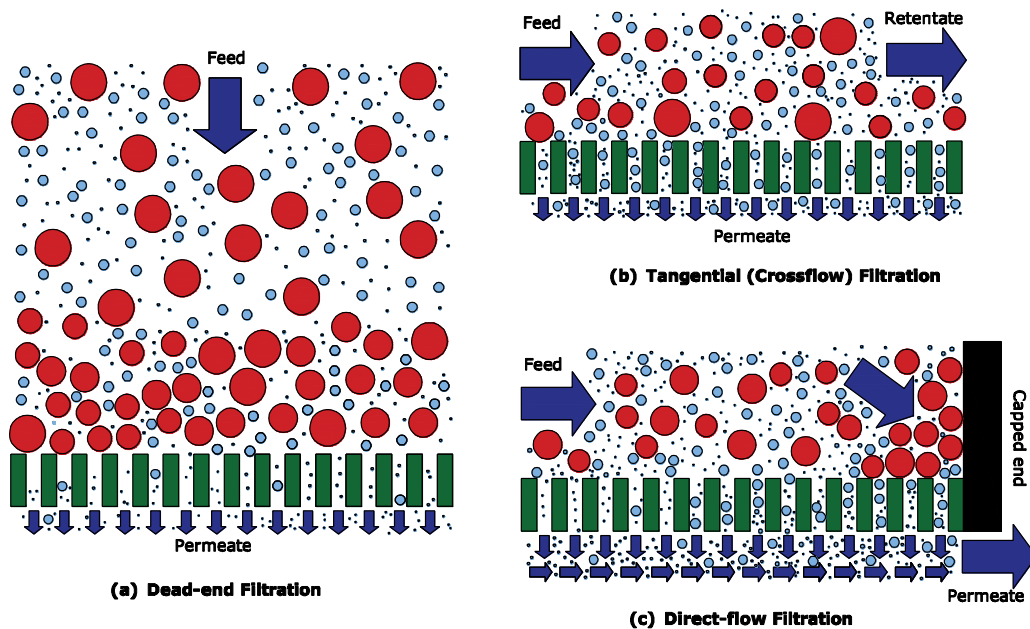
In membrane applications, cleaning is a vital step to remove fouling and maintain membrane permeability and selectivity so that the plant can maintain its design capacity (D'souza and Mawson, 2005). In practice, there are various cleaning approaches to mitigating the effects of membrane fouling. Either foulants are forced away from the membrane by hydrodynamic changes, or fouling is reduced by introducing chemical agents to the cleaning process (Pearce, 2011). Therefore, the fouling control and cleaning strategies can be summarised into two main categories: physical or mechanical cleaning, and chemical cleaning. Physical or mechanical cleaning can be achieved by introducing a higher shear rate during the operation, or in some membrane operations periodical backwashing can be applied on the permeate side to push part of the accumulated foulants away from the membrane (Trägårdh, 1989). Sometimes, membrane

relaxation, air scour, and forward flush can also be involved for physical/mechanical cleaning efforts (Miller et al., 2013, Yan-jun et al., 2000). The efficiency of physical/mechanical cleaning not only depends on the type of fouling, but also on the cleaning frequency and applied pressures (Galaj et al., 1984).

Often, clogging near the membrane surface can only be partially loosened or removed by physical cleaning processes. On a fairly regular basis, chemical cleaning with a cleaning agent should be incorporated to enhance cleaning efficiency. During chemical cleaning processes, the choice of cleaning agents and operating conditions not only depends on the type of particles deposited on the membrane surface, but also on the chemical and thermal resistance of the membrane (Trägårdh, 1989). Therefore, chemical cleaning process should be designed to minimize the adverse effects of cleaning agents on membrane life, the disposal of cleaning chemicals, the volume of water used and wastewater produced, and the disruption to normal membrane operations (Daufin et al., 1991b, Daufin et al., 1991a, Lawrence et al., 1998, Wagner, 2001). The major chemical agents used in membrane operations are alkalis, acids, enzymes, surfactants, sequestrants, and disinfectants (D'souza and Mawson, 2005). Sometimes, cleaning solutions are tailored with a combination of several agents to remove specific foulants. In the food industry, alkali (e.g. NaOH) and chlorine soak are used to treat organic fouling, acid soak (e.g. HCl, H<sub>2</sub>SO<sub>4</sub>, Citric) is used to tackle inorganic fouling, and biocide soak (e.g. Cl<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>) is normal for the treatment of bio-fouling (Pearce, 2011). In practice, both the physical and chemical cleaning are aiming to remove fouling deposits, restore the normal capacity, ensure adequate flux and filtration performance in subsequent operations. Therefore, an ideal cleaning system in membrane operations is usually achieved by a combination of appropriate physical and chemical cleaning processes.

## 2.6 Membrane flow configurations

Membrane filtration processes can be designed to operate in three ways: dead-end, crossflow, and direct-flow (Figure 2.3). In classical dead-end filtration the flow and filtration direction are both normal to the membrane surface, while in crossflow mode the flow is parallel to the membrane surface with permeation being orthogonal to the feed flow (Noble and Stern, 1995). Although in the last century the membrane industry was driven through the adoption of crossflow filtration, the modern water membrane market is based increasingly on the use of direct-flow filtration, which combines a filtration cycle operated in dead-end mode with a regular intermittent backwash.



**Figure 2.3:** Schematics of (a) dead-end, (b) crossflow and (c) direct-flow filtration. Please note in particular for (b) and (c) aspect ratio (length/radius width) is not to scale.

### **2.6.1 Dead-end filtration**

The classical dead-end filtration was originally applied for filtering a precipitate with a single filter paper (Noble and Stern, 1995). Essentially all of the fluid feed flow emerges as permeate in dead-end mode thus the filtration efficiency is relatively high. Also, very low energy is demanded in dead-end operation, it therefore becomes an attractive option in direct ultrafiltration of secondary effluents (Vera et al., 2015). However, the thickness of contaminants layer on the membrane surface constantly increase as more solute particles settles on the layer (Banerjee et al., 2012). This outcome is concentration polarization and significant membrane fouling as discussed in Section 2.5.

### **2.6.2 Crossflow filtration**

In crossflow mode the flow is parallel to the membrane surface with permeation being orthogonal to the feed flow, thus only a small fraction of the fluid actually passing across the membrane flows through it. The advantage of crossflow filtration is that the foulant concentration does not increase excessively on the membrane surface due to shear stress and inertial lift from the crossflow (Chang et al., 2002). The crossflow sweeps the contaminants downstream and limits the build-up of particles at the surface. This significantly reduces concentration polarization and membrane fouling (discussed in Section 2.5), which enables relatively high permeability and reasonable fluxes to be maintained. However, the pressure drop in the feed channel incurs a relatively high energy cost (Pearce, 2011), and this mode of operation is not suitable in large scale water treatment plants. Also, as some filtration duties only have a low-solids loading crossflow is not necessarily required.

### **2.6.3 Direct-flow filtration**

Since direct-flow filtration offers an efficient combination of the benefits of both dead-end and crossflow filtration, it has been broadly applied in the modern membrane market especially when solids loading is low. During the filtration cycle, one end of the crossflow device is effectively capped so that all the fluid is forced to pass through the membrane walls. This is normally achieved through near-permanent closing of the relevant valve. To ensure sustainable operation in direct-flow filtration short automatic backwashes at regular frequencies are incorporated to make fouling mainly reversible (B. Balannec 2005) and typically the capped-end remains closed. If necessary, low dose of disinfectant/oxidant can be added to the backwash water of direct-flow filtration to remove foulants and restore membrane permeability (Li, 2008). As discussed, the pressure drop in the feed channel incurs a relatively high energy cost in crossflow filtration which would not be suitable for large scale water treatment applications. Direct-flow filtration is more energy efficient than crossflow filtration since the inflow velocity is modest because all of the feed is filtered. Thus the pressure drop along the feed channel during the filtration cycle can be very small. Also it permits the use of smaller diameter fibres which enables the filtration area per unit volume to be increased. Additionally, with the elimination of the need to withstand high pressure drop crossflow, direct-flow provides the module with the possibility of much higher packing densities than other UF and MF products, which in turn reduces the overall costs (Pearce, 2011). It therefore forms the dominant method for producing potable water from ground or surface water (Blankert et al., 2006). A summary of flow configuration in these three filtration formats is given in Table 2.1.

**Table 2.1:** Summary of flow situations in three filtration modes

|                                  | <b>Dead-end</b>           | <b>Crossflow</b>              | <b>Direct-flow</b>                        |
|----------------------------------|---------------------------|-------------------------------|-------------------------------------------|
| <b>Strength of feed flow</b>     | Feed flow = Permeate flow | Feed flow $\gg$ Permeate flow | Feed flow = Permeate flow                 |
| <b>Tangential flow</b>           | None                      | Yes, strong                   | Yes, weak and variable <sup>1</sup>       |
| <b>Permeate rate</b>             | Low permeate rate         | High permeate rate            | High net permeate rate                    |
| <b>Orientation of permeate</b>   | Same direction            | Orthogonal                    | Orthogonal                                |
| <b>Means of reducing fouling</b> | None                      | Shear due to crossflow        | Frequent periodic backwashes <sup>2</sup> |

Although direct-flow filtration has the advantage of cost-competitive and viable alternative to conventional methods, most recent studies have focused on the classical technique of cross-flow filtration format, there remains a general lack of information on the direct-flow filtration behaviours.

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<sup>1</sup> Tangential flow in direct-flow filtration exceeds weak and is towards moderate, but becomes strong in backwash.

<sup>2</sup> Backwash creates crossflow to reverse pore plugging and concentration polarization so that particles can be removed from both the pore throat and the cake layer.

## **2.7 Conclusion**

In this chapter, the relevant literature on the membrane filtration history and membrane process development has been reviewed, followed by an emphasis on the membrane applications in water treatment industry. Membrane materials, modules and fundamentals in the water industry has been extensively studied. Especially, membrane fouling and cleaning challenges in the water treatment industry were discussed. The underlying fundamentals in membrane flow configuration has been introduced so that the readers understand the current and promising future market of direct-flow filtration. From the review, the development of modern membrane market based on direct-flow filtration is still at a relative young age. Therefore, there exists the need for research approaches to address the operational and design guidelines within direct-flow filtration.

# **Chapter 3**

## **Literature Review of Direct-Flow**

### **Filtration**

Having given a general contextual introduction in Chapter 2, this chapter focuses upon existing literature that is particularly pertinent to the focus of this thesis, namely direct-flow filtration. It firstly introduces the two filtration formats and the two operating modes within direct-flow filtration, and then discusses specific fouling issues and cleaning measures inherent in this flow configuration. Lastly, this chapter outlines research works in existing literature supporting the background of this thesis and thus interprets the objective and necessity of this research.

## **3.1 Direct-flow filtration fundamentals**

### **3.1.1 Module formats**

As mentioned in Section 2.4.2, there are two formats for a hollow fibre membrane module and they are based on whether the feed is on the inside of the fibre lumen or on the outside. The formats are respectively known as pressure driven inside feed (PDI) and pressure driven outside feed (PDO). Of interest to the users of membrane products, the two formats have different strengths and weaknesses in operation due to their different operational characteristics.

The inside feed format has the advantage of accommodating the contaminants from the feed in the fibre lumen and providing controlled hydrodynamics (Pearce, 2011). Since the entire surface of the membrane is continuously scoured by the crossflow of the fluid, inside feed format allows for improved fluid management, which minimizes the fouling effects (Porter, 1989). Another advantage is that a more even flow distribution of the feed can be achieved thus the tolerance to non-uniform packing is higher with the inside feed format (Drioli and Barbieri, 2011). Furthermore, the requirement of mechanical strength of the module casing is reduced due to lower pressures on the shell side during inside feed filtration (Drioli and Barbieri, 2011). The downside is that the surface area based on the inner diameter is much less than that based on the outer diameter. Also, it is practically not possible to generate the turbulence to mitigate cake layer formation due to the small inner diameter and relatively low water velocity, therefore membranes in PDI modules are somewhat more subject to plugging and the potential for the lumen to become clogged is increased (Allgeier et al., 2005). Although PDI modules might suffer from integrity loss during operational processes, they do provide long term stability and records in commercial

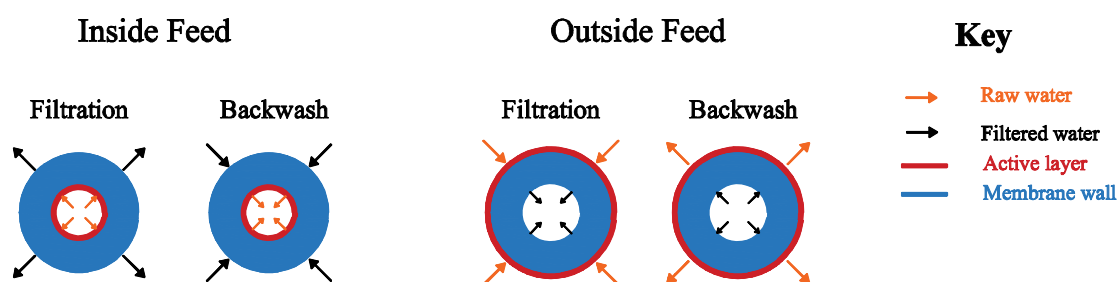
practice reveal that the modules operate well under low pressures with a moderate or conservative design flux (Pearce, 2011).

For the outside feed format, the advantages are tolerance of a high solids concentration in the feed and a higher surface area (Yoon, 2015). The downside is that the hydrodynamics are spatially variable and not so easily controlled, e.g. the channelling problem in outside feed format causes a reduction in the mass transfer coefficient (Mulder, 2012, Wu and Chen, 2000, Allgeier et al., 2005, Pearce, 2011). Since the surface of the membrane is not as effectively scoured as in inside feed format, outside feed operation requires more frequent backwash cleaning to address fouling issues (Hoek and Tarabara, 2013). To counter its poor efficiency in backwash, outside feed formats utilise air scour during backwash which is effective but adds another process operation.

An important variant of the outside feed configuration is the immersion of a bundle of fibres into a tank of the water to be treated, without module containment, and to apply a vacuum on the permeate side. This is known as the submerged format.

The different requirements of inside and outside feed formats have led to a different selection of polymer materials. Two materials have come to dominate the industry, namely polyethersulfone (PES) and polyvinylidene difluoride (PVDF). Most inside feed products utilise PES which has high permeability and good strength. The outside feed format is dominated by PVDF which has good flexibility allowing it to tolerate air scour, but it has lower permeability. The surface area advantage of the outside feed format is somewhat lost by having to use a flexible material with lower permeability.

Figure 3.1 shows that during the filtration cycle the fibre is subject to expansion due to the pressure of the feed. In backwash, the fibre is subject to compression. The reverse is true for the outside format. Expansion of the inside format during the filtration cycle will tend to enlarge the pores. If pressures are too high, fouling effects such as pore plugging may be enhanced. Conversely, the backwash operates typically at higher flow, perhaps 2.5 times that of the filtration cycle. Any foulants trapped in the pores during the filtration cycle may be held by pinching due to slight contraction during backwash.



**Figure 3.1:** Process flows of the two formats as seen in cross-section of the hollow fibre modules.

Commercial practice has shown that inside feed format plants operate well if pressures are kept low by using a moderate or conservative design flux. Under these conditions, backwash is effective and the plant has good long-term stability. However, if the design flux is too high and pressures rise during a sequence of filtration cycles, the backwash pressures can quickly become excessive. The removal of foulants by backwash along may be ineffective and irreversible fouling may develop. In the worst cases, the pressures could become so high that the fibre loses integrity, becoming crushed by pressure spikes (Gijssbertsen-Abrahamse et al., 2006).

Outside format plants also have limitations if pressures become too high, since the trans-membrane pressure can rise sharply to display a characteristic saw-tooth pattern during the feed cycle if operation is too aggressive. The compression of the fibre during the filtration cycle means that the attempts to maintain the flux can be confounded since resistance increases, reducing the sizes of pores and hence the permeability. Additionally it may lead to compression of any cake filtration layer. Although the foulants may be effectively removed during the backwash/air scour cycle, energy and chemical costs can become excessive (Field and Pearce, 2011). The problems for these plants are therefore not so much loss of integrity but excessive operational costs.

Some earlier plants of both formats suffered from problems since expensive membranes and inexperience encouraged the use of excessive design fluxes. Outside feed format plants suffered from excessive operational costs and inside feed formats could potentially suffer from integrity loss or require fibre repair procedures too frequently. When high design flux was combined with early stage product weakness, excessive fibre breakage could occur, and a small percentage of plants suffered from this specific problem (Pearce, 2011).

In practice, the pros and cons of the two formats are therefore finely balanced and both approaches have strong adherents. Indeed, both types of product are strongly represented in the membrane market. The details of the operating characteristics for both formats are summarised in Table 3.1.

**Table 3.1:** Operating characteristics and limitations in inside and outside formats

|                                  | <b>Inside feed</b>                       | <b>Outside feed</b>                                               |
|----------------------------------|------------------------------------------|-------------------------------------------------------------------|
| <b>Membrane area/volume</b>      | Low                                      | High                                                              |
| <b>Permeability</b>              | High                                     | Low                                                               |
| <b>Polymer materials</b>         | PES: high permeability and good strength | PVDF: good flexibility to tolerate air scour but low permeability |
| <b>Hydrodynamics</b>             | Good maintain                            | Spatially variable                                                |
| <b>Cake formation</b>            | Easy lumen clogging                      | Thinner layer formation                                           |
| <b>Backwash</b>                  | Effective                                | Low efficiency, air scour cycle needed                            |
| <b>Channel problem</b>           | No, or very little                       | Yes                                                               |
| <b>Maldistribution of fibres</b> | Good tolerance                           | Channel problem enhanced                                          |
| <b>Integrity loss</b>            | Yes                                      | Not so much                                                       |
| <b>Operating pressure</b>        | Low                                      | Low to moderate                                                   |
| <b>Design flux</b>               | Moderate or conservative                 | Low to moderate                                                   |
| <b>Long term stability</b>       | Good but require fibre repair frequently | Good                                                              |
| <b>Cost</b>                      | Maintenance cost                         | Excessive operational cost                                        |

These days, products have improved and fluxes tend to be lower, but challenging feeds can still raise concerns. For example episodes of poor feed water quality may combine with lower temperatures and/or temporary equipment failure (such as a sticking valve) to increase the stress on a plant. This thesis therefore sets out to model the hydrodynamic efficiency of the inside feed format module. The ultimate aim is to identify operational triggers such as the maximum allowable filtration cycle pressure to ensure the long term operational resilience of these plants.

### 3.1.2 Operating modes

Hollow fibre membrane filtrations can be performed under two conditions: constant pressure (CP) and constant flux (CF). During constant pressure operation, the overall module transmembrane pressure (M-TMP) is fixed by applying a steady pressure to the feed flow and withdrawing the permeate at the atmospheric ambient. Unlike constant pressure operation, in CF operation the imposed permeate flux is maintained for the feed flow at constant flux filtration. Although the current literature research has been directed on the former operation in the last two decades, the latter one is more favoured in commercial practice (Iritam et al., 2015, van de Ven et al., 2008).

As generally reported in recent studies (Iritam et al., 2015, van de Ven et al., 2008, Sun et al., 2008, Miller et al., 2013, Miller et al., 2014b, Miller et al., 2014a), flux declines over time under constant pressure operation and pressure (and/or transmembrane pressure) increases in constant flux filtration. Field et al. applied the concept of critical flux during their experiments and observed that the overall mass transfer resistance of membrane and the fouling layer was generally low for operation at constant flux and high for constant pressure filtration (Field et al., 1995). The authors suggested constant flux filtration for industry since fouling can be controlled compared with constant pressure operation. Miller et al. also reached the same conclusion by studying the fouling mechanisms under two conditions and found that CF operation (provided fluxes are not too high) provides an operational mechanism to abate fouling formation, while rapid fouling was observed at the start of constant pressure experiments (Miller et al., 2014a). Despite the advantage of reducing enormous fouling during the filtration cycle, Le-Clech et al. also pointed out that constant flux filtration is more cost effective (Le-Clech et al., 2006). However, the authors did have concerns in constant flux filtration because evidence of

irreversibility was found in the deposition layer. Meanwhile, they also mentioned the benefits of the substantial deposition layer generated in constant flux operation to be acted as a pre-filter for particles. The overview of the comparison of constant flux and constant pressure in recent literatures is presented in Table 3.2.

**Table 3.2:** Overview of filtration variations at constant flux and constant pressure operations

| Filtration | Format | Mode      | Details                                                                                                                                                                                                                   | Main Findings                                                                                                                                                                                                                                                                                                                                                                                                                                                      | Ref                                                               |
|------------|--------|-----------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------|
| Dead-end   | /      | CP and CF | <ul style="list-style-type: none"> <li>• Microfiltration</li> <li>• CP experiments operated at 98 to 294 kPa</li> <li>• CF tests ranges from 1152 to 2088 L m h</li> </ul>                                                | <ul style="list-style-type: none"> <li>• Filtration flux dramatically declines over time in CP.</li> <li>• TMP rises over time in CF.</li> </ul>                                                                                                                                                                                                                                                                                                                   | (Iritam et al., 2015)                                             |
| Crossflow  | PDI    | CF        | <ul style="list-style-type: none"> <li>• Ultrafiltration</li> <li>• The filtration flux ranges from 20 L m h to 110 L m h, while the backwash operates at 250 L m h</li> </ul>                                            | <ul style="list-style-type: none"> <li>• There exits an initial increase in axial pressure drop.</li> <li>• Higher pressure drop observed at higher fluxes.</li> </ul>                                                                                                                                                                                                                                                                                             | (van de Ven et al., 2008)                                         |
| Dead-end   | /      | CF        | <ul style="list-style-type: none"> <li>• Microfiltration</li> <li>• Permeate flux ranges from 577 L m h to 2293 L m h</li> </ul>                                                                                          | <ul style="list-style-type: none"> <li>• Pressure increases during CF filtration at both low and high fluxes.</li> <li>• TMP increases with flux slowly first and rapidly later at high fluxes.</li> <li>• TMP increases slowly and fouling is almost negligible at low fluxes.</li> </ul>                                                                                                                                                                         | (Sun et al., 2008)                                                |
| Crossflow  | PDO    | CP and CF | <ul style="list-style-type: none"> <li>• Flat-sheet ultrafiltration</li> <li>• Feed pressure 210 kPa in CP experiments</li> <li>• CF experiments were performed at five fluxes: 25, 40, 55, 85, and 100 L m h.</li> </ul> | <ul style="list-style-type: none"> <li>• Decrease in permeate flux with filtration time at CP process.</li> <li>• CF: initial TMP increases with higher flux.</li> <li>• Modest TMP increase at low flux and rapid TMP increase at high flux (above threshold flux) during CF filtration.</li> <li>• Filtration resistance during CF rises to the same value at which CP resistance reached a plateau, after which the CF resistance increases rapidly.</li> </ul> | (Miller et al., 2014a, Miller et al., 2014b, Miller et al., 2013) |

|             |     |           |                                                                                                                                                                                                                                                                                |                                                                                                                                                                                                                                                                                                                                                                            |                                    |
|-------------|-----|-----------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------|
| Crossflow   | PDO | CP and CF | <ul style="list-style-type: none"> <li>• Membrane bioreactor</li> <li>• CP operations first start at 0.4 bar for 30 min to obtain a stabilised flux.</li> <li>• CF starts from below 50 L m h and once stabilised TMP observed, flux increases 10 L m h every hour.</li> </ul> | <ul style="list-style-type: none"> <li>• Compared to CP experiment, CF avoids over-fouling during the initial stage of filtration.</li> <li>• In long term cross-flow filtration, it is preferred to operate at CF than at CP.</li> <li>• CF operation avoids excessive fouling of membranes as well as being cost effective for submerged membrane operations.</li> </ul> | (Defrance and Jaffrin, 1999)       |
| Crossflow   | PDO | CF        | <ul style="list-style-type: none"> <li>• CF experiments performed from 259 to 540 L m h with a starting pressure 210 kPa.</li> </ul>                                                                                                                                           | <ul style="list-style-type: none"> <li>• TMP increases throughout the entire filtration process due to protein aggregation and deposition.</li> <li>• The capacity of the membrane system is a strong function of the constant volumetric flowrate and high flow rates enhances high specific resistance.</li> </ul>                                                       | (Ho and Zydny, 2002)               |
| Crossflow   | PDI | CP and CF | <ul style="list-style-type: none"> <li>• CF experiments targeted at 149 L m h.</li> <li>• CP tests are regulated by a pressure gauge from 0 to 690 kPa.</li> </ul>                                                                                                             | <ul style="list-style-type: none"> <li>• CP mode is characterised by higher specific fluxes compared with those in CF.</li> <li>• Modelling results suggest preferential use of CP mode as particle size decreases.</li> </ul>                                                                                                                                             | (Tarabara et al., 2002)            |
| Crossflow   | /   | CP and CF | <ul style="list-style-type: none"> <li>• CP tests operated at 34.5, 69.0 and 103.5 kPa.</li> <li>• CF filtration performed from 30 to 130 L m h.</li> </ul>                                                                                                                    | <ul style="list-style-type: none"> <li>• Resistance evolution is well correlated with flux in CF filtration, while it is observed a rather constant resistance in CP filtration.</li> </ul>                                                                                                                                                                                | (Sioutopoulos and Karabelas, 2016) |
| Direct-flow | PDI | CF        | <ul style="list-style-type: none"> <li>• CF filtrations compared from 40 to 100 L m h while other geometrical parameters manipulated.</li> </ul>                                                                                                                               | <ul style="list-style-type: none"> <li>• Initial TMP increases as fluxes increases at CF filtration.</li> <li>• During CF filtration, the TMP decreases initially and then rises again towards the capped-end.</li> </ul>                                                                                                                                                  | (Herterich, 2015)                  |

Since constant flux filtration has been much less reported in the literature presumably due to the complexity and challenge under laboratory operation, the reason why they are popular for industrial filtration systems is not clear. This thesis therefore sets out to investigate filtration under constant flux via various modelling simulations.

### **3.1.3 Membrane materials and module designs**

As mentioned in Section 3.1.1, PES and PVDF are the major membrane materials in direct-flow filtration, with the former favoured in inside feed format and the latter favoured in outside feed format. The main advantage of PVDF is longer membrane life due to low fibre breaks. Together with air scour, PVDF is suitable for a wide range of feed streams. Due to its strong tolerance of chlorine, PVDF is usually applied in MBRs, while chlorine tolerance in PES is only sufficient for surface water, seawater, and wastewater treatment (Pearce, 2011). On the other hand, PES combines excellent permeability and a narrow pore size distribution within UF application which operate effectively in removing organics. The relatively high permeability of PES permits cost-effective operation both in terms of capital cost and operational maintenances. Although PVDF is more expensive than PES and has lower permeability, it can be successfully applied in outside feed configuration to avoid these disadvantages by providing a higher membrane area per unit volume. In addition, PVDF allows higher chlorine concentration to be used in cleaning in place (CIP).

Apart from the intrinsic characteristics of the membrane polymer materials, membrane filtration performance is also influenced by the pore size, pore density, and membrane morphology (Li, 2008, Cogan and Chellam, 2009, Tung et al., 2010, Miller et al., 2014a). Although the pore size

distribution of PVDF is wider than PES, it is usually of less significance in general water or wastewater treatment applications. Membrane fibre selection tends to be mainly influenced by other factors, among which design of fibre geometry (length, diameter and wall thickness) is an important one. For example, if fibre diameter increases, fibre wall thickness has to increase accordingly to maintain the membrane strength, and this can result in loss of membrane area and the drop in membrane permeability (Drioli and Barbieri, 2011, de Wit et al., 2017). In general, the membrane industry makes fibres with diameters in the range of 0.7 – 1.0 mm to ensure robust, self-supporting, high permeability in their applications (Pearce, 2011). This therefore determines a typical fibre design with around 1.5 m length for inside feed configuration and a slightly longer length for outside feed due to different backwash flowrates (Pearce, 2011). Details of commercial optimization in membrane module designs will be discussed later in Chapter 4.

Another influential factor of membrane filtration performance is the operating conditions. In addition to the module design, operation in membrane filtration systems requires careful selection of imposed flux values to achieve cost-effective design so that fouling can be controlled at an acceptable level (Berubier, 2007). A very low flux will increase the capital cost of the membrane system due to higher membrane area needed to push the throughput. On the other hand, a very high flux will lead to high operating and maintenance cost since membrane fouling increases exponentially with flux once a threshold flux is exceeded (Field and Pearce, 2011). At high fluxes, particles accumulate to the surface more quickly and the resulted high TMP will compress the cake layer and encourage particles to plug the pores. As a result, the use of high driving pressures and frequent chemical cleaning become necessary in high flux operations (Pearce, 2011).

## **3.2 Fouling and cleaning in direct-flow filtration**

### **3.2.1 Fouling in direct-flow filtration**

As introduced in Section 2.5, fouling in membrane filtrations is complex with multiple interactions between various fouling components in the feed and between the components and the membrane surface. Within different membrane modules and operations, different fouling characteristics and problems will be observed due to the differing nature of the flow and the filtration hydrodynamics. In direct-flow filtration, the use of a hollow fibre membrane module with a capped-end introduces axial flow and spatial filtration variations into the system, which therefore implicates a non-uniform fouling distribution along the fibre. As reported in the literature (Fane and Fell, 1987, Carroll and Booker, 2000, Chang et al., 2006, Li et al., 2015), the rate and mechanism of fouling during direct-flow filtration depend on their fibre length, diameter and other module design features. The authors found that axial fouling differences are huge at the initial stages of operation but become less significant as filtration proceeds. Especially, fouling is much more severe towards the capped-end (Pearce, 2011, Lv et al., 2017). The unique axial fouling features during direct-flow filtration therefore requires unique selection of cleaning procedures.

### **3.2.2 Cleaning processes in direct-flow filtration**

During direct-flow filtration operations, there are three fouling control and cleaning strategies: prevention, maintenance and recovery. Prevention normally applies a physical cleaning method including backwash, membrane relaxation, air scour, or forward flush (Miller et al., 2013, Yan-

jun et al., 2000). Maintenance usually involves the use of chemicals during the cleaning process, e.g. chemical wash, chemical enhanced backwash (CEB) (Baars et al., 2005). The typical recovery procedure is clean-in-place (CIP) which is relatively time consuming and expensive since specific cleaning formulations are used. CIP often uses some of the same chemicals as used in the chemical wash, thus also classifies as chemical cleaning. However CIP generally involves multiple stages.

### **3.2.2.1 Physical cleaning**

In direct-flow filtration, the intermittent cleaning process used during the filtration cycle is referred as the backwash cleaning process. Backwash cleaning is usually incorporated on a regular basis (i.e. 1-4 times/ hour) in direct-flow filtration to maintain long term operational stability. It is mainly designed to remove loosely attached foulants in a simple and inexpensive way. It controls the build-up of fouling by kinetically expelling particles from the membrane surface, i.e. it creates both reverse pore flow and surface shear to dislodge surface attachment (Le-Clech et al., 2006). The conditions used in the backwash process and the mechanism of the operation may change between membrane formats. The flowrates of backwash water can also vary significantly in different cases, normally 2.5 times the feed flow (Pearce, 2011). One should note that backwashing of other module configurations is not recommended because pressurization of the back side of the membrane could lead to delamination of the membrane itself from the substrate, especially in RO membranes (Hoek and Tarabara, 2013).

The physical backwash process may remove more than 90% of the foulants, but it is not fully effective (Tung et al., 2010). Especially in direct-flow filtration, fouling is much more severe near

the capped-end compared to the entrance, which leads to the reduction of physical cleaning efficiency towards the capped-end (Lv et al., 2017).

Sometimes air flush and/or air scour may be combined with liquid flow in either the filtration or backwash cycle. For inside feed configuration, air flush can be used after the high velocity backwash flow to expel water from lumen. Alternatively, the air can be introduced to the fibre lumen by creating a slug flow of air pockets either during forward filtration or backwash cleaning (Serra et al., 1999, Cabassud et al., 2001, Pearce, 2011). While air flush is applied in inside feed formats, air scour is more likely to be used in outside feed filtration (Fane et al., 2002, Fane et al., 2005). The air scour combats the fouling effects by loosening particles and vibrating them to detach the membrane surface. This cleaning technique can also be conducted with either forward flush or backwash flow (Pearce, 2011), which enhances the cleaning with an even more aggressive effect than simple backwash. The filtration maybe ceased when air scouring is processed, this is known as membrane relaxation (Vera et al., 2015).

In some filtration formats, the use of crossflow flushing is also possible to address fouling. During the low flowrate forward flush, crossflow creates high shear stress to displace particles on the feed side so that the particle concentration at the membrane surface can be reduced. By preventing an excessive particle concentration, the risk of plugging the feed channel is effectively reduced. The use of low flowrate crossflow flushing is particularly beneficial for large particle filtrations (Pearce, 2011). It can be undertaken either during the filtration cycle or as part of the backwash cleaning process. Due to cost implications, the low flowrate crossflow is only applied when necessary.

### 3.2.2.2 Chemical cleaning

In some cases, clogging near the membrane surface can only be partially loosened or removed by physical cleaning processes, therefore introducing chemical cleaning is necessary. On a fairly regular basis, the addition of chemicals are required to improve the efficiency of particle removal, and periodically, a full cleaning-in-place procedure is required for the membrane systems. The mechanism of chemical cleaning is different to physical approaches. Physical processes generally apply hydraulic and mechanical forces to the fouling layer through high velocities, whereas chemical cleaning is target to hydraulically irreversible foulants, i.e. the strong parts of the fouling (Kim et al., 2010, Shi et al., 2014). Chemical cleaning mainly depends on chemical reactions to combat foulants from the membrane system. In practice, it is normal to use chemical agents for cleaning on a periodic basis as a maintenance strategy, either being dosed into the backwash or to the feed forward flush. Compared with physical cleaning, the enhanced flow (either in backwash or feed forward) with chemicals require a few minutes of extended contact between the membrane to allow cleaning reactions to occur.

Chemical cleaning involves: 1. Chemical enhanced backwash; 2. Maintenance cleaning with higher chemical concentration; 3. Intensive (or recovery) chemical cleaning, i.e. clean-in-place (Le-Clech et al., 2006). The first two chemical wash process may be operated from between several times per day to once per week, while clean-in-place may only be once or twice a year (Pearce, 2011). These maintenance cleaning procedures are aimed at maintaining the membrane permeability and to reduce the frequency of further intense cleaning. As discussed in Section 2.5.3, the most common-used chemical agents include alkali (e.g. NaOH) or chlorine soak to treat organic fouling, acid soak (e.g. HCl, H<sub>2</sub>SO<sub>4</sub>, Citric) to tackle inorganic fouling and biocide soak (e.g. Cl<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>) for the treatment of bio-fouling (Pearce, 2011).

When the filtration process is no longer sustainable, e.g. an elevated TMP is observed, clean-in-place is necessary. It is needed as part of the restoration procedure after a long period operation to tackle the heavy or tenacious fouling remaining on the membranes. Clean-in-place procedures are normally employed between once in several months to once a year (Le-Clech et al., 2006). The same chemical agents are used as for the previous two chemical wash processes. However, the soak time of the cleaning is no longer a few minutes but extended to 30 minutes to an hour so that the chemical cleaning action have enough time to complete (Pearce, 2011). Additionally, the concentration of the chemical agent used is normally higher than chemical enhanced backwash. Feed side recirculation may be involved in cross-flow filtration to ensure the chemical concentration is maintained at the membrane surface and to introduce shear stress to the flow so that the velocity will help to dislodge the foulants (Pearce, 2009). In direct-flow filtration the feed side recirculation is not achievable, instead, clean-in-procedure can be conducted in a filtration cycle mode to create partial crossflow velocity to the system (Pearce, 2011). This operation may not be fully effective in direct-flow filtration due to its capped-end design. Therefore, fouling and cleaning control has to be specifically discussed pertinent to flow configuration, module format, and operating conditions in the membrane system.

### **3.3 Module optimization**

Although many industrial projects have employed the direct-flow filtration format, there has been few published studies on the associated flow regimes and filtration behaviours. In particular there are limited entries for the inside feed format direct-flow filtration as given in Table 3.3 which includes some results in crossflow mode.

**Table 3.3:** Overview of optimal hollow fibre module design for PDI modules.

| Filtration Modes | Feed    | Operating Details                                                                                                             | Module Dimensions                                                                                                                                                      | Variables investigated                                                              | Main Findings                                                                                                                                                                                                                                                                                                                                                                                                             | Ref                        |
|------------------|---------|-------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------|
| Direct-flow      | Inside  | <ul style="list-style-type: none"> <li>• DuPont B-9 module</li> <li>• No backwash considered</li> <li>• Pure water</li> </ul> | $b = 0.042\text{mm}$<br>$a = 0.021\text{mm}$<br>$L = 765\text{mm}$ or $311\text{mm}$<br>$\epsilon = 0.45$ or $0.51$                                                    | $L$                                                                                 | <ul style="list-style-type: none"> <li>• Filtration productivity of hollow fibre modules can be increased significantly either by opening both fibre ends or by fabricating shorter modules.</li> </ul>                                                                                                                                                                                                                   | (Soltanieh and Gill, 1983) |
| Direct-flow      | Outside | <ul style="list-style-type: none"> <li>• Pure water</li> <li>• With frequent backwash</li> </ul>                              | Standard parameters<br>$d_o = 0.85\text{mm}$<br>$d_o/d_i = 1.75$<br>$L = 1.2\text{m}$<br>$\epsilon = 0.36$<br>$k_m = 3.5 \times 10^{-10}\text{m Pa}^{-1}\text{s}^{-1}$ | One changed at a time<br>$d_o$<br>$d_o/d_i$<br>$L$<br>$\epsilon$<br>$k_m$<br>$\phi$ | <ul style="list-style-type: none"> <li>• Fibre properties and its design parameters have large influence in the filtration behaviours.</li> <li>• There is an optimum fibre diameter and void fraction due to the competition between an increase in filtration area and increase in pressure drop.</li> <li>• Any increase in fibre length beyond a certain value has little effect on increasing throughput.</li> </ul> | (Serra et al., 1998)       |
| Crossflow        | Outside | <ul style="list-style-type: none"> <li>• Submerged hollow fibre module</li> </ul>                                             | $d_i/d_o = 1/2$<br>$L = 0.8\text{-}4\text{ m}$                                                                                                                         | $d_i$<br>$d_o$<br>$L$<br>$\phi$                                                     | <ul style="list-style-type: none"> <li>• The positive effect resulted by increasing the fibre length is dominant over the negative effect by reducing the fibre numbers (reducing the packing density).</li> <li>• High packing density will result in increase of fouling due to the unfavourable hydrodynamic conditions within the fibre bundles.</li> </ul>                                                           | (Yoon et al., 2004)        |

|             |                    |                                                                                                                                                                                                                         |                                                                                                                            |                                                            |                                                                                                                                                                                                                                                                                                                                                                |                                              |
|-------------|--------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------|
| Direct-flow | Outside            | <ul style="list-style-type: none"> <li>• Constant flux filtration</li> <li>• Effect of particle accumulation on the membrane surface considered</li> </ul>                                                              | $d_i/d_o = 0.39/0.65$ or $0.2/0.5$<br>$L=930$ mm<br>$R_m=7.06 \times 10^{11}$ or $1.134 \times 10^{12} \text{ m}^{-1}$     | $d_o/d_i$                                                  | <ul style="list-style-type: none"> <li>• The non-uniform features of the filtration along the fibre will increase with decrease in fibre size (<math>d_o/d_i</math>) and increase in imposed average flux.</li> </ul>                                                                                                                                          | (Chang et al., 2006)                         |
| Crossflow   | Inside             | <ul style="list-style-type: none"> <li>• Laboratory scale microfiltration unit</li> <li>• Constant module inlet flow rate</li> <li>• Feed consists of a dilute solution containing rigid spherical particles</li> </ul> | $a=0.1, 0.29, 0.375$ mm<br>$L=0.305$ m                                                                                     | $a$                                                        | <ul style="list-style-type: none"> <li>• The permeate flux (averaged over the length) was found to vary by as much as 65% depending on the lumen radius.</li> <li>• Both cake layer and membrane resistances are similar in magnitude with an optimal lumen radius that is dependent upon the average permeate flux.</li> </ul>                                | (Mondor and Moresoli, 2006)                  |
| Crossflow   | Outside and Inside | <ul style="list-style-type: none"> <li>• Hollow fibre module immersed in a reactor filled with water</li> </ul>                                                                                                         | $d_i=0.25$ mm<br>$d_o=0.43$ mm<br>$\phi=0.4, 0.6, 0.8$                                                                     | $\phi$                                                     | <ul style="list-style-type: none"> <li>• At very high packing density, filtration flux will decrease dramatically and filtration behaviour will occur preferentially at the bottom of the fibre.</li> <li>• High packing density will result in non-uniform filtration along the fibre and this lead to uneven particle deposition and cake growth.</li> </ul> | (Gunther et al., 2010, Gunther et al., 2012) |
| Direct-flow | Outside            | <ul style="list-style-type: none"> <li>• Clean water used as feed liquid</li> </ul>                                                                                                                                     | Standard parameters<br>$d_i=0.7$ mm<br>$d_o=1.3$ mm<br>$\Gamma=600 \mu\text{m}$<br>$L=2$ m<br>$R_m=10^{11} \text{ m}^{-1}$ | One changed at a time<br>$d_i$<br>$d_o$<br>$\Gamma$<br>$L$ | <ul style="list-style-type: none"> <li>• The design of fibre dimensions should be determined by considering the lumen and shell side pressure drops.</li> <li>• Short fibres with large inner diameter and small thickness (relatively small outer diameter), as long as the fibre mechanical strength is allowed, are found to be efficient.</li> </ul>       | (Kim et al., 2012a, Kim et al., 2012b)       |

|             |         |                                                                                                                                                                                                              |                                                                                                                                  |                   |                                                                                                                                                                                                                                                                                                                                 |                     |
|-------------|---------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------|-------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------|
| Crossflow   | Inside  | <ul style="list-style-type: none"> <li>• Backflushing flow and backflushing pressure studied</li> <li>• Operated under constant flow or constant pressure condition</li> </ul>                               | $d_i=0.6\text{mm}$<br>$d_o=1.10\text{mm}$<br>$L=0.6, 1.2 \text{ and } 1.8\text{m}$                                               | $L$               | <ul style="list-style-type: none"> <li>• An increase of the fibre length will result in a corresponding increase of the flow or pressure to achieve the same backwash effect.</li> <li>• The cleaning time should be extended for longer fibres.</li> </ul>                                                                     | (Wang et al., 2014) |
| Direct-flow | Outside | <ul style="list-style-type: none"> <li>• Submerged hollow fibre module</li> <li>• Flow in lumen is assumed laminar flow in stationary state</li> </ul>                                                       | $d_i=0.5\text{-}1 \text{ mm}$<br>$d_i/d_o=1/2$<br>$L=1, 1.5 \text{ and } 2 \text{ m}$<br>$R_m = 6 \times 10^{11} \text{ m}^{-1}$ | $d_i$<br>$L$      | <ul style="list-style-type: none"> <li>• An increase in fibre length led to more non-uniform local flux distribution and more severe membrane fouling.</li> <li>• A reasonable long fibre length and larger fibre diameter is preferred to increase the water production efficiency at fixed average operating flux.</li> </ul> | (Li et al., 2015)   |
| Direct-flow | Inside  | <ul style="list-style-type: none"> <li>• 2-D channel and 3-D filter pipe</li> <li>• Fibre wall thickness ignored</li> <li>• Not a real filtration unit</li> <li>• Mathematical model investigated</li> </ul> | Dimensionless quantities (See Section 3.4)                                                                                       | $k_m$<br>$\gamma$ | <ul style="list-style-type: none"> <li>• TMP is larger at the entrance and capped end, with a dip in the middle</li> <li>• Low permeability is preferred</li> <li>• Module separation distance influences the filtration performance</li> </ul>                                                                                 | (Herterich, 2015)   |

Note: Fibre outside diameter  $d_o$ , fibre inside diameter  $d_i$ , fibre inside radius  $a$ , fibre outside radius  $b$ , fibre length  $L$ , fibre permeance  $k_m$ , void fraction  $\epsilon$ , fibre packing density  $\phi$ , spacing parameter

$\gamma$ , and imposed average flux  $J_{av}$  will be defined correspondently in Chapter 4 and 5.

As presented in Table 3.3, there are a few attempts to analyse the influences of geometry of hollow fibre module design on the overall filtration behaviours mostly with regard to crossflow operation. Agreement has been reached that a shorter length fibre with larger lumen diameter is desirable so as to increase the filtration throughput per unit membrane area (Serra et al., 1998, Soltanieh and Gill, 1983, Kim et al., 2012b, Kim et al., 2012a, Wang et al., 2014, Li et al., 2015). Long fibres with small inner diameters will generate large differences in local flux and leads to a rapid increase in transmembrane pressure (Kim et al., 2012b, Kim et al., 2012a). Additionally, any increase in fibre length beyond a certain value will lead to more non-uniform local flux distribution and severe membrane fouling (Li et al., 2015). Moreover, the negative effect of having longer fibres will be even worse at high module packing (low void fraction) because the filtration flux will decrease dramatically and filtration behaviour will occur preferentially at the capped-end (Yoon et al., 2004, Gunther et al., 2010, Gunther et al., 2012). However, some researchers have found that longer fibres foul more slowly than shorter fibres due to lower fluxes (Carroll and Booker, 2000). Therefore the selection of optimized module design including module packing (void fraction) and fibre length is crucial to the overall filtration performance. Also the ratio of inside to outside diameter has been shown to have an influence with the filtration productivity being different at various ratios (Chang et al., 2006, Serra et al., 1998). This indicates the potential importance of fibre wall thickness. To date, although a general understanding has been achieved by researchers (Mondor and Moresoli, 2006, de Ven et al., 2008, Chang et al., 2006, Oh et al., 2007b, Serra et al., 1998, Kim et al., 2012b, Kim et al., 2012a, Gunther et al., 2010, Gunther et al., 2012, Soltanieh and Gill, 1983, Li et al., 2015, Wang et al., 2014, Yoon et al., 2004) that the geometry of the fibre as well as operating conditions is crucial to the membrane filtration efficiency, there remains the need for systematic modelling at full scale. In particular an

understanding of the effects of fibre wall thickness, void fraction and imposed average fluxes is needed.

Although properties of the feed components (e.g. particle size, size distribution and fluid properties) and transport phenomena may be key contributors to the filtration performance and membrane fouling, the decline in permeate flowrate is reported to be largely influenced by module design features until substantial fouling has occurred (Carroll and Booker, 2000). Therefore this thesis set out to investigate the dependence of filtration performance and fouling behaviours on fibre dimensions and module designs under conditions where axial feature are significant. The study starts by building upon an existing literature model (Herterich, 2015) which will be introduced in next section.

Firstly this thesis adopts a 2D mathematical model from literature to develop a direct-flow filtration model within a 3D axisymmetric pipe. Later in Chapter 5 a new mathematical model is developed with realistic industrially relevant dimensions, including a finite wall thickness. The simple mathematical 2D model from the literature are presented here so that the readers can fully understand the research background of this thesis.

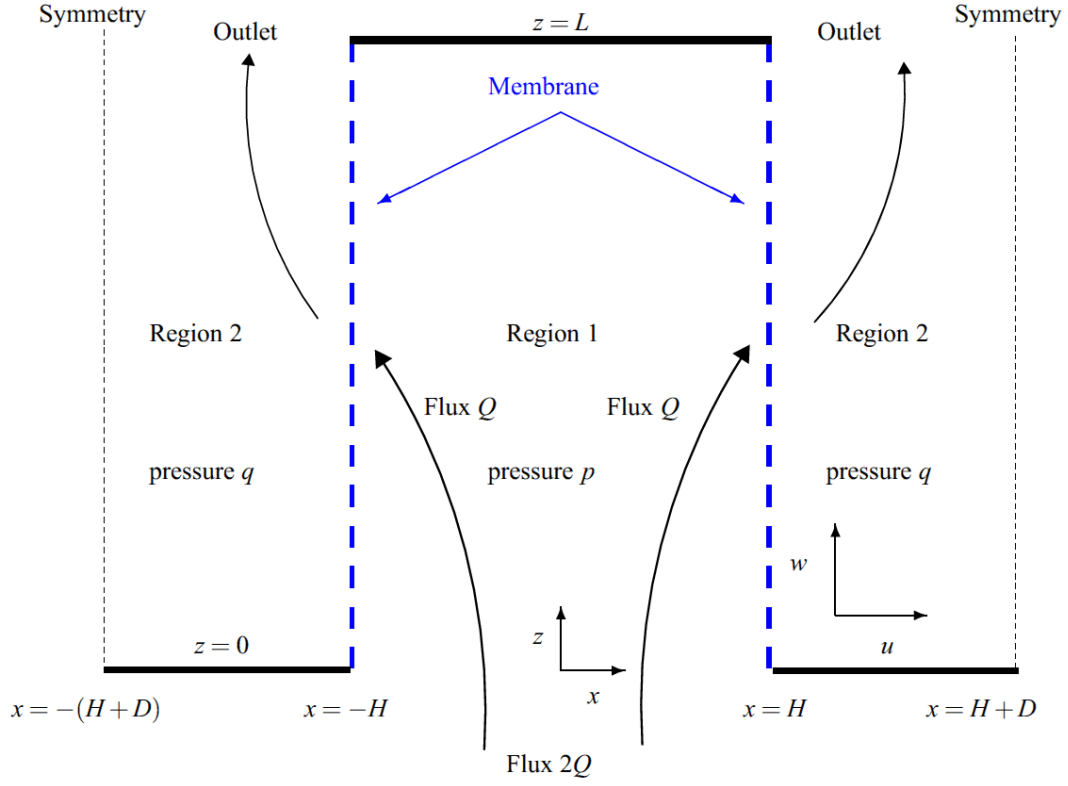
### **3.4 2D direct-flow channel**

In existing literature, studies have approached towards direct-flow filtration investigation. Hurwitz modelled the direct-flow process by considering the flow inside a single porous tube with a capped-end by solving the flow in the asymptotic limits of low permeability and low Reynolds numbers (Hurwitz, 1989). Sanaei et al. derived a theoretical model for the flow behaviour in a pleated filter containing a series of permeable pleated fibres to explore the impact

of pore constriction on filter blocking mechanisms (Sanaei et al., 2016). However, most studies have not considered the coupling effect due to the flow on the permeate side.

A recent mathematical study (Herterich, 2015) as mentioned in Table 3.3 developed a 2D direct-flow model to explore the variation in the axial dependence of transmembrane pressure differences. However, the author did not take the wall thickness into account and only covered a limited set of module geometric parameters. In hollow fibre membrane modules, the wall thickness occupies a notable portion of the overall device space (circa 30%) so in real engineering it influences the module performance. By building upon this mathematical model, but not avoiding the wall thickness, this thesis explores direct-flow filtration for a range of a realistic hollow fibre module dimensions and operating conditions for pressure driven inside flows.

Herterich (2015) started his modelling by considering a 2D analogue of the direct-flow device in which the flow enters a single 2D channel (Figure 3.2), of length  $L$  and width  $H$ , with permeable walls. The wall permeability,  $k$ , is constant, and the end of the channel is capped. A Cartesian coordinate system  $(x, z)$  is applied to represent the system, where  $x$  denotes the direction perpendicular to the permeable channel walls and  $z$  denotes the distance along the channel. Fluid enters the channel (Region 1) at a fixed flux  $2Q$ , which passes through the membrane side walls, and into the permeate region (Region 2). Since the end of the channel is blocked, the entire fluid must flow through the permeable walls from Region 1 into Region 2 (net flux  $Q$  through each side of the channel by symmetry).



**Figure 3.2:** Schematic of the flow within a single direct-flow 2D channel. Solid lines indicate impermeable walls, blue dashed lines indicate permeable walls, and the dotted line indicates symmetry in the flow. Flow enters from the bottom and passes through the permeable walls. Beyond the capped end, the fluid continues to an outlet following the black arrows. The inside of the membrane-walled channel is referred as Region 1, while that outside is Region 2 (Herterich, 2015).

For an array of such channels, with a centre-centre separation  $2(H + D)$ , a symmetry condition can be imposed at  $x = \pm(H + D)$ . The fluid velocity field in the  $(x, z)$  direction is  $\vec{u} = (u(x, z), w(x, z))$ , while the (spatially variable) pressure inside the channel is  $p$  with that outside the channel denoted by  $q$ . The quantity of most interest is the TMP,  $p - q$  at  $x = \pm H$ ; The greater the spatial uniformity in the TMP, the more uniformly the membrane area is used for filtration. The aspect ratio of the module:  $\delta = H/L \ll 1$  and the module space parameter  $\ell =$

$D/H$ . Since the reduced Reynolds number is small and the channel is thin, the fluid flow obeys the lubrication equations (Herterich et al., 2014, Herterich, 2015). After non-dimensionalizing the variables, i.e. scaling with regard to the channel length,  $L$ , and the typical axial velocity,  $W$ , the modelling equations were presented in non-dimensional form as follows:

In Region 1 ( $0 < x < 1$  and  $0 < z < 1$ ), the lubrication equations are

$$\frac{\partial u}{\partial x} + \frac{\partial w}{\partial z} = 0, \quad (3.1)$$

$$\frac{\partial^2 w}{\partial x^2} = \frac{dp}{dz}. \quad (3.2)$$

The equations are subject to typical boundary conditions that reflect the symmetry of the flow, the flux of fluid and no flow through the impermeable end. A filtration velocity is assumed as proportional to the transmembrane pressure difference and permeability of the membrane (i.e. Darcy flow) (Kedem and Katchalsky, 1958). In general, at a permeable wall there is a tangential slip velocity, whose magnitude is determined by a Neumann boundary condition such as that given in (Beavers and Joseph, 1967). However, it has been found that this slip is not significant for a wide range of membranes (Altena and Belfort, 1984, Shipley et al., 2010) and so a no-slip boundary condition was assumed. The boundary conditions are expressed as

Symmetry:

$$\frac{\partial u}{\partial x} = u = 0, \quad \text{on } x = 0, \quad (3.3)$$

No axial wall slip:

$$w = 0, \quad \text{on } x = 1, \quad (3.4)$$

Permeate flow:

$$u = k(p - q), \quad \text{on } x = 1, \quad (3.5)$$

Fluid flux:

$$\int_0^1 w \, dx = Q, \quad \text{at } z = 0, \quad (3.6)$$

No Fluid outflow:

$$w = 0, \quad \text{at } z = 1. \quad (3.7)$$

As the capped end is impermeable all injected fluid must pass through the membrane, therefore

$$\int_0^1 u \, dz = Q \quad \text{at } x = 1. \quad (3.8)$$

Similarly in Region 2 ( $1 < x < 1 + \ell$  and  $0 < z < 1$ ), the fluid also obeys the lubrication equations

$$\frac{\partial u}{\partial x} + \frac{\partial w}{\partial z} = 0, \quad (3.9)$$

$$\frac{\partial^2 w}{\partial x^2} = \frac{dq}{dz}. \quad (3.10)$$

The boundary conditions are expressed as

Symmetry:

$$\frac{\partial w}{\partial x} = u = 0, \quad \text{on } x = 1 + \ell, \quad (3.11)$$

No axial wall slip:

$$w = 0, \quad \text{on } x = 1, \quad (3.12)$$

Permeate flow:

$$u = k(p - q), \quad \text{on } x = 1, \quad (3.13)$$

Fluid flux:

$$w = 0, \quad \text{at } z = 0, \quad (3.14)$$

No Fluid outflow:

$$q = 0, \quad \text{at } z = 1. \quad (3.15)$$

Note that the permeate-flow boundary conditions are the same, enforcing continuity of flux across the membrane.

The author solved the equations analytically using both sets of symmetry and no axial wall-slip conditions to obtain velocity profiles in terms of the pressures in both Regions. In Region 1,

$$w = \frac{1}{2} \frac{dp}{dz} (x^2 - 1), \quad (3.16)$$

$$u = \frac{1}{6} \frac{d^2 p}{dz^2} (3 - x^2)x, \quad (3.17)$$

while in Region 2,

$$w = \frac{dq}{dz} \left[ \frac{1}{2} x^2 - (1 + \ell)x + \ell + \frac{1}{2} \right], \quad (3.18)$$

$$u = \frac{d^2 q}{dz^2} \left[ -\frac{1}{6} x^3 + \frac{1}{2} (1 + \ell)x^2 - \left( \ell + \frac{1}{2} \right) x - \frac{1}{3} (1 + \ell)^3 \right. \\ \left. + \left( \ell + \frac{1}{2} \right) (\ell + 1) \right]. \quad (3.19)$$

Substituting the transverse velocity expression into the boundary conditions for the permeate flux through the membrane, gives a set of second-order ordinary differential equations (ODEs) for the pressures,  $p$  and  $q$ . Combine the velocities and the boundary conditions, the system of ODEs is solved analytically

$$p(z) = \frac{1}{1 + \ell^3} [\ell^3 \mathcal{M}(z) + 3Q(1 - z) + \mathcal{M}_0], \quad (3.20)$$

$$q(z) = \frac{1}{1 + \ell^3} [-\mathcal{M}(z) + 3Q(1 - z) + \mathcal{M}_0], \quad (3.21)$$

where

$$\mathcal{M}(z) = \text{sech}(C_1) \left\{ \mathcal{M}_0 \cosh(C_1 z) + \frac{3Q}{C_1} \sinh[C_1(1 - z)] \right\}, \quad (3.22)$$

$$\mathcal{M}_0 = \frac{3Q}{C_1} \left[ \frac{1}{\ell^3} \coth(C_1) + \text{csch}(C_1) \right], \quad (3.23)$$

$$C_1 = \sqrt{3k \left( 1 + \frac{1}{\ell^3} \right)}. \quad (3.24)$$

One of the implications of the above equations is that the local TMP ( $\mathcal{M}(z)$ ) is dependent upon the relative channel size. If both feed and permeate channels are the same (i.e.  $\ell = 1$ ) then the TMP profile shows a distinct minimum but the variation between the minimum and maximum value is modest. If  $\ell < 1$ , the curve becomes skewed and the TMP profile shows the largest value at the impermeable capped-end, whereas for  $\ell > 1$ , the section of the membrane walls nearest the entrance of the channel has the largest TMP values. Such behaviours are investigated in a greater detail for more realistic geometries in Chapter 4 and 5.

## **3.5 Conclusion**

In this chapter, a review has been given specifically to direct-flow filtration, including the module formats, operating modes, materials and modules, fouling and cleaning issues. Specifically, a comprehensive literature survey has been carried out to address the importance of the module design in controlling the filtration performance unique to direct-flow filtration systems. From the survey, the development of modern membrane market is based on direct-flow filtration which is still at a relative young age. The underlying operational consideration and design aspects are not completely understood and there remains gap and challenge to tackle the fouling issues in direct-flow filtration operations, especially in the inside feed format.

This thesis therefore sets out to investigate the dependence of filtration performance and fouling behaviours upon fibre/module designs for direct-flow filtration under various operating conditions.

# Chapter 4

## Simple Direct-Flow Filtration Devices

This chapter aims to develop a simple direct-flow filtration model in a 3D pipe to describe direct-flow filtration behaviours. Without considering the wall thickness effect, the 3D pipe model aims to predict the operating regimes of direct-flow filtration devices so that one can maximize the spatial uniformity in the transmembrane pressure and thus optimize the use of the entire membrane area. The results will be a useful simplification in the subsequent chapters, especially for the contents in Chapter 5.

## 4.1 Introduction

In Chapter 3, a 2D direct-flow filtration model developed in existing literature (Herterich, 2015) was introduced. The author considered the behaviour of the pressure and TMP profiles in a 2D direct-flow filtration channel for various module spacing parameters, fluxes, and permeabilities in a dimensionless way. This chapter develops a model in an 3D axisymmetric pipe to further investigate and quantify direct-flow filtration performance by examining the uniformity and productivity of the filtration under given operating conditions. To compare our modelling results with the existing literature, the model in this chapter will also be presented in a dimensionless way.

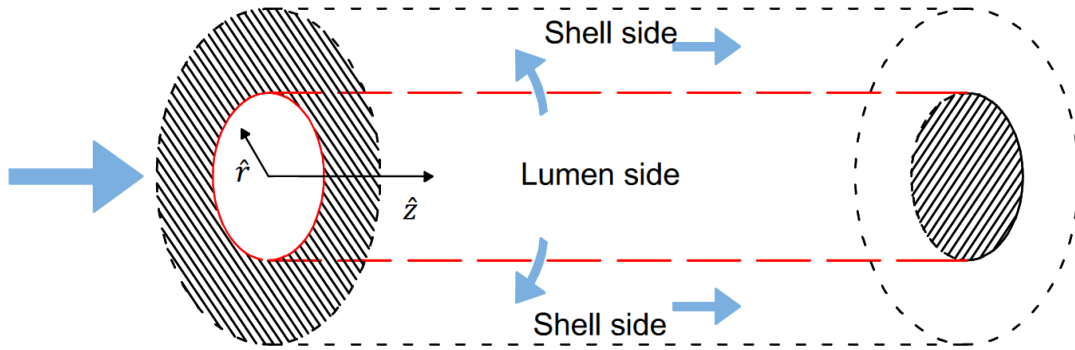
## 4.2 Model development: direct-flow pipe

Figure 4.1 illustrates the 3D axisymmetric direct-flow pipe geometry. The pipe has a permeable wall (dashed red lines) with a cross-section radius of  $\hat{R}$  and pipe length  $\hat{L}$ . The aspect ratio of this model is  $\delta = \frac{\hat{R}}{\hat{L}} \ll 1$ . The hashed black areas indicate non-permeable walls where fluid cannot pass through. In this axisymmetric direct-flow pipe geometry, the position of the symmetry boundary is no longer simple and will, in general, depend on the packing of the pipes (square, hexagonal, etc.). For simplicity, a symmetry condition is assumed in this pipe model so that an axisymmetric coordinate system  $(\hat{r}, \hat{z})$  can be used at a particular radial coordinate. The radial coordinate  $\hat{r}$  replaces the Cartesian coordinate  $\hat{x}$  and the  $\hat{z}$  coordinate remains unchanged from that shown in Figure 3.2. The pressure in the lumen side is  $\hat{p}$ , and the pressure in the shell side is  $\hat{q}$ . The fluid inflow is  $\hat{Q}$  and the permeable wall permeability is  $\hat{k}$ . The fluid velocity along  $\hat{z}$

direction is  $\hat{w}$  and the fluid velocity along the  $\hat{r}$  direction is  $\hat{u}$ . To complete the specification of the system assumed that the radial position of the dotted black line is  $\hat{D}$ . Thus non-dimensionalization of the parameters gives:

$$\begin{aligned} z &= \frac{1}{L} \hat{z}, & r &= \frac{1}{\delta L} \hat{r}, & w &= \frac{1}{W} \hat{w}, & u &= \frac{1}{\delta W} \hat{u} \\ p &= \frac{\delta^2 L}{\mu W} \hat{p}, & q &= \frac{\delta^2 L}{\mu W} \hat{q}, & Q &= \frac{1}{\delta^2 L^2 W} \hat{Q}, & k &= \frac{\mu}{\delta^3 L} \hat{k}. \end{aligned} \quad (4.1)$$

The lumen side region can be denoted as  $0 \leq r \leq 1$ , the module spacing parameter in this 3D pipe model is  $\ell = D/R$  and therefore the shell side region can be denoted as  $0 \leq r \leq 1 + \ell$ .



**Figure 4.1:** Schematic of the flow within a single 3D direct-flow axisymmetric pipe. Red dashed lines indicate the permeable walls, the black dashed lines indicate the ‘symmetry’ between two pipes, and the hashed areas indicate non-permeable areas. Flow enters from the lumen side and passes through the permeable walls. Beyond the capped-end, the fluid continues to an outlet following the blue arrows.

The aspect ratio is small, therefore the fluid in this model also obeys the lubrication equations. In Region 1,

$$\frac{1}{r} \frac{\partial}{\partial r} (ru) + \frac{\partial w}{\partial z} = 0, \quad (4.2)$$

$$\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial w}{\partial r} \right) = \frac{dp}{dz}, \quad (4.3)$$

For  $0 < r < 1$  and  $0 < z < 1$ . The boundary conditions are:

$$\text{Symmetry:} \quad \frac{\partial w}{\partial r} = 0; u = 0, \quad \text{on } r = 0, \quad (4.4)$$

$$\text{No axial wall slip:} \quad w = 0, \quad \text{on } r = 1, \quad (4.5)$$

$$\text{Permeate flow:} \quad u = k(p - q), \quad \text{on } r = 1, \quad (4.6)$$

$$\text{Fluid flux:} \quad 2\pi \int_0^1 wr \, dr = Q, \quad \text{at } z = 0, \quad (4.7)$$

$$\text{No fluid outflow:} \quad w = 0, \quad \text{at } z = 1. \quad (4.8)$$

In Region 2, the fluid again obeys the lubrication equations,

$$\frac{1}{r} \frac{\partial}{\partial r} (ru) + \frac{\partial w}{\partial z} = 0, \quad (4.9)$$

$$\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial w}{\partial r} \right) = \frac{dq}{dz}, \quad (4.10)$$

For  $1 < r < 1 + \ell$  and  $0 < z < 1$ . For this region, the boundary conditions read,

$$\text{Symmetry:} \quad \frac{\partial w}{\partial r} = 0; u = 0, \quad \text{on } r = 1 + \ell, \quad (4.11)$$

$$\text{No axial wall slip:} \quad w = 0, \quad \text{on } r = 1, \quad (4.12)$$

$$\text{Permeate flow:} \quad u = k(p - q), \quad \text{on } r = 1, \quad (4.13)$$

$$\text{Fluid flux:} \quad w = 0, \quad \text{at } z = 0, \quad (4.14)$$

$$\text{No fluid outflow:} \quad q = 0, \quad \text{at } z = 1. \quad (4.15)$$

The equations for 3D pipe can be solved analytically by using both sets of symmetry conditions together with the no-slip wall conditions. The fluid flow is, in terms of the pressure gradient, as follows: in Region 1,

$$w = \frac{1}{4} \frac{dp}{dz} (r^2 - 1), \quad (4.16)$$

$$u = \frac{1}{16} \frac{d^2 p}{dz^2} (2r - r^3), \quad (4.17)$$

In Region 2,

$$w = \frac{1}{4} \frac{dq}{dz} [r^2 - 2(1 + \ell)^2 \log r - 1], \quad (4.18)$$

$$u = \frac{1}{16} \frac{d^2 q}{dz^2} \left\{ 2r - r^3 + (1 + \ell)^2 \left[ -2r + 4r \log r - (1 + \ell)^2 \left[ -\frac{3}{r} + \frac{4}{r} \log(1 + \ell) \right] - \frac{2}{r} \right] \right\}. \quad (4.19)$$

Substituting the transverse velocity expression into the boundary conditions for the permeate flux through the membrane, gives another set of second-order ordinary differential equations (ODEs) for the pressures,  $p$  and  $q$ . Combine the velocities and the boundary conditions, the system of ODEs is solved analytically to give

$$p(z) = \frac{1}{1 + \Lambda} \left[ \Lambda \mathcal{N}(z) + \frac{8Q}{\pi} (1 - z) + \mathcal{N}_0 \right], \quad (4.20)$$

$$q(z) = \frac{1}{1 + \Lambda} \left[ -\mathcal{N}(z) + \frac{8Q}{\pi} (1 - z) + \mathcal{N}_0 \right], \quad (4.21)$$

where

$$\mathcal{N}(z) = \text{sech}(C_2) \left\{ \mathcal{N}_0 \cosh(C_2 z) + \frac{8Q}{\pi C_2} \sinh[C_2(1 - z)] \right\}, \quad (4.22)$$

$$\mathcal{N}_0 = \frac{C_2 Q}{2\pi k(1 + \Lambda)} \coth(C_2) + \frac{8Q}{\pi C_2} \text{csch}(C_2), \quad (4.23)$$

$$C_2 = \sqrt{16k \left( 1 + \frac{1}{\Lambda} \right)}, \quad (4.24)$$

$$\begin{aligned} \Lambda = \Lambda(\ell) = & -1 + 4(1 + \ell)^2 \\ & - [3 - 4 \log(1 + \ell)](1 + \ell)^4 > 0. \end{aligned} \quad (4.25)$$

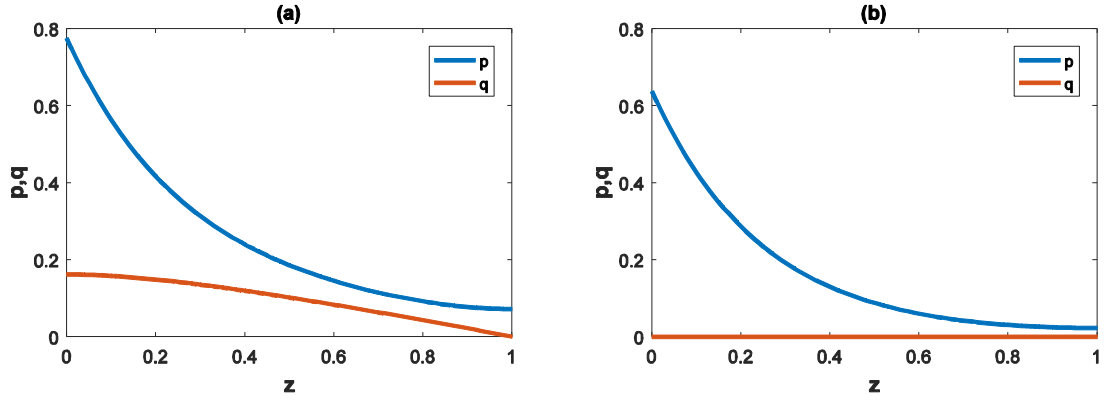
### 4.3 Discussion of results and optimization

In all filtration scenarios (dead-end, crossflow, and direct-flow), the main quantity of interest is the volumetric flux across the porous membrane, which depends on the local transmembrane pressure: the more spatially uniform the TMP, the more effectively the membrane area is used

during filtration. As reported in literature, the TMP profile is a complex function of operating flux, membrane wall permeability and, crucially, the packing density of the fibres within the device (Karode, 2001, Herterich, 2015). Although many authors have studied the impact of particle accumulation upon these parameters, little emphasis has been placed on explaining the influence of pressure profiles, especially during direct-flow filtrations. Therefore this study started with a mathematical approach demonstrating how the pressure profiles changes in response to fibre spacing and operating conditions. To simplify this research, the axisymmetric direct-flow pipe model is studied under non-fouling conditions, i.e. all the fluid is considered as clean water. Considering clean water keeps the permeability constant and thus all non-uniform distribution of local flux only arises from the non-uniformity of the transmembrane pressure differences.

### **4.3.1 Pressure and TMP profiles**

Firstly, the behaviours of the pressure profiles in the lumen and shell side are studied under various module spacing parameters. As shown in Figure 4.2, when  $\ell = 1$ , there is a negative pressure gradient in the  $z$ -direction on both sides with a generally steeper gradient in the lumen side. The pressure in the lumen is higher than the pressure in the shell side which indicates there is a flow of fluid in the positive  $z$ -direction on both side of the membrane and a flow through the membrane, as expected. When  $\ell = 10$ , the outside pressure,  $q$ , is effectively zero everywhere, equivalent to a quiescent bath (i.e. its pressure is unaffected by the flow from the pipe).

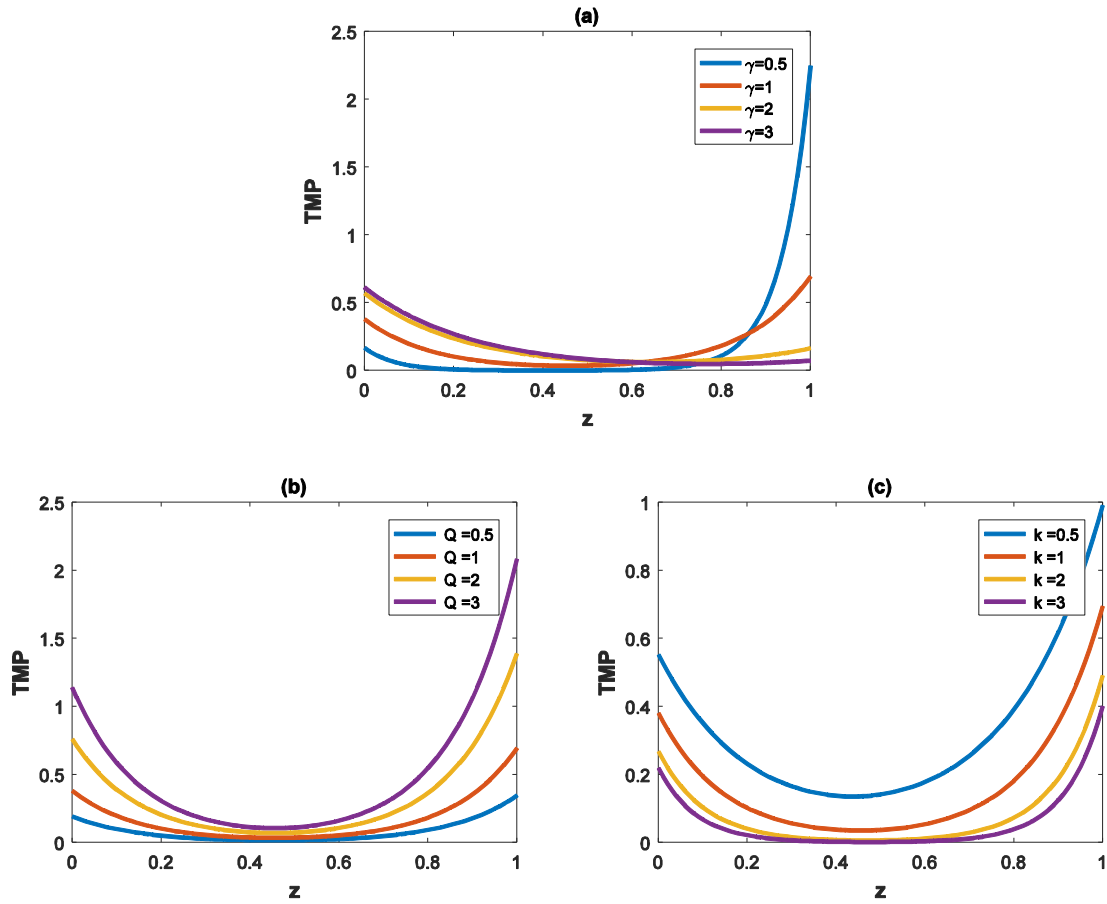


**Figure 4.2:** Variation of fluid pressure in the lumen side,  $p$ , and in the shell side,  $q$ , under the condition of permeability  $k = 1$  and the flux  $Q = 1$ , with dimensionless module spacing parameters **(a)**  $\ell = 1$  and **(b)**  $\ell = 10$ .

The pressure profiles of the axisymmetric pipe model (Figure 4.2) show qualitative similarities to that of the 2D direct-flow channel in literature, but differ in detail. Compare to the 2D channel, the gradient of the lumen side pressure is steeper in this 3D pipe model and filtration tends to be higher at the inlet and less towards the capped-end. The difference between 2D channel and 3D pipe directed this study towards a focus on the geometrical differences. For the 2D channels (as presented in Section 3.4) the module spacing parameter,  $\ell$ , could also be counted as a cross-sectional area ratio of Region 2 to Region 1. However, the situation differs in this pipe configuration,  $\ell = D/R = 1$  does not indicate an equal cross-sectional area for the lumen and the shell side of the pipe. Therefore, another dimensionless module spacing parameter,  $\gamma$ , for the pipe configuration on area basis is defined:

$$\gamma = \frac{\text{Area in shell side}}{\text{Area in lumen side}} = \frac{(1 + \ell)^2 - 1}{1} = 2\ell + \ell^2. \quad (4.26)$$

As discussed, the quantity of real interest is not  $p$  and  $q$  separately but rather the TMP,  $p - q$ , which is simply given by the function  $\mathcal{N}(z)$ : the more spatially uniform the TMP the more effective the use of the available membrane area. Therefore, TMP profiles are studied at various conditions of  $\gamma$ ,  $Q$ , and  $k$  (in Figure 4.3).



**Figure 4.3:** Profiles of transmembrane pressure difference (TMP), in a capped-end axisymmetric pipe. Variation with **(a)** module spacing parameter,  $\gamma$  (with  $Q = 1$ ,  $k = 1$ ); **(b)** flux,  $Q$  (with  $\gamma = 1$ ,  $k = 1$ ); and **(c)** permeability,  $k$ , (with  $\gamma = 1$ ,  $Q = 1$ ).

As shown in Figure 4.3a, varying the module spacing parameter,  $\gamma$ , affects not only the magnitude of the TMP but also its shape across the module length,  $z$ . When  $\gamma > 1$ , the permeate (shell) side area is larger than the feed (lumen) side and the pipe walls nearest the entrance of the module has the largest TMP and then the TMP decreases monotonically, as the permeate side behaves like a quiescent bath (as seen in Figure 4.2b). When  $\gamma \sim 1$  (permeate side area is equal to the feed side area), the TMP assumes a profile with a central minimum. For  $\gamma < 1$ , when shell side is relatively small compared with the feed side, the TMP is largest at the impermeable capped-end. This results in the majority of the filtration occurring further down the pipe length (towards the capped-end), which is a situation to be avoided since a large area of the membrane is not being used to its full potential.

In both Figure 4.3b and c, the TMP profiles are generally having a convex shape with a central minimum, which also indicates that more of the fluid is filtered near the entrance and capped end. As  $Q$  increases and  $k$  decreases, the convex shape becomes even more obvious. For small values of  $Q$ , the TMP is small but still relatively large at the ends. When  $Q$  is larger, the TMP retains the similar qualitative shape, but varies in absolute terms to a larger degree along the pipe length,  $z$ . The TMP has a qualitative similar profile at varying  $k$  values, although higher pressures are required for smaller permeabilities (Figure 4.3c). This should be expected because in direct-flow filtration with constant flux operation, all the fluid must be filtered through the permeable membrane walls. The variation between minimum and maximum TMPs increases with decreasing  $k$  values.

When  $Q$  is large and  $\gamma$  is small, the TMP generally retains the qualitative shape, but varies to a larger degree absolutely (i.e. a larger variation between the minimum and maximum TMP values) along the pipe length for larger flux,  $Q$ , and lower permeabilities,  $k$ . Therefore under

constant flux operation in direct-flow filtration, this suggests that using low-permeability material and operating at low fluxes results in the filtration area being more evenly

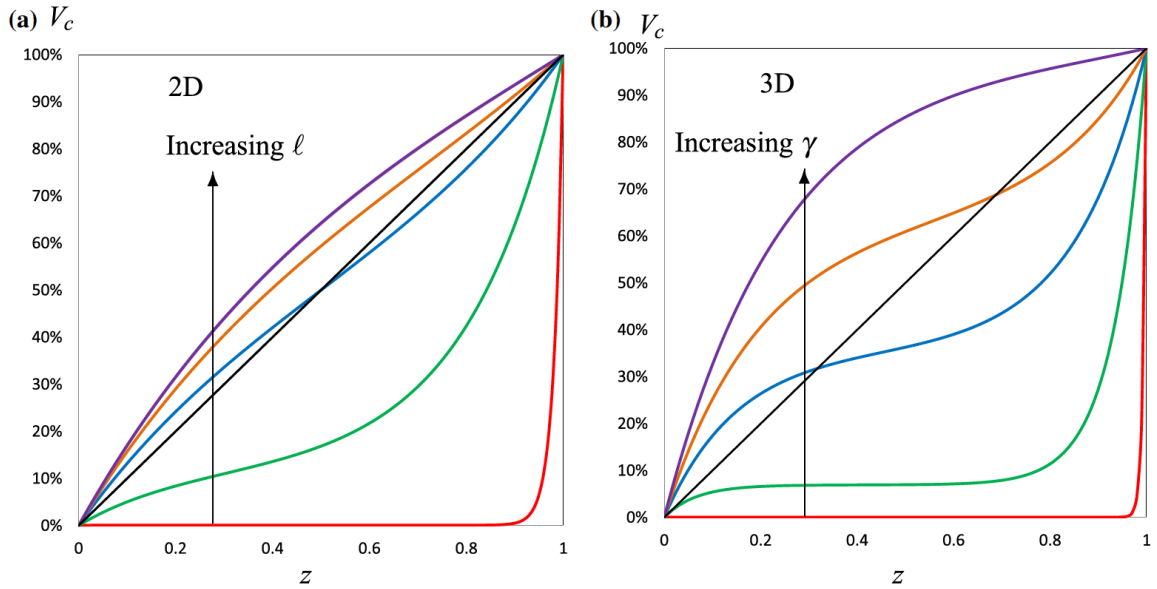
### 4.3.2 Cumulative fluid volume

This study has so far considered the behaviour of the pressure and TMP profiles in an axisymmetric direct-flow pipe, both lumen side and shell side for a range of (a) module spacing parameter,  $\gamma$ ; (b) fluxes,  $Q$ ; and (c) permeabilities,  $k$ . In this section, the study continues to explore how the axisymmetric pipe model can be used to understand how a direct-flow device can be arranged to operate optimally and, in particular, making good use of the available membrane area.

As discussed for small module spacing parameter values, the TMP is low for most of the permeable walls and then rapidly rises to a high value as the capped-end is approached ( $z = 1$ ). In this case, the majority of the filtration occurs at the end of the pipe and highlights the constraints that the impermeable capped-end enforces on the TMP. In order to investigate direct-flow filtration devices on the geometrical configurations in both 2D channel and 3D axisymmetric pipe, the following studies will consider both the characterized areal spacing parameters,  $\ell$  (in 2D) and  $\gamma$  (in 3D).

In Figure 4.4, the accumulated filtration volume percentage,  $V_c$ , clearly demonstrates the restrictions that arise when the module spacing parameter is too small. With a spacing parameter of 0.1, there is almost no flux through the membrane wall over the first 95% of the length and all of the filtration essentially occurs in the last 5% near the impermeable capped-end. For both 2D and 3D, this is a situation to be avoided since a large area of the membrane is not being utilized

and the very high local permeate flux at the end would lead to intense particle deposition. It is important to note that when the module spacing parameter becomes too small the packing arrangement chosen (e.g. a hexagonal or square array) in 3D pipe model plays a role. At this point the symmetry boundary conditions will no longer be appropriate and would need to be solved for the flow in the complete array using computational fluid dynamics tools. When the module spacing parameter is large, the feed channels or pipes are widely separated, and any further increase in the separation will have only a small effect on the flux profiles.



**Figure 4.4:** The cumulative fluid volume transported through the fibre walls,  $V_c$  (%), versus axial distance,  $z$ , in **(a)** a 2D channel (as shown in Section 3.4) and **(b)** an axisymmetric pipe for various values of module spacing parameters ( $\ell$  and  $\gamma$  respectively). In both cases, the varying quantity assumes the values 0.1, 0.5, 1, 1.5 and 5, respectively at  $Q = 1$  and  $k = 1$ . (Optimal performance would correspond to a linearly increasing cumulative flux, as shown by the straight black line.)

In the opposing regime of large areal spacing parameter, one should also note that in both 2D and 3D plots (Figure 4.4), the profiles of percentage volume of cumulative filtered fluid will overlap when the module spacing parameter is large (i.e.  $\ell = 5$  with  $\ell = 10$  and  $\gamma = 5$  with  $\gamma = 10$ ). When  $\ell$  in 2D or  $\gamma$  in 3D are large (typically greater than 2), the feed channels or pipes are widely separated and thus further increase of  $\ell$  or  $\gamma$  larger than 2 will have a very small effect on the flux profile. Continuing to increase the module spacing parameter value beyond approximately 3 has only a small influence on the TMP plots and the overall filtration process. However the space in a module is not being used effectively.

### 4.3.3 Filtration uniformity and productivity

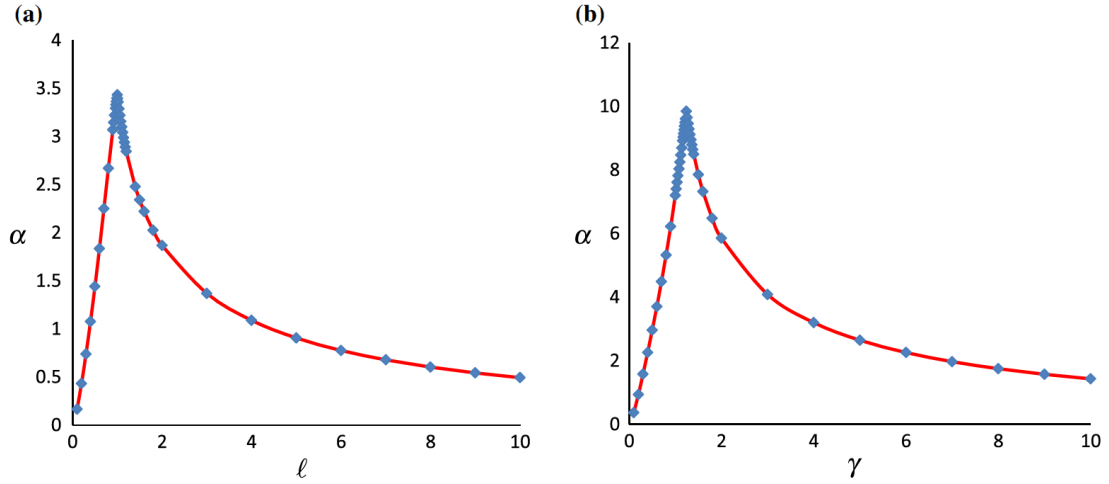
The results of previous analysis show that when fibres are packed too closely to one another the membrane area is not effectively used in a uniform manner. However, when the fibres are spaced far apart, although the area is more uniformly used, fewer fibres can be packed into a single direct-flow device. Attention is now given to the question of how closely the fibres should be placed to optimize the operating efficiency of a direct-flow device. This study framed this mathematically by introducing the design variable

$$\alpha = \frac{P_0}{P_m} \frac{Q}{(1 + S_p)}, \quad (4.27)$$

where  $S_p$  is the dimensionless module spacing parameter ( $\ell$  in 2D and  $\gamma$  in 3D),  $Q$  is the fluid flux as defined earlier,  $P_m$  is defined as the maximum pressure difference,  $P_0 - q_L$ , for a chosen set of conditions, and  $P_0$  is the operating pressure at the inlet. The term  $Q/P_m$  is a constant for a given channel or pipe and equates to a mean dimensionless permeability. Thus,  $P_0 Q/P_m$

expresses the productive capacity for a given set of conditions. Overall, the parameter  $\alpha$  is a measure of the productive capacity per unit size of the module, i.e. a module productivity factor. Approaches that maximizes this factor were determined.

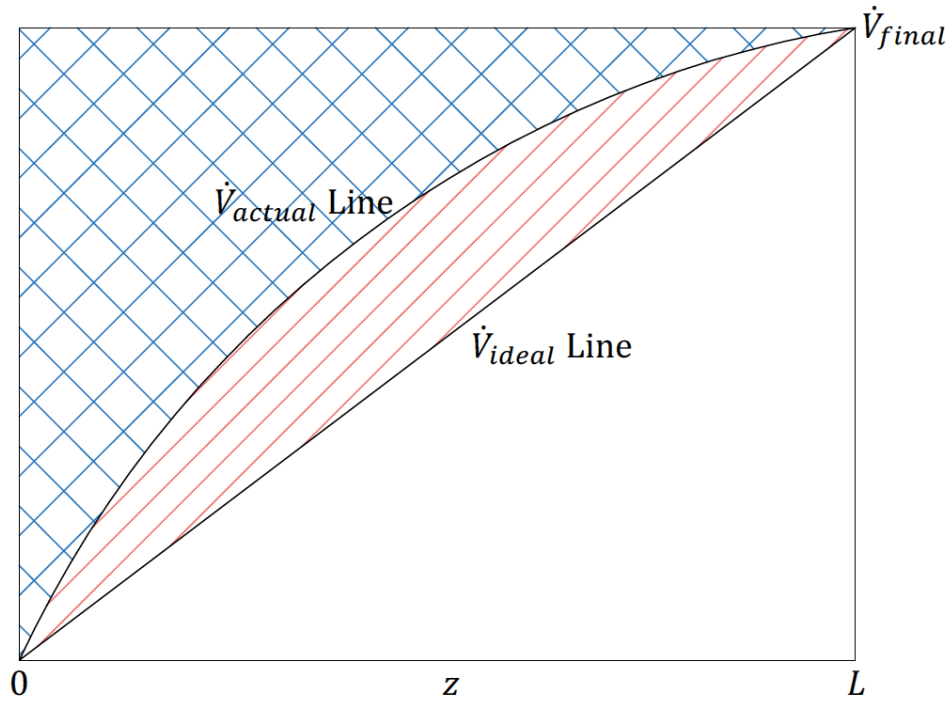
This study found that an optimal module spacing parameter  $S_p$  exists that maximizes the module productivity in both the 2D and 3D filtration systems (in particular we find  $S_p^{opt} \approx 1$  in 2D and  $S_p^{opt} \approx 1.24$  in 3D, see Figure 4.5).



**Figure 4.5:** The normalized module productivity,  $\alpha$ , versus different values of dimensionless module spacing parameters **(a)**  $\ell$  in a 2D channel (as shown in Section 3.4) and **(b)**  $\gamma$  in an axisymmetric pipe respectively. In both cases,  $Q = 1$  and  $k = 1$ .

Herein the uniformity of the filtration behaviour is quantified through the parameter  $\varphi$ . The definition of this uniformity parameter,  $\varphi$ , is explained in accord with Figure 4.6. In Figure 4.6, the actual cumulative fluid volume line separates the triangular area into blue hatched area and red hatched area. Therefore

$$\begin{aligned}
\varphi &= \frac{\text{blue area}}{\text{triangular area}} = 1 - \frac{\int_0^L \Delta V \, dz}{\frac{1}{2} L V_{final}} \\
&= 1 - \frac{2}{L V_{final}} \int_0^L \Delta V \, dz.
\end{aligned} \tag{4.28}$$



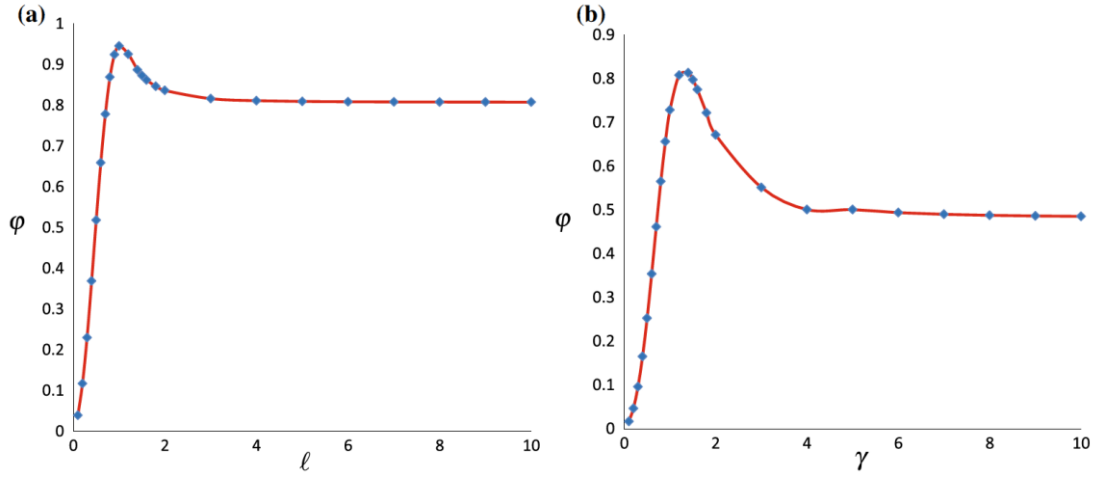
**Figure 4.6:** Illustration of the filtration uniformity parameter definition according to the cumulative fluid volume line (The  $\dot{V}_{actual}$  line can be below the diagonal and also be ‘S’ shaped, as shown later in Figure 4.8).

Applying the non-dimensionalized parameters in this chapter, the filtration uniformity parameter,  $\varphi$ , is defined for the study in this chapter:

$$\varphi = 1 - \frac{2}{V_{final}} \int_0^1 \Delta V(z) dz, \quad (4.29)$$

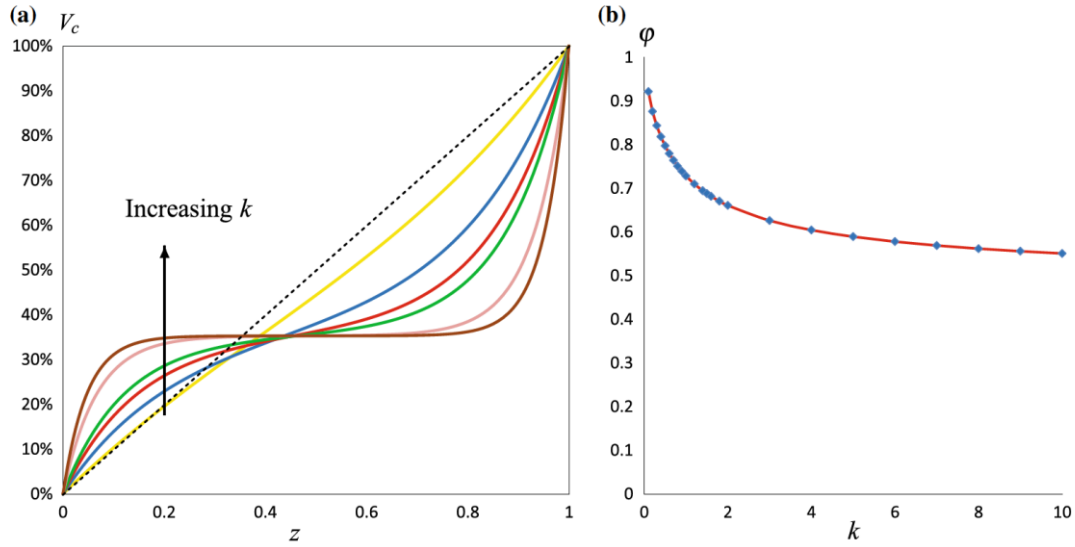
where  $\Delta V(z) = |V_{ideal} - V_c(z)|$  with  $V_{final}$  the total filtration volume,  $V_{ideal} = z V_{final}$  the cumulative volume that would be filtered along the membrane under strictly uniform (idealized) filtration, and  $V_c$  is the actual cumulative volume that has been filtered (as seen in Figure 4.4). When  $\varphi = 1$  the system filters the fluid uniformly across the entire fibre medium; when  $\varphi = 0$  all the fluid is filtered only at a single position in the fibre.

At low module spacing parameters, the uniformity of the filtration behaviour is low and increases as  $S_p$  increases (Figure 4.7). As for the module productivity,  $\alpha$ , the study finds that the filtration uniformity,  $\varphi$ , is optimized for module spacing parameter values of around 1 both in the 2D channel and axisymmetric pipe systems. In a 2D channel, the results find that  $\varphi > 0.8$  for all  $\ell \gtrsim 0.7$  and the peak occurs at  $\ell \approx 1$  where  $\varphi \approx 0.95$ . This is consistent with Figure 4.4a, which indicates the most uniform filtration performance when  $\ell$  is near 1. For a pipe,  $\varphi > 0.7$  for  $0.95 \lesssim \gamma \lesssim 1.9$  and the peak occurs at  $\gamma \approx 1.3$  where  $\varphi \approx 0.82$ . The values of the spacing parameter that create 95% uniformity in a 2D channel and 82% uniformity in pipe are also close to those that maximize the module productivity factor,  $\alpha$ . If one continuously increases  $S_p$  beyond unity, the uniformity of the filtration behaviour will begin to fall again, asymptoting to the value attained for an individual pipe surrounded by an infinite bath of fluid.



**Figure 4.7:** The measure of the uniformity of the overall filtration,  $\varphi$ , versus different values of module spacing parameters **(a)**  $\ell$  in a 2D channel (as shown in Section 3.4) and **(b)**  $\gamma$  in an axisymmetric pipe respectively. In both cases, the filtration is setup at  $Q = 1$  and  $k = 1$ .

The uniformity in filtration is also influenced by the membrane permeability,  $k$ , with smaller permeabilities giving rise to more uniform filtration (Figure 4.8). However, one should avoid too low a permeability in a real engineering situation as this will result in either low throughputs or the need for a high inlet pressure. When the wall permeability is large, the central portion of the filter will contribute little to the overall filtration, with the membrane surface near the beginning and the end providing the main contribution, as seen in Figure 4.8a. For example, when  $k = 10$ , around 30% of the total flux passes through the membrane in the portion  $0 \leq z \leq 0.2$ ; filtration then decreases with a small TMP, before the majority of the remaining fluid is filtered in the region  $z \gtrsim 0.8$ .



**Figure 4.8:** (a) The cumulative fluid volume transported through the fibre walls (%) versus axial distance,  $z$ , in an axisymmetric pipe with different values of permeability  $k$ ; (b) The measure of the uniformity of the overall filtration,  $\phi$ , in a pipe versus permeability  $k$ . In (a), the varying quantity assumes the values 0.1, 0.5, 1, 1.5, 5, and 10, respectively (the black dashed line depicts the optimal filtration case). In both cases, the filtration is setup at  $Q = 1$  and  $\gamma = 1$ .

As reported in literature (Herterich, 2015), the inlet pressure (i.e. minimum pump outlet pressure) is directly proportional to the flux,  $Q$ , and decreases with increasing permeability,  $k$ . As the module spacing parameter decreases, more of the fluid passes through a smaller area of the membrane, which also results in a higher operating pressure. Therefore, optimal direct-flow modules with a capped-end should operate at low fluxes, be manufactured from a material with a relatively low permeability, and be arranged such that the ratio of the inside fibre to the outside is approximately unity. Such modules would ensure a relatively uniform filtration across the entire available membrane fibre area in the system.

## 4.4 Conclusion

This chapter derived a mathematical model for the fluid flow in an axisymmetric direct-flow pipe model composed of a series of hollow fibres arranged within a single device, each with a capped-end. The main focus was to investigate the influence of the proximity of neighbouring fibres on the flow behaviour and the filtration efficiency. This generalizes the models developed for a single hollow fibre with a capped-end (Hurwitz, 1989), a 2D direct-flow Channel with a capped-end (Herterich, 2015), and models where neighbouring fibres are adjacent to one another (Sanaei et al., 2016).

The model is able to predict the operating regimes that optimize the uniformity of fluid transport across the whole membrane area, thus maximizing the efficiency of operation. This chapter defined a module spacing parameter based on the of cross-sectional areas of the feed and permeate side. It showed that, to maximize the uniformity of use of the filtration area, capped-end filter modules should operate at low fluxes, be composed of a filter material with a relatively low permeability and have neighbouring fibres spaced such that the ratio of volumes on the feed and permeate side are approximately equal (i.e. module spacing parameter values of approximately unity).

For small module spacing parameters, this chapter showed that a large portion of the available membrane space is not utilized, and the majority of the filtration occurs at or near the impermeable capped-end. Module spacing parameters of around 1 will result in a close to optimal distribution of available membrane area in the modules, when operating in a regime that optimizes the pressure applied and the space occupied by the system. The lower the permeability the higher the uniformity in the filtration system. However, this needs to be weighed against the

fact that as the permeability falls a higher pump pressure is required to drive the fluid across the filter.

This chapter has proposed two important metrics and a strategy for optimizing them, but for engineering design it is also useful to have real units considered. Also for industry, other factors may play a role, principally cost. Other functions could be constructed with respect to other optima which may be determined in a similar manner. The results of this chapter are useful for the following chapters and offers a deeper understanding of the filtration process in simple way. However, one should also be aware that what is uniform or reasonably uniform for this system may not be uniform for when the flow is reversed (backflow) in the same system. For evaluation of the overall filtration cycle (direct filtration followed by short sharp backflow), further research will be carried out in Chapter 5 and 6 to investigate the backflow in the same modules.

# **Chapter 5**

## **Direct-Flow Hollow Fibre Filtration**

### **Modules**

This chapter focuses on the operational considerations and design aspects of realistic direct-flow filtration membrane modules (with finite fibre wall thickness), specifically for the inside feed format. It investigates the axial dependence of the TMP and the cumulative filtration volume under non-fouling conditions. Geometrical parameters including fibre radius, fibre wall thickness, fibre length, module void fraction and imposed average flux are varied over a broad range values encompassing the values applied in today's water industry. The performance parameters (productivity and uniformity) defined in Chapter 4 are re-defined here for realistic modules. Suggested design ranges for fibre diameter, fibre length, fibre wall thickness and packing density will be determined. Taguchi's method is applied to determine how much each parameter contributes to the overall filtration performance.

## 5.1 Introduction

In Chapter 4, simple direct-flow filtration in 3D axisymmetric pipe model was studied and the strategies for optimizing the direct-flow filtration were proposed. The dimensionless model was used to explore the variation in the axial dependency of TMP and predicted the operating regimes of the device that maximize the spatial uniformity in TMP and thereby optimize the use of the entire membrane area. However, membrane wall thickness was not taken into account and the contents only covered a limited set of module geometric parameters. Nevertheless, it did highlight that for PDI operation a large portion of the available membrane area is not used when the fibres are packed too closely together; in this case the majority of the filtration becomes localized near the impermeable capped-end. Chapter 4 introduced two performance parameters: the uniformity of filtration was quantified through the parameter,  $\varphi$ , and the module productivity was analysed mathematically through the parameter,  $\alpha$ . The former is retained for use herein this chapter whilst the latter is more usefully re-defined as normalized module productivity,  $\beta$ , as given later in this chapter.

Now in hollow fibre membrane modules the wall thickness occupies a notable portion of the overall device space (circa 30%) so in real engineering it influences the module performance. By building upon the mathematical model presented in Chapter 4, but avoiding the thin wall assumption, this chapter explores direct-flow filtration for a range of realistic hollow fibre membrane module dimensions and operating conditions for pressure driven inside flows.

## 5.2 Filtration parameters

In this chapter, the direct-flow filtration model will be analysed under typical industrial operating conditions to quantify the overall filtration performance by explicitly examining various geometric parameters, including inside fibre radius,  $a$ , wall thickness,  $\Gamma$ , void fraction,  $\epsilon$ , and fibre length,  $L$ , at various imposed average fluxes,  $J_{av}$ . In contrast to Chapter 4, this approach examines the TMP profiles established by the fluid mechanics for a range of realistic industrially relevant values rather than use dimensionless quantities.

The two performance parameters are defined and used as measures of merit. The normalized module productivity,  $\beta$ , is defined as:

$$\beta = \frac{J_{av}}{k_m P_p} (1 - \epsilon), \quad (5.1)$$

where  $k_m$  is the permeance through the membrane walls, which is used as 300 L m h/bar (L m h represents litres per m<sup>2</sup> per h) in this thesis (Pearce, 2011),  $P_p$  is the pump driving (inlet) pressure (with respect to the ambient pressure as reference) observed during the filtration process,  $J_{av}$  is the imposed average flux, and  $\epsilon$  is the void fraction.  $P_p$  also indicates the difference in pressure between feed and filtrate and thus quantifies the driving force through the membrane.

$\frac{J_{av}}{k_m P_p}$  is a measure of the actual average flux to that which would occur if all of the filtration were at the inlet pressure (assuming permeate side was uniformly at ambient pressure), and  $1 - \epsilon$  is a measure of the area available for filtration to the maximum theoretically available (see supplementary contents in Appendix B). The filtration uniformity factor,  $\varphi$ , used herein (from Chapter 4 Figure 4.6), is:

$$\varphi = 1 - \frac{2}{L \dot{V}_L} \int_0^L \Delta \dot{V}(z) dz, \quad (5.2)$$

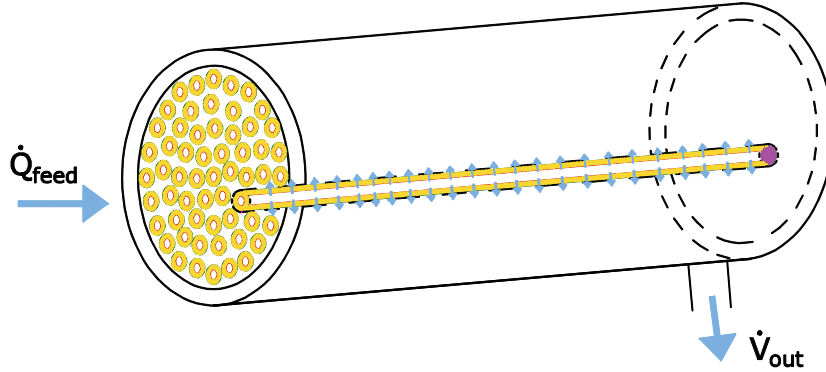
where  $\dot{V}_L = \int_0^L 2\pi a J(z) dz$  is defined as the overall cumulative flowrate exiting at the capped-end (  $\dot{V}_L = \dot{Q} = 2\pi a L J_{av}$  ), and  $\Delta \dot{V}(z) = |\dot{V}_{ideal}(z) - \dot{V}_c(z)|$  with  $\dot{V}_c(z) = \int_0^z 2\pi a J(z) dz$  which is the actual cumulative flowrate of permeate at axial position  $z$  and  $\dot{V}_{ideal}(z) = \frac{z \dot{V}_L}{L}$  is the cumulative flowrate of permeate at axial position  $z$  that would be filtered through the membrane under strictly uniform (idealised) filtration conditions.

Additionally, the advanced Taguchi method will be introduced to explicitly indicate the relative contribution of each geometrical parameter on the filtration performance parameters,  $\beta$  and  $\varphi$ .

## 5.3 Model development

### 5.3.1 Setup

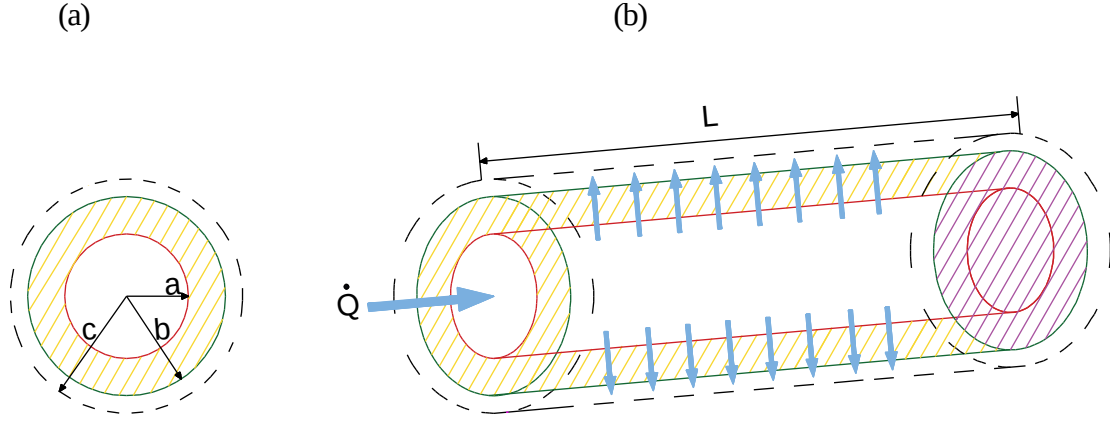
A hollow fibre membrane system includes bundles of small porous hollow fibres packed into a shell module (shown in Figure 5.1), which is similar in configuration to a shell and tube heat exchanger. The key difference in direct-flow filtration is that there is no outlet on the tube side and no inlet on the shell side in the pressure driven inside format. The opposite is true for the pressure driven outside format.



**Figure 5.1:** Schematic of capped-ended hollow fibre membrane module. Please note aspect ratio (length/fibre diameter) is not to scale.

The single fibre as highlighted in Figure 5.1 is more closely examined in Figure 5.2. The fibre has an internal radius  $a$ , an outer radius  $b$  and a length  $L$ . For simplicity, a symmetry condition is applied so that a radial coordinate system  $(r, z)$  can be used. In this axisymmetric coordinates,  $r$  denotes the radial coordinate and  $z$  represents the distance along the fibre length  $L$ . The yellow hashed annulus area represents the permeable wall of the fibre, with the inside fibre wall shown as the red circle and the outer fibre wall shown as the green circle. According to (Happel, 1959) and (Elmore and Lipscomb, 1995), each single fibre can be envisioned as being bounded by an imaginary cylinder with zero-stress outer surface centred around the fibre. This assumption is sometimes known as the equivalent annulus approximation. This imaginary boundary is indicated with a dotted circle of radius  $c$  in Figure 5.2a. A constant permeate flow rate  $\dot{Q} = 2\pi a L J_{av}$  is applied to the model fibre based on the inside filtration area. For such an isolated fibre this study consider the region inside the fibre as lumen side, the region outside as shell side. As shown in Figure 5.2b, there is inside feed. The pressure in the lumen side is  $p$ , and in the shell

side is  $q$ . As discussed in previous chapters, the quantity of most interest is the TMP,  $p - q$ . The TMP profile directly indicates how the filtration process proceeds varies along the fibre length.



**Figure 5.2:** Schematic of a single fibre as used in model development (Section 5.3) **(a)** cross sectional view; **(b)** overall side view with flow direction indicated. The aspect ratio (length/fibre diameter) is not to scale.

Industrial modules typically have a length of around 1.5m, the inside radius of the HFM is around 0.4mm, therefore the aspect ratio (radius/length) of the module  $\delta = a/L \ll 1$ . The fluid velocity in  $r$  direction is  $u$ , and velocity along the fibre length direction is  $w$ . As explained in Chapter 3, the Reynolds number of current model is reconsidered as  $Re = (2apw)/\mu$ , where  $\rho$  is the density of the fluid (taken to be water),  $w$  is the typical axial velocity where the average is used in Reynolds number calculation, and  $\mu$  is the kinematic viscosity. With imposed average flux of 70 L m h, the Reynolds number calculated with the above values of length and radius is 131, the reduced Reynolds number  $\delta Re = O(10^{-2}) \ll 1$ . Therefore, lubrication equations can be applied.

### 5.3.2 Lubrication equations

For the isolated fibre model (shown in Figure 5.1), this study considers the region inside the fibre as lumen side, the region outside as shell side. For lumen side (for  $0 \leq r \leq a$  and  $0 \leq z \leq L$ ), the lubrication equations can be written as

$$\frac{1}{r} \frac{\partial}{\partial r} (ru) + \frac{\partial w}{\partial z} = 0, \quad (5.3)$$

$$\mu \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial w}{\partial r} \right) = \frac{dp}{dz}. \quad (5.4)$$

The boundary conditions for lumen side can be defined as

$$\text{Symmetry:} \quad \frac{\partial w}{\partial r} = 0; u = 0, \quad \text{when } r = 0, \quad (5.5)$$

$$\text{No axial wall slip:} \quad w = 0, \quad \text{when } r = a, \quad (5.6)$$

$$\text{Permeate flow:} \quad u = \frac{k(p - q)}{\mu a \ln \frac{b}{a}}, \quad \text{when } r = a, \quad (5.7)$$

$$\text{Fluid inflow:} \quad 2\pi \int_0^a wr \, dr = \dot{Q}, \quad \text{when } z = 0, \quad (5.8)$$

$$\text{No fluid outflow:} \quad w = 0, \quad \text{when } z = L. \quad (5.9)$$

Shell side region (for  $b \leq r \leq c$  and  $0 \leq z \leq L$ ) fluid also obeys the lubrication equations

$$\frac{1}{r} \frac{\partial}{\partial r} (ru) + \frac{\partial w}{\partial z} = 0, \quad (5.10)$$

$$\mu \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial w}{\partial r} \right) = \frac{dq}{dz}. \quad (5.11)$$

The boundary conditions for shell side can be defined as

$$\text{Zero stress surface:} \quad \frac{\partial w}{\partial r} = 0; u = 0, \quad \text{when } r = c, \quad (5.12)$$

$$\text{No axial wall slip:} \quad w = 0, \quad \text{when } r = b, \quad (5.13)$$

$$\text{Permeate flow:} \quad u = \frac{k(p - q)}{\mu b \ln \frac{b}{a}}, \quad \text{when } r = b, \quad (5.14)$$

$$\text{No fluid inflow:} \quad w = 0, \quad \text{when } z = 0, \quad (5.15)$$

$$\text{Fixed outlet pressure:} \quad q = q_L = 1 \text{ atm} \quad \text{when } z = L. \quad (5.16)$$

The equations can be solved analytically to get the transverse velocity expressions. For lumen side the results find,

$$w = \frac{1}{4\mu} \frac{dp}{dz} (r^2 - a^2), \quad (5.17)$$

$$u = \frac{1}{16\mu} \frac{d^2 p}{dz^2} (2a^2 r - r^3), \quad (5.18)$$

while on shell side,

$$w = \frac{1}{4\mu} \frac{dq}{dz} \left( r^2 - 2c^2 \ln \frac{r}{b} - b^2 \right), \quad (5.19)$$

$$u = \frac{1}{16\mu} \frac{d^2 q}{dz^2} \left\{ 2b^2 r - r^3 + c^2 \left[ -2r + 4r \ln \frac{r}{b} - \frac{2b^2}{r} + c^2 \left( \frac{3}{r} - \frac{4}{r} \ln \frac{c}{b} \right) \right] \right\}. \quad (5.20)$$

Substituting the boundary equations into the velocity expressions, a system of ODEs can be found:

$$\frac{d^2 p}{dz^2} = \frac{16k(p - q)}{\Lambda_1}, \quad \frac{dp}{dz} \Big|_{z=0} = -\frac{8\mu\dot{Q}}{\pi a^4}, \quad \frac{dp}{dz} \Big|_{z=L} = 0, \quad (5.21)$$

$$\frac{d^2 q}{dz^2} = -\frac{16k(p - q)}{\Lambda_2}, \quad \frac{dq}{dz} \Big|_{z=0} = 0, \quad q|_{z=L} = q_L = 1\text{atm}, \quad (5.22)$$

where

$$\Lambda_1 = a^4 \ln \frac{b}{a}, \quad (5.23)$$

$$\Lambda_2 = -b^4 \ln \frac{b}{a} + c^2 \ln \frac{b}{a} \left[ 4b^2 - c^2 \left( 3 - 4 \ln \frac{c}{b} \right) \right], \quad (5.24)$$

$$\Lambda_3 = -\frac{8\mu\dot{Q}}{\pi a^4}. \quad (5.25)$$

The ODEs can be solved analytically to obtain expressions for the pressures:

$$p(z) = \frac{1}{\Lambda_1 + \Lambda_2} [\Lambda_2 \mathcal{N}(z) - \Lambda_1 \Lambda_3 (L - z) + \mathcal{N}_0] + q_L, \quad (5.26)$$

$$q(z) = \frac{1}{\Lambda_1 + \Lambda_2} [-\Lambda_1 \mathcal{N}(z) - \Lambda_1 \Lambda_3 (L - z) + \mathcal{N}_0] + q_L, \quad (5.27)$$

where

$$\mathcal{N}(z) = \text{TMP} = -\frac{\Lambda_3}{C_1} \text{Csch}(C_1 L) \left[ \frac{\Lambda_1}{\Lambda_2} \text{Cosh}(C_1 z) + \text{Cosh}[C_1(L - z)] \right], \quad (5.28)$$

$$\mathcal{N}_0 = -\frac{\Lambda_1 \Lambda_3}{C_1} \left[ \frac{\Lambda_1}{\Lambda_2} \text{Coth}(C_1 L) + \text{Csch}(C_1 L) \right], \quad (5.29)$$

$$C_1 = \sqrt{16k \left( \frac{1}{\Lambda_1} + \frac{1}{\Lambda_2} \right)}. \quad (5.30)$$

From Darcy's law, the relationship between the membrane permeance  $k_m$ , the membrane permeability  $k$  (based on inner fibre area) and the membrane resistance  $R_m$ , are defined respectively ( $J$  denotes for the local flux across the membrane, the calculation in this chapter used the imposed average flux  $J_{av}$ )

$$k_m = \frac{J}{\text{TMP}}, \quad (5.31)$$

$$J = \frac{\text{TMP}}{\mu R_m}, \quad (5.32)$$

$$k = \frac{a}{R_m} \ln\left(\frac{b}{a}\right). \quad (5.33)$$

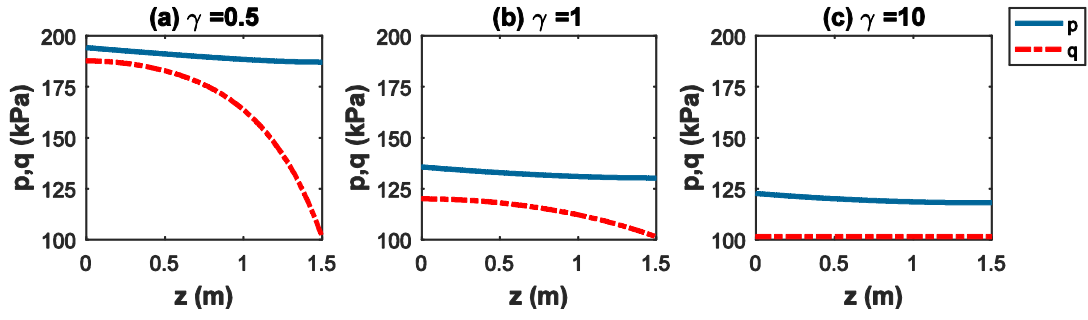
## 5.4 Results and discussion

Before displaying results indicating the influence of void fraction, fibre diameter and other parameters, an overview of the pressure profiles is given using the parameter  $\gamma$  which parallel that used in Chapter 4 where:

$$\gamma = \frac{\text{area in the shell side}}{\text{area in the lumen side}} = \frac{\pi c^2 - \pi b^2}{\pi a^2} = \frac{c^2 - b^2}{a^2}. \quad (5.34)$$

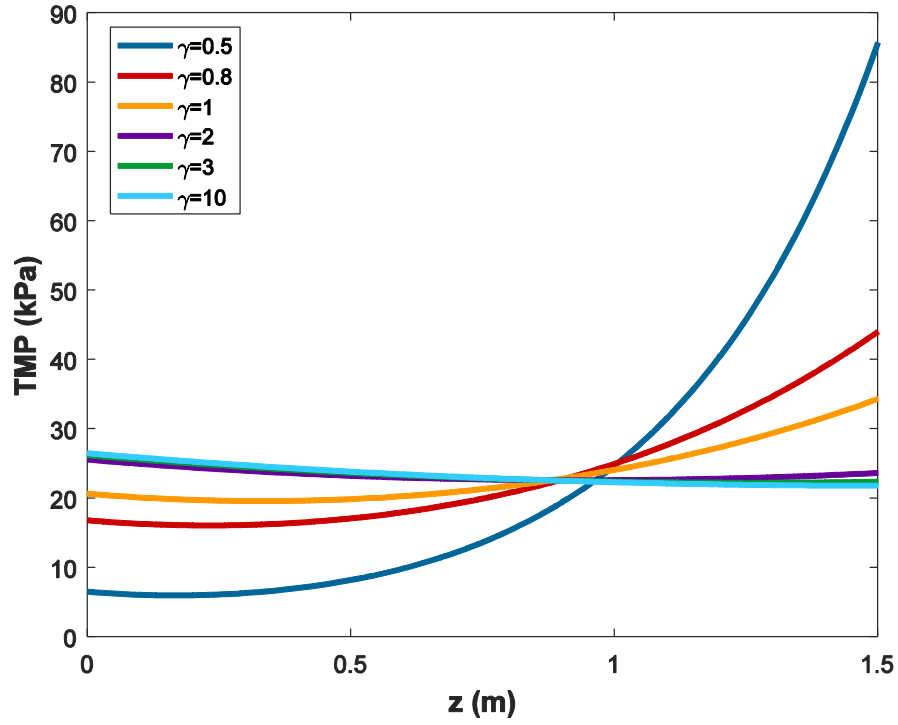
### 5.4.1 Influence of fibre spacing on axial pressure profiles

Figure 5.3 shows three patterns of pressure profiles. When the spacing on the shell side is large as in Figure 5.3c the shell side behaves as a quiescent bath that is essentially at the exit pressure over the whole length. With equal areas on the two sides (Figure 5.3b) the difference of  $p - q$  (i.e. TMP) varies modestly along the fibre length. Too smaller a value of  $\gamma$  (Figure 5.3a) introduces a significant hydrodynamic resistance on the shell side and this results in a major maldistribution of TMP and hence of fluxes.



**Figure 5.3:** Variation of fluid pressure in the lumen side,  $p$ , and in the shell side,  $q$ . Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70 \text{ L m h}$  and  $k_m = 300 \text{ L m h/bar}$  with spacing parameters **(a)**  $\gamma = 0.5$ , **(b)**  $\gamma = 1$  and **(c)**  $\gamma = 10$ .

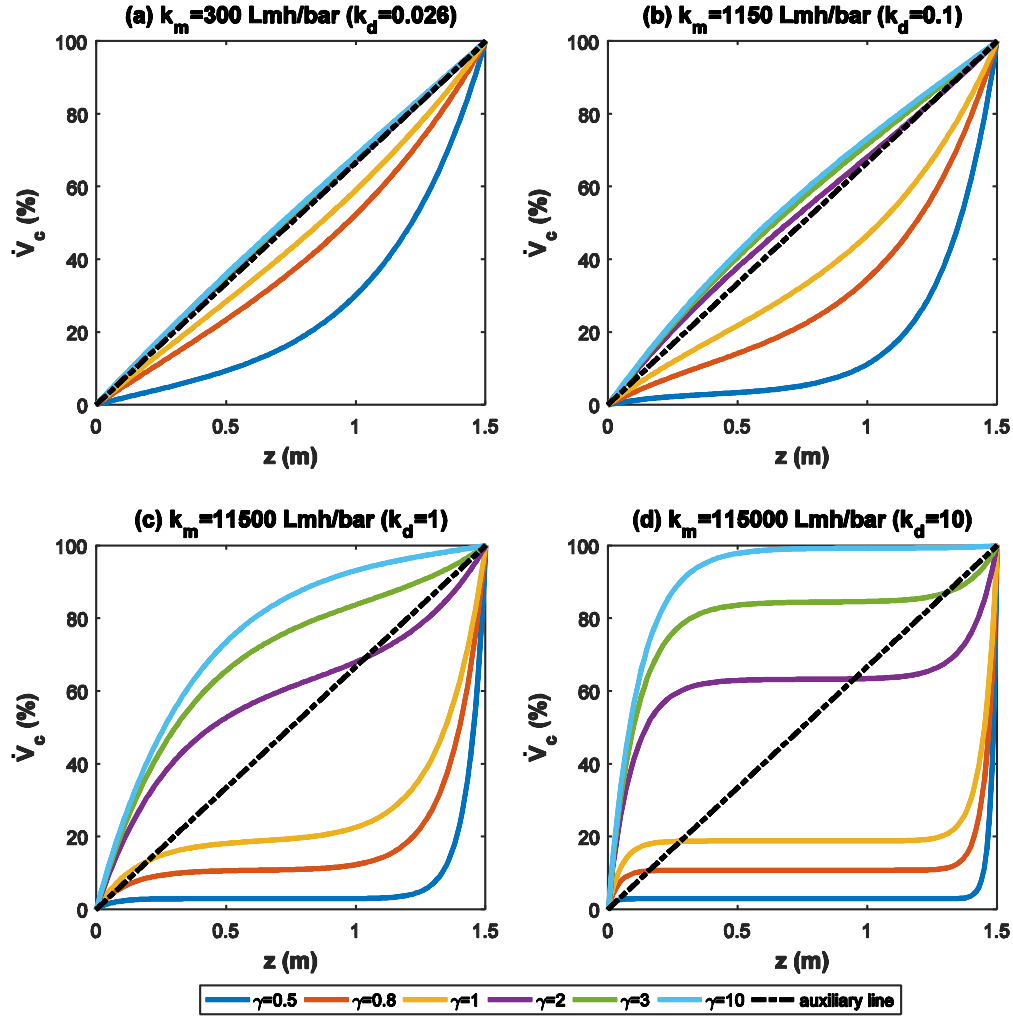
As shown in Figure 5.4, with a small module spacing parameter  $\gamma = 0.5$ , TMP increases along the fibre length and there is a large variation between minimum and maximum values. For  $\gamma$  between 1 and 3, the TMP profiles indicate relative uniform distribution but one should note that the local flux at  $z = 0$  becomes higher than that at the capped-end when  $\gamma > 2$ . However, continuing to increase  $\gamma$  value beyond approximately 3 has only a small influence on the TMP plots and the overall filtration process (green line and light blue line overlap completely as shown in Figure 5.4). This is in agreement with a previous conclusion made in Chapter 4 where there was zero wall thickness.



**Figure 5.4:** The effect of spacing parameter,  $\gamma$ , upon the transmembrane pressure difference (TMP) profiles versus axial distance,  $z$  at different spacing parameter  $\gamma$ . Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70 \text{ L m h}$  and  $k_m = 300 \text{ L m h/bar}$ .

In previous dimensionless model developed in Chapter 4, the equivalent dimensionless variable of permeance,  $k_m$ , was set from 0.1 to 10. For fixed parameters  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$  and  $J_{av} = 70 \text{ L m h}$ , a value of 1 implies a permeance of  $11500 \text{ L m h/bar}$ , while a value of 0.1 equates  $1150 \text{ L m h/bar}$  which is still higher by a factor of 3.83, than  $300 \text{ L m h/bar}$  found in commercial operations. The permeance  $k_m = 300 \text{ L m h/bar}$  used in this thesis is equivalent to the dimensionless variable used in Chapter 4 having a value of 0.026 which is outside of the range considered. Hence, the representative dimensionless value used in previous chapter is not applicable in the real water industry. It was found that lower permeance has the effect of creating better uniformity and this is illustrated in Figure 5.5. The

extreme non-uniformity noted previously can now be unambiguously attributed to the permeance values chosen. For typical industrial values of the parameters (Figure 5.5) a permeance of 300 L m h/bar gives only modest spatial variation if  $\gamma \gg 0.8$ .



**Figure 5.5:** The effect of spacing parameter,  $\gamma$ , upon the cumulative volume transported through the fibre walls,  $\dot{V}_c$  (%), versus axial distance,  $z$ . Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ , and  $J_{av} = 70$  L m h with permeances (a)  $k_m = 300$  L m h/bar, (b)  $k_m = 1150$  L m h/bar, (c)  $k_m = 11500$  L m h/bar, and (d)  $k_m = 115000$  L m h/bar.<sup>3</sup>

<sup>3</sup> Permeance,  $k_m$ , is transferred to equivalent value of dimensionless variable  $k_d$ , which is defined as dimensionless permeability,  $k$ , in Chapter 4.

### 5.4.2 Selection of design parameters

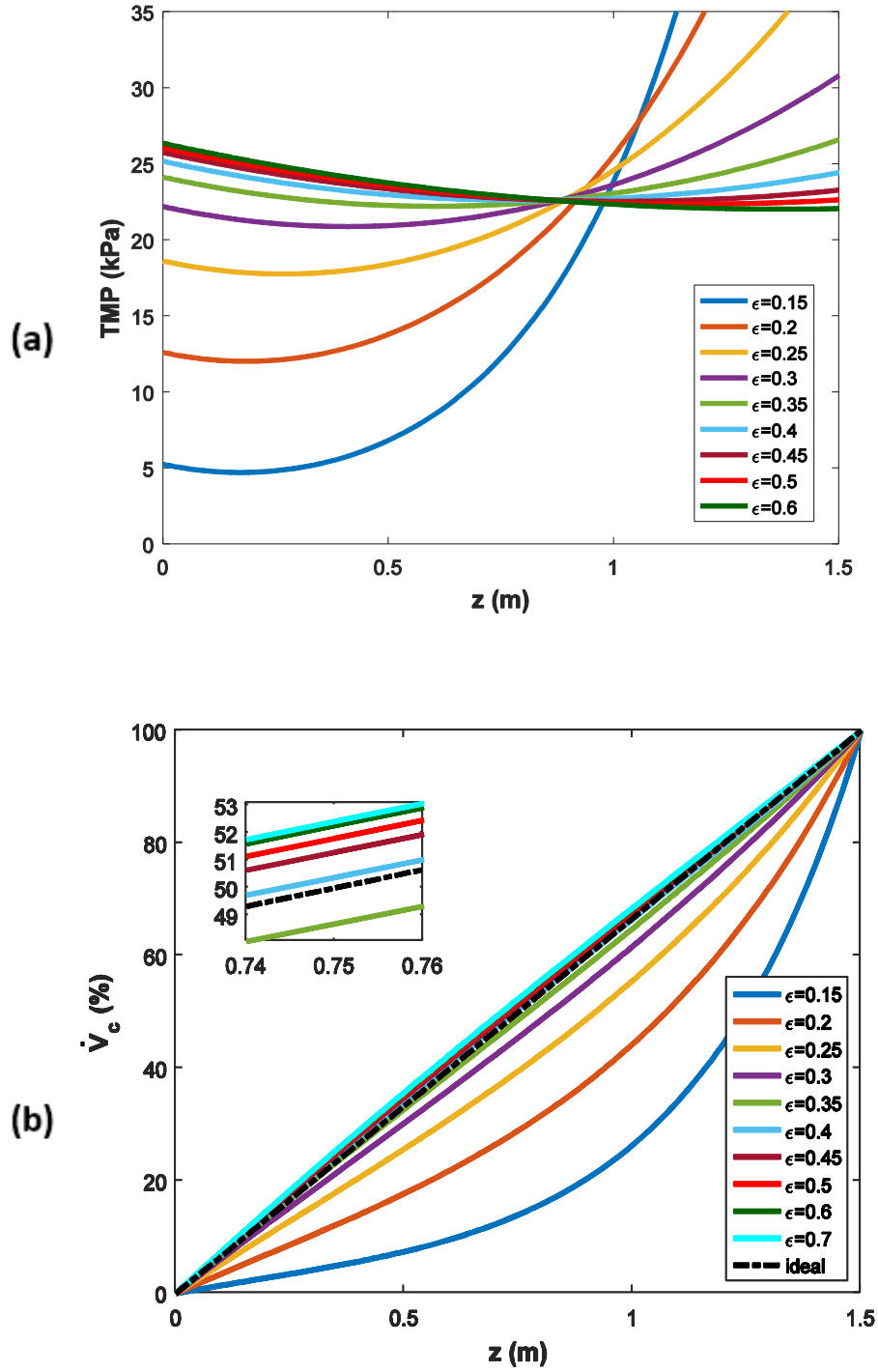
In real membrane manufacturing the module spacing parameter,  $\gamma$ , cannot be adjusted directly, instead, a hollow-fibre module is usually designed with regard to void fraction,  $\epsilon$ , which is defined with respect to the fibre area on the shell side as:

$$\epsilon = \frac{\text{free area in the shell side}}{\text{total area}} = \frac{\pi c^2 - \pi b^2}{\pi c^2} = \frac{c^2 - b^2}{c^2}. \quad (5.35)$$

The lowest achievable void fraction for square array is  $\epsilon = 0.215$  while for equilateral triangular array is  $\epsilon = 0.093$ . In order to alter the overall void fraction,  $\epsilon$ , fibre properties including inside fibre radius,  $a$ , wall thickness,  $\Gamma$ , and fibre length,  $L$ , have therefore to be examined. Typically, the fibre wall thickness is designed as two thirds of the fibre inner radius to maintain the mechanical strength (Pearce, 2011). Although the effect of fibre wall thickness upon the filtration behaviour is studied in this chapter, it will not be emphasised in this thesis. A typical commercial value of  $\Gamma = 0.25\text{mm}$  is used in this chapter. Permeance,  $k_m$ , is not investigated further in this chapter, as permeability,  $k$ , is based on an industrial value for a clean membrane. This chapter seeks explicitly to examine the filtration behaviour based on the effect of geometric factors and fluid inflow. Thus the main variables considered are: module void fraction,  $\epsilon$ , inside fibre radius,  $a$ , fibre length,  $L$ , and imposed average fluxes,  $J_{av}$ .

### 5.4.2.1 Influence of void fraction

In Figure 5.6a, TMP profiles are compared at different void fraction,  $\epsilon$ , with other parameters set at: inner radius  $a = 0.4\text{mm}$ , wall thickness  $r = 0.25\text{mm}$ , fibre length  $L = 1.5\text{m}$ , imposed average flux  $J_{av} = 70\text{ L m h}$  and permeance  $k_m = 300\text{ L m h/bar}$ . These are the values for the standard case. The profiles for  $\epsilon = 0.15$ ,  $0.2$  and  $0.25$  are not fully shown in Figure 5.6a so as not to compress other details. At small  $\epsilon$  values, the fluid hardly goes through the permeate wall near the entrance and thus the majority of the filtration process takes place near the capped-end. When  $\epsilon = 0.25$  (yellow line), a rapid increase of TMP is observed near the capped-end, which results in the local flux at capped-end is over twice that at the entrance. This can be even worse for much smaller  $\epsilon$  values (i.e. dark blue line  $\epsilon = 0.15$ ) as indicated in the cumulative fluid volume plots (Figure 5.6b). At  $\epsilon = 0.15$ , 80% of the fluid passes through the membrane over the last 20% fibre length.

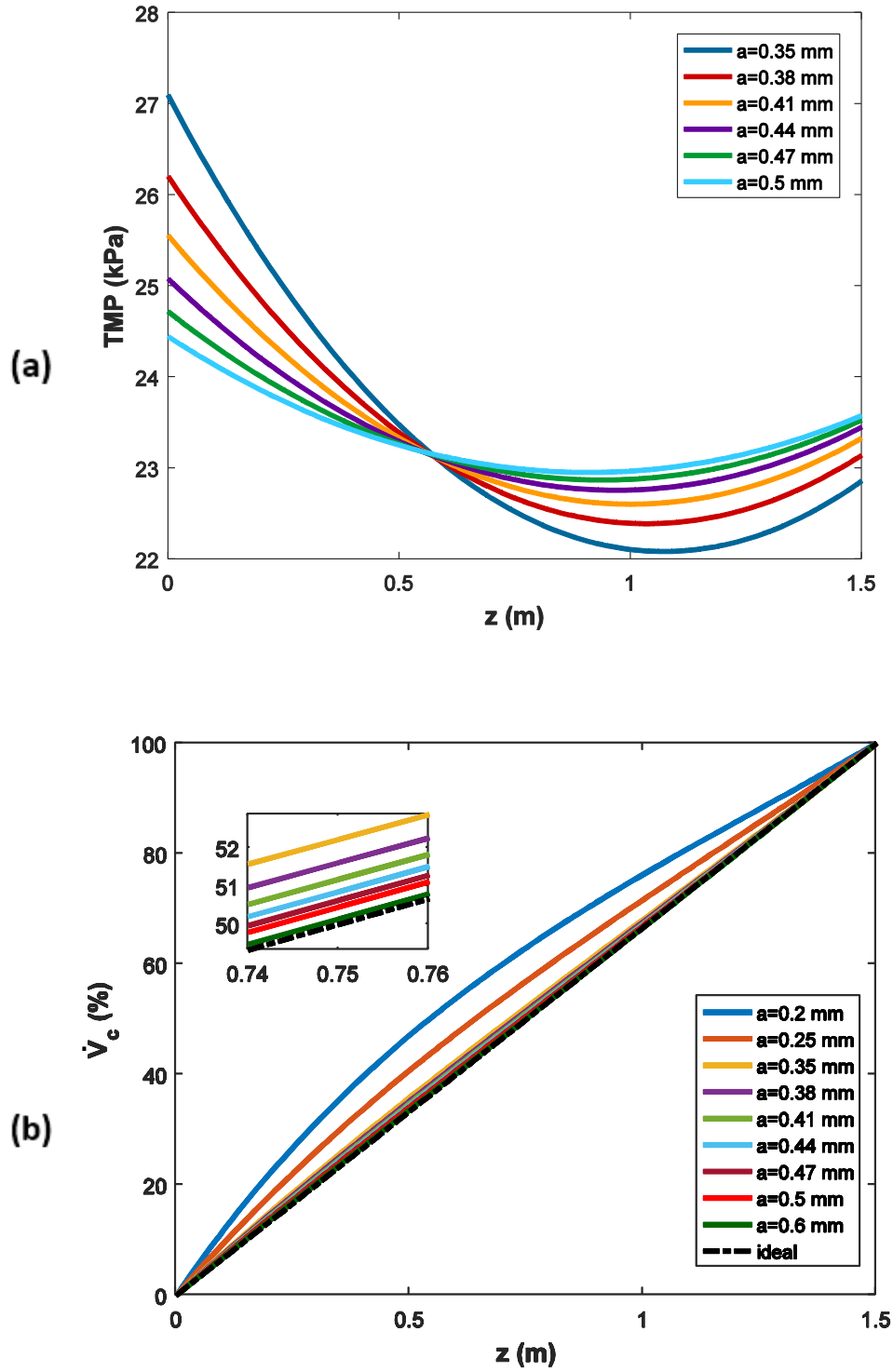


**Figure 5.6:** The effect of void fraction,  $\epsilon$ , upon **(a)** the transmembrane pressure difference (TMP) profiles and **(b)** the cumulative volume transported through the fibre walls,  $\dot{V}_c$  (%), versus axial distance,  $z$ . Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70\text{ L m h}$  and  $k_m = 300\text{ L m h/bar}$ .

In theory, the lowest achievable void fraction for square array is  $\epsilon = 0.215$ , while for equilateral triangular array is  $\epsilon = 0.093$ , however, the practical minimum void fraction in commercial products is  $\epsilon = 0.4$ . In practice the maximum pressure tolerance to avoid permeability loss due to active layer compression or fibre damage is 140 kPa to 300 kPa, and the maximum TMP to ensure stable low fouling operation is 70 kPa (Pearce, 2011). A value of  $\epsilon = 0.15$  is impractical as it would involve a maximum lumen side pressure of circa 210 kPa and a corresponding maximum TMP of 95 kPa. Increasing  $\epsilon$  promotes uniform filtration, but increasing  $\epsilon$  beyond approximately 0.5 will hardly change the cumulative volume curves (Figure 5.6b). As such increase will reduce the filtration area the module productivity will reduce. The resulting trade-off between uniformity and productivity is discussed later in Section 5.4.3.

#### 5.4.2.2 Influence of fibre inner radius

Figure 5.7a shows TMP profiles for different inner radii,  $a$ , with other parameters fixed. All TMP profiles show negative gradient with increasing axial distance and minima around 1m fibre length. The axial variation of TMP is greater for small  $a$ ; the range of  $a$  encompasses the industrial range of 0.4 to 0.45 mm. When  $a = 0.35$  mm, a maximum TMP of 27.1 kPa is found at  $z = 0$  and a minimum TMP of 22.1 kPa is found at  $z = 1.08$ m (A greater variation is found for smaller  $\epsilon$ ). The filtration processes in Figure 5.7b show more non-uniform distribution at small  $a$  values. Increasing  $a$  values can minimize the differences between the maximum and minimum TMP values thus promoting a more uniform filtration. (Kim et al., 2012b) reached a similar conclusion and recommended a fibre of larger inner diameter and small wall thickness as long as it were of sufficient mechanical strength.

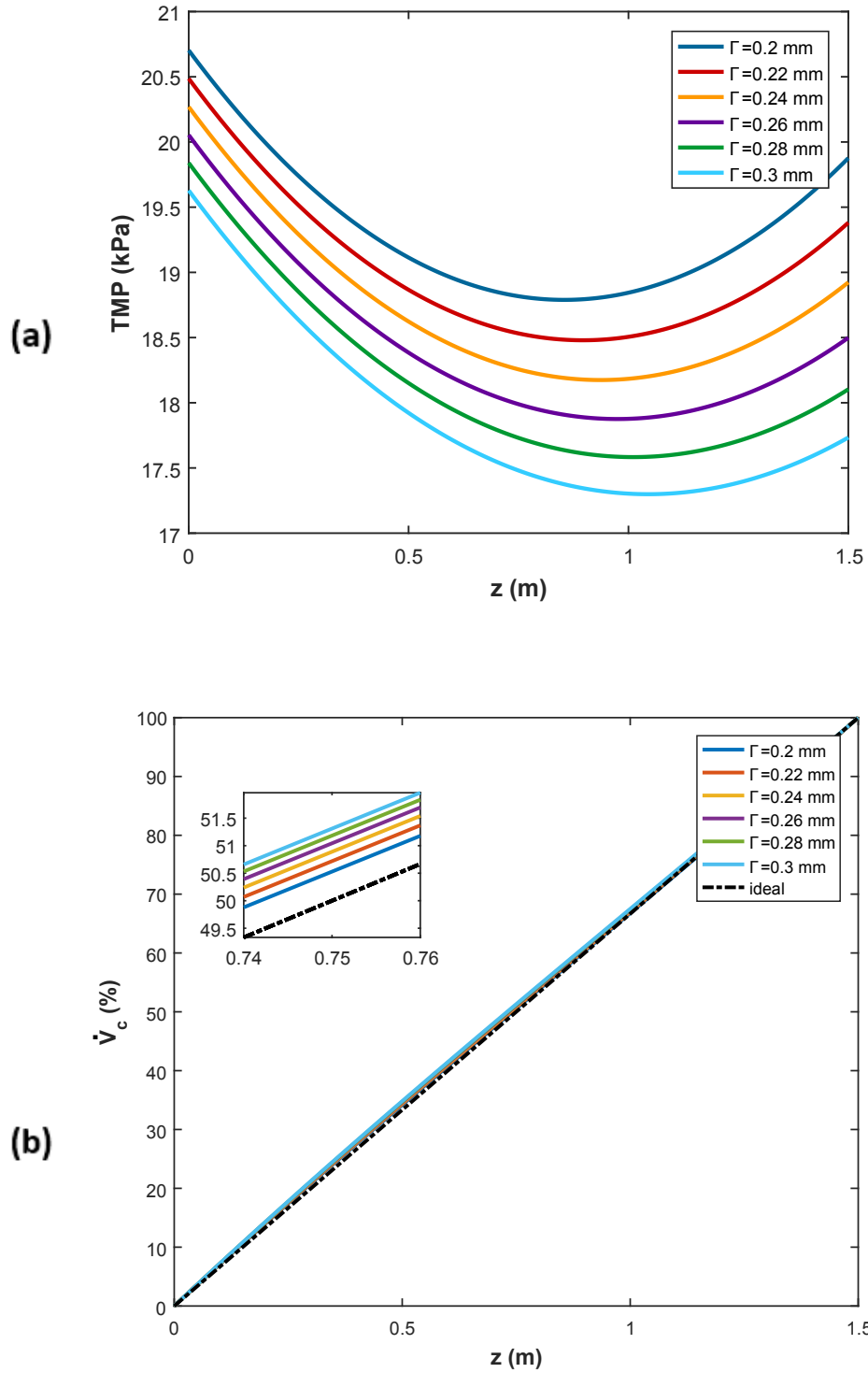


**Figure 5.7:** The effect of inner radius,  $a$ , upon **(a)** the transmembrane pressure difference (TMP) profiles and **(b)** the cumulative volume transported through the fibre walls,  $\dot{V}_c$  (%), versus axial distance,  $z$ . Fixed parameters:  $\epsilon = 0.45$ ,  $\Gamma=0.25$ mm,  $L = 1.5$ m,  $J_{av} = 70$  L m h and  $k_m = 300$  L m h/bar.

### 5.4.2.3 Influence of fibre wall thickness

As shown in Figure 5.8a, the TMP profiles show similarly negative gradient first and then increase gradually near the impermeable capped-end, which results in a saucer shape without any intersection between lines. With increasing  $\Gamma$  values, the profiles retain a qualitatively similar shape but show more TMP variation along the fibre length with thicker walls. Specifically, when  $\Gamma = 0.2$  mm the TMP value at the impermeable capped-end is even higher than the TMP value at  $z = 0$  when  $\Gamma = 0.3$  mm. Therefore, thinner fibre wall effectively minimizes the difference of maximum and minimum TMP values.

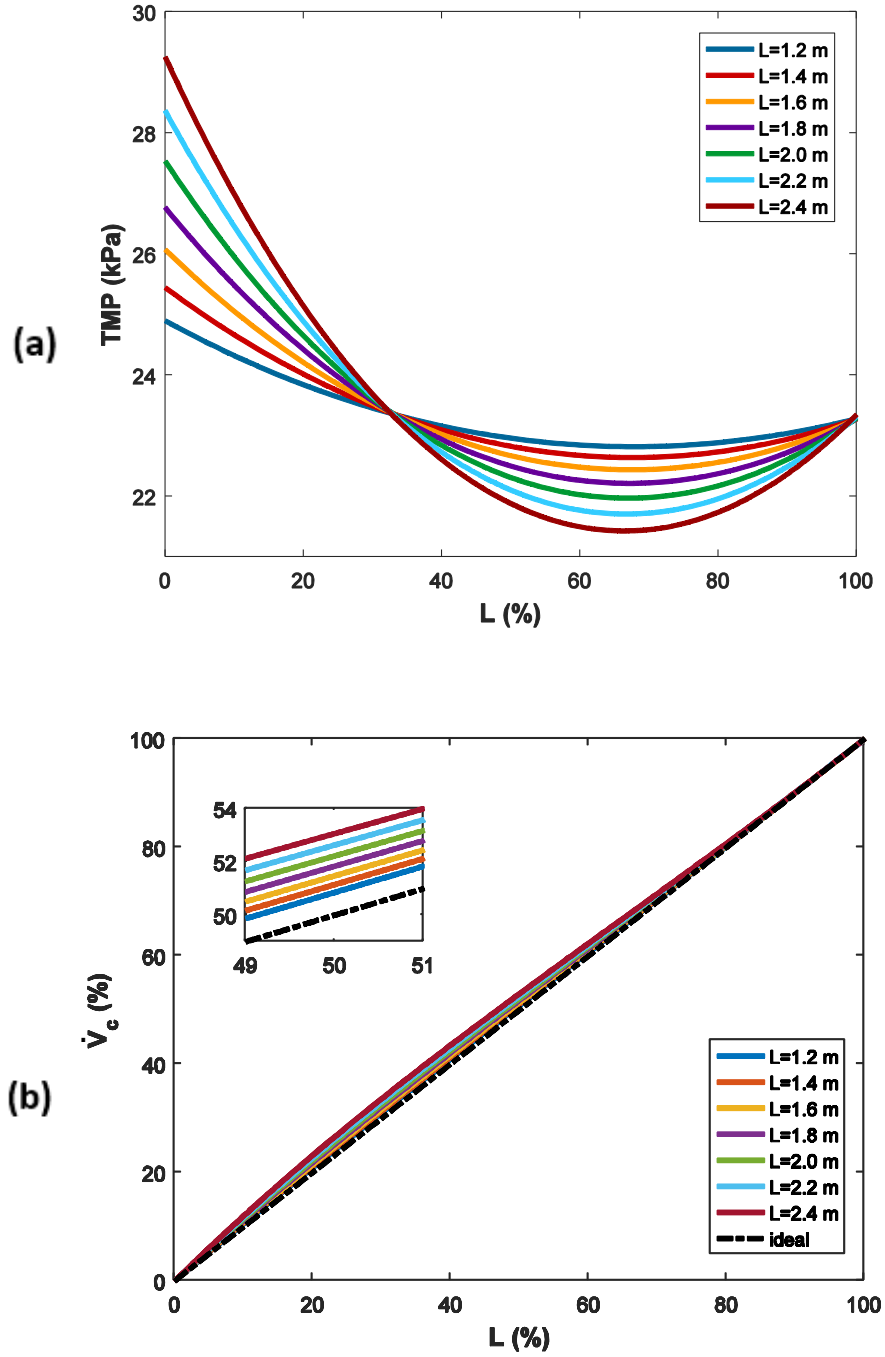
The profiles in both Figure 5.8a and b indicate that a thinner fibre wall promotes uniform distribution of local flux. However, the wall thickness variations show less impact on the overall filtration process than other parameters, i.e. void fraction (Figure 5.6), fibre inner radius (Figure 5.7) and fibre length (later in Section 5.4.2.4). Therefore, the effect of fibre wall thickness will not be emphasised in the rest of this thesis. In commercial products, although a very thin fibre wall can ensure more uniform distribution of the filtration behaviour, it is always avoided because it lowers the mechanical strength (Baley, 2002). In practice, the fibre wall thickness is usually two thirds of the fibre inner radius.



**Figure 5.8:** The effect of wall thickness,  $\Gamma$ , upon (a) the transmembrane pressure difference (TMP) profiles and (b) the cumulative volume transported through the fibre walls,  $\dot{V}_c$  (%), versus axial distance,  $z$ . Fixed parameters:  $a = 0.45\text{mm}$ ,  $\epsilon = 0.45$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70\text{ L m h}$  and  $k_m = 300\text{ L m h/bar}$ .

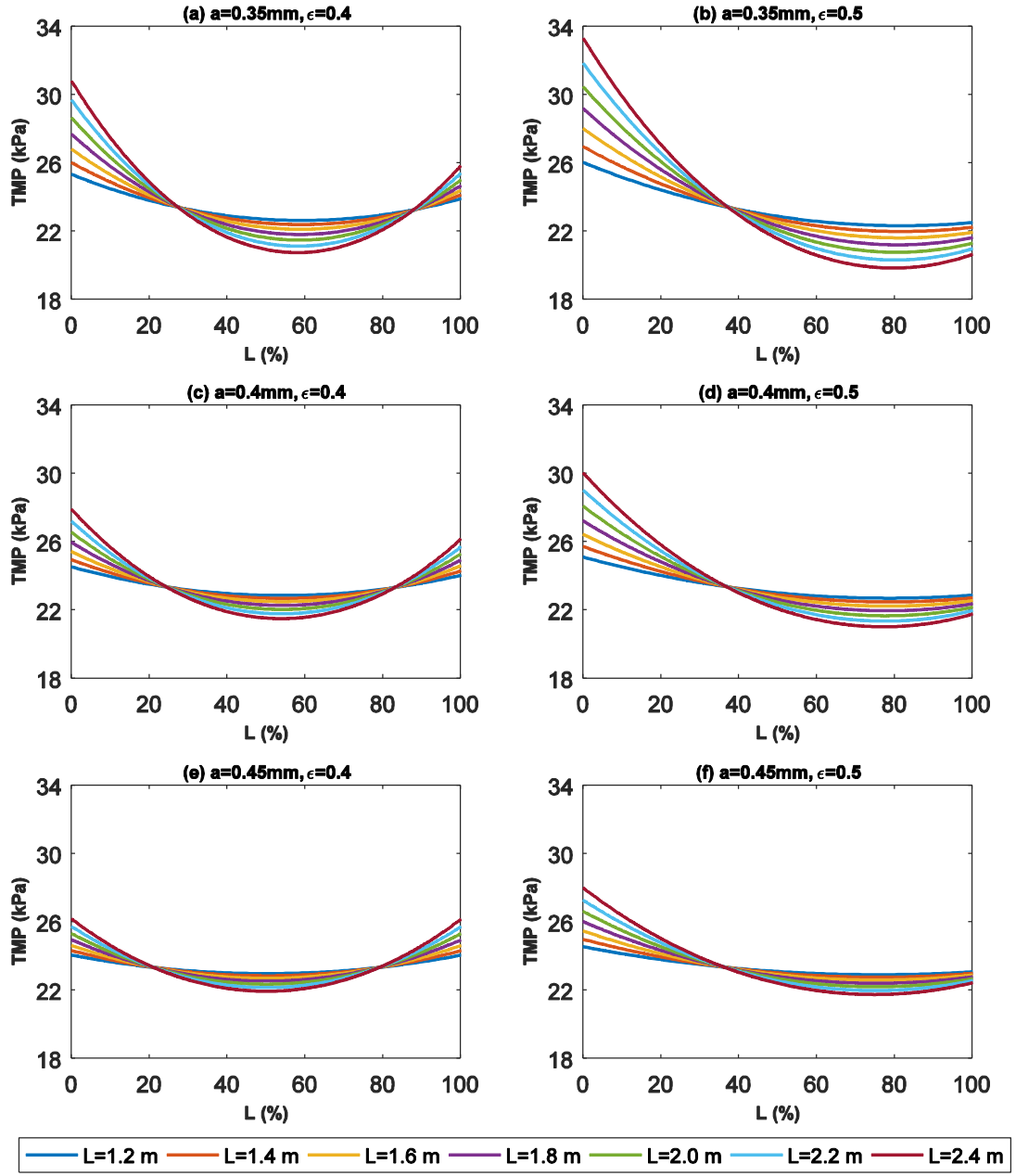
#### 5.4.2.4 Influence of fibre length

Figure 5.9a shows TMP profiles for various fibre length,  $L$ , with other parameters fixed. The concave curves with a minimum around 67% of the length of the fibre show a similar pattern to that given above for the effect of fibre radius on TMP profiles as seen in Figure 5.7, the longer the fibre the greater the TMP variation. The TMP variation of 5.9 kPa at  $L = 2.4\text{m}$  is over triple that of 1.6 kPa for  $L = 1.2\text{m}$ . A shorter fibre is preferred but module productivity will be reduced which is a trade-off discussed more fully later in Section 5.4.3.



**Figure 5.9:** The effect of fibre length,  $L$ , upon (a) the transmembrane pressure difference (TMP) profiles and (b) the cumulative volume transported through the fibre walls,  $\dot{V}_c$  (%), versus percentage of the fibre length at different fibre length,  $L$  (%). Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $\epsilon = 0.45$ ,  $J_{av} = 70 \text{ L m h}$  and  $k_m = 300 \text{ L m h/bar}$ .

The effect of fibre length on TMP profiles at various inner radii and module void fractions are compared in Figure 5.10. The plots reveal that increasing inner radius can minimise the difference between maximum and minimum TMPs. The effect of changes in the module void fraction are small if the fibre length is  $L = 1.2\text{m}$ . For longer fibres increasing  $\epsilon$  increases the TMP at the inlet. Now as the average flux and permeance are fixed the *average* TMP is the same for every case covered in Figure 5.10, so modules with higher TMPs at inlet will also have lower TMPs elsewhere along the module i.e. the variation in TMP increases. Although Figure 5.9a and Figure 5.10 seem to suggest unique crossover points at particular values of % length, it is found on close examination that they are spread over a small range, e.g. from around 26.5% to 28% and 87% to 89% in Figure 5.10a and an even tighter spread of just 0.3% (36.0 to 36.3%) in Figure 5.10f. We seek to explain this observation. Firstly consider two curves one with a high inlet TMP and one with a lower inlet TMP. Clearly the former will form more of a ‘U’ and the latter will be flatter. These curves will have two crossover points. Now add an intermediate curve and consider it with respect to the original two curves. For each pairing there will be two crossover points but there is no particular reason why these would be unique values and upon closer inspection they are not unique values.



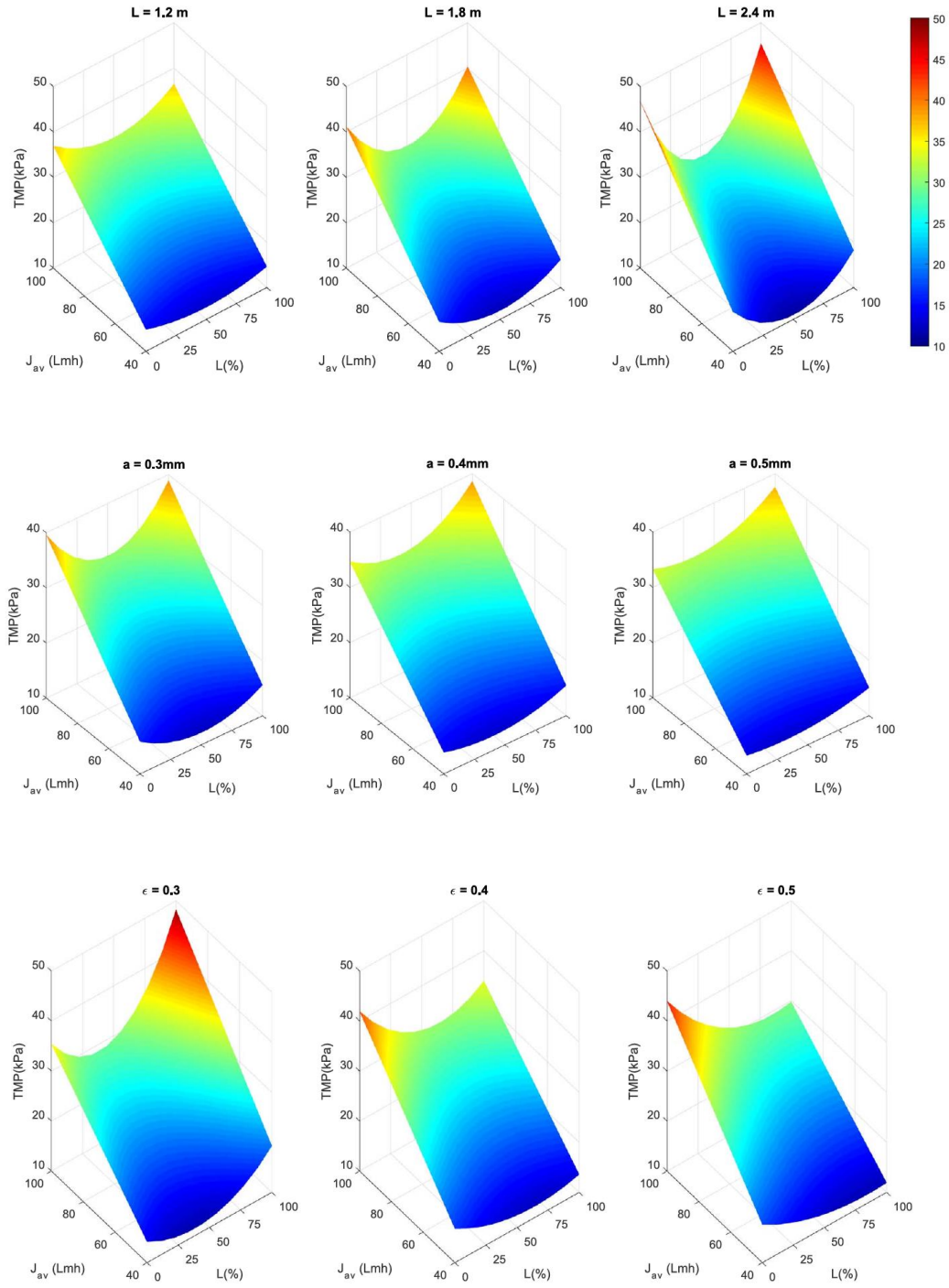
**Figure 5.10:** The effect of fibre length,  $L$ , upon the transmembrane pressure difference (TMP) profiles versus percentage of the fibre length at different fibre length,  $L$  (%). Fixed parameters:  $\Gamma = 0.25\text{mm}$ ,  $J_{av} = 70 \text{ L m h}$  and  $k_m = 300 \text{ L m h/bar}$ . (a)  $a = 0.35\text{mm}$ ,  $\epsilon = 0.4$ , (b)  $a = 0.35\text{mm}$ ,  $\epsilon = 0.5$ , (c)  $a = 0.4\text{mm}$ ,  $\epsilon = 0.4$ , (d)  $a = 0.4\text{mm}$ ,  $\epsilon = 0.5$ , (e)  $a = 0.45\text{mm}$ ,  $\epsilon = 0.4$ , (f)  $a = 0.45\text{mm}$ ,  $\epsilon = 0.5$ .

#### 5.4.2.5 Influence of imposed average flux

In Figure 5.11, surface plots of TMP profiles at different imposed average flux,  $J_{av}$ , (based on the inside fibre area  $2\pi aL$ ) are investigated at various fibre lengths, with other parameters fixed as given in caption. Unlike previous figures the standard parameters in Figure 5.11 are  $a = 0.3$  mm,  $\epsilon = 0.35$ , and  $L = 1.6$  m except when specifically varied. The first three subplots examine the effect of changing  $L$ , the middle set the effect of  $a$ , and the last set the effect of  $\epsilon$ . Negative gradient of the TMP profiles are observed for the first two-thirds of the fibre length. After a minimum value around this point TMPs then slightly increase until the end of the fibre.

For a fibre of given diameter and length, the only way of increasing the output is to increase the imposed average flux. However, very high imposed average fluxes will intensify fouling issues. As (Field and Pearce, 2011, Field et al., 1995) pointed too high a starting flux will lead to over-fouling. Although fouling cannot be avoided in practice, a relatively lower flow can restrain particles build-up on the membrane surface. Furthermore with a highly compressible layer a higher driving force (provided by increasing TMP) can be largely offset by the increased resistance resulting from a compressible layer.

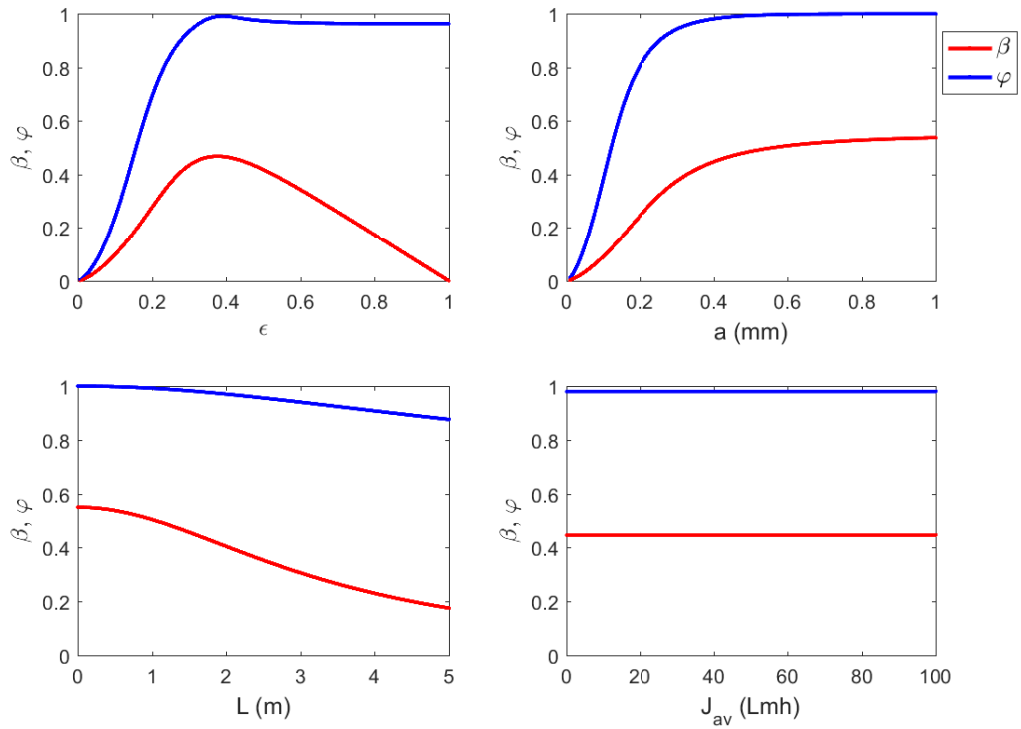
As shown in Figure 5.11 very long fibres lead to significantly more curvature in the TMP profiles. Thus the flux distribution will be uneven and at the capped-end there will be a coupling of a relative high solids loading with a relatively high local flux and this will lead to locally high fouling. Initially for a few cycles this will not be problematic but it is not efficient in the long term. Other plots show that relatively small inner fibre radius and relatively small void fraction also lead to greater curvature especially at higher fluxes.



**Figure 5.11:** The effect of imposed average flux,  $J_{av}$ , upon the transmembrane pressure difference (TMP) profiles versus percentage of the fibre length,  $L$  (%). Except where indicated standard parameters were as follows:  $a = 0.3$  mm,  $\epsilon = 0.35$ ,  $\Gamma = 0.25$  mm,  $L = 1.6$  m and  $k_m = 300$  L m h/bar.

### 5.4.3 Filtration performance

This chapter has so far considered the effects of several design parameters on the TMP profiles and the fluid filtration distribution. However, the overall filtration performance should be quantified both in terms of its productivity and uniformity. The latter is taken to be a proxy for an ability to curtail fouling effects. Thus performance parameters of the normalized module productivity,  $\beta$ , and the filtration uniformity factor,  $\varphi$ , are presented together in Figure 5.12.



**Figure 5.12:** The normalized module productivity,  $\beta$ , and the uniformity of the filtration behaviour,  $\varphi$ , of the filtration process versus the design control parameters. **(a)** void fraction,  $\epsilon$ , **(b)** inner radius,  $a$ , **(c)** fibre length,  $L$ , and **(d)** imposed average flux,  $J_{av}$ , respectively. Standard parameters:  $a = 0.4$  mm,  $\epsilon = 0.45$ ,  $\Gamma = 0.25$  mm,  $L = 1.6$  m,  $J_{av} = 70$  L m h and  $k_m = 300$  L m h/bar.

In Figure 5.12, the normalized module productivity factor,  $\beta$ , is shown plotted against each design parameters. An optimum range around 0.3 – 0.45 of void fraction is suggested by the curves in Figure 5.12a. Further increases reduce the overall module productivity. In Figure 5.12b, below 0.5,  $\beta$  is proportional to the inside fibre radius which validates the suggestion that the larger the inner radius the more uniform the filtration process will be achieved (as shown in Figure 5.7). However further beyond 0.5 the overall filtration productivity will not change. In Figure 5.12d, higher imposed average flux effectively increases the local flux along the fibre length thus more output can be achieved through the filtration process.

In Figure 5.12, the filtration uniformity factor,  $\varphi$ , is plotted against each design parameter. As discussed before, higher  $\varphi$  values indicate more uniform distribution of the local flux, the idealised universal uniform would be  $\varphi = 1$ . In Figure 5.12a and b increasing the void fraction and the inside fibre radius the overall filtration process will lead to more uniformity (as seen also in Figure 5.6b and Figure 5.7b). In Figure 5.12c, the uniformity of the overall filtration process will weaken as the fibre length increases, as seen in Figure 5.9b previously. In Figure 5.12d, the uniformity of the filtration is shown as being independent of the imposed average flux. The adverse effect of high fluxes and thus higher throughput will become apparent when non-uniformity of fibre spacing is taken into account and allowance is made for the pressure losses associated with exit flows. Table 5.1 provides an overall summary and clearly shows that the model in this chapter can be validated against industrial practice that has evolved over the last 20 years. Industrial practice has been detailed elsewhere (Table 5.2 of (Pearce, 2011)) and the commercial ranges given in Table 5.1 are based mainly on this source.

**Table 5.1:** Summary of table of modelling results in this chapter.

| Design parameters                       | Void fraction                  | Inner radius (mm)      | Wall thickness (mm)      | Fibre length (m)      | Imposed average flux (L m h) |
|-----------------------------------------|--------------------------------|------------------------|--------------------------|-----------------------|------------------------------|
| <b>Productivity</b>                     | $0.3 \leq \epsilon \leq 0.45$  | $a \geq 0.35$          | $r \uparrow$             | $L \leq 2$            | No obvious effect            |
| <b>Uniformity</b>                       | $0.35 \leq \epsilon \leq 0.45$ | $a \geq 0.4$           | $0.15 \leq r \leq 0.3$   | $L \leq 1.8$          | No obvious effect            |
| <b>Commercial ranges (Pearce, 2011)</b> | $0.4 \leq \epsilon \leq 0.45$  | $0.4 \leq a \leq 0.45$ | $r \approx \frac{2}{3}a$ | $1.5 \leq L \leq 1.8$ | $60 \leq J_{av} \leq 80$     |
| <b>Concerns</b>                         | Overall module size            | Mechanical strength    | Mechanical strength      | Overall module size   | Membrane fouling             |

For void fraction the modelling results in this chapter suggest a range of 0.35 – 0.45 and industrial practice is 0.4 – 0.45. For inner radius the modelling results suggest that the value should be  $> 0.4$  mm and naturally one would not design for this to be much larger than necessary as there is a desire to have as much surface area per unit volume as practicable. Thus this thesis suggests close to 0.4 mm and industrial practice is 0.4 – 0.45 mm. For fibre length the modelling results suggest that the value should be  $< 1.8$  m and because potting is relatively expensive one would not design for this to be much shorter; industrial practice is 1.5 – 1.8 m.

#### 5.4.4 Taguchi method

To combine the total influence of four control parameters on the overall module productivity and the uniformity distribution of the local flux, Taguchi method (Taguchi and Taguchi, 1987) is applied in this study.

Typical values of the four variables, taken from industrial practice are displayed in Table 5.2 (Pearce, 2011). As there are 4 parameters influencing the overall filtration performance and each parameter corresponds to at least 4 levels, a traditional approach could involve at least 256 ( $4^4$ ) different sets of calculation. Taguchi Method (Taguchi and Taguchi, 1987) is applied so as to simplify the investigation process. A similar Taguchi approach has been applied by (Xu et al., 2013) to replace an original set of 1024 experiments with a set of 16. Before applying this experimental design method to the study in this thesis, each control factor is individually examined to determine the parameters' range. Based upon the typical applied range given in Table 5.2, values shown in Table 5.3 were chosen for the Taguchi design method. The investigation of 4 different levels of the 4 control parameters (Table 5.3) are then readily assigned to 16 runs by the orthogonal array  $L_{16} (4^4)$  as shown in Table 5.4.

**Table 5.2:** Control parameters and their industrially relevant range.

| Parameters                            | Applied level range |
|---------------------------------------|---------------------|
| <b>A Inside radius (mm)</b>           | 0.4 ~ 0.45          |
| <b>B Void fraction</b>                | 0.4 ~ 0.5           |
| <b>C Fibre length (m)</b>             | 1.5 ~ 1.8           |
| <b>D Imposed average flux (L m h)</b> | 60 ~ 80             |

**Table 5.3:** Control parameters and their levels used in this study.

| Parameters                          | Level 1 | Level 2 | Level 3 | Level 4 |
|-------------------------------------|---------|---------|---------|---------|
| <b>A Inside radius (mm)</b>         | 0.35    | 0.4     | 0.45    | 0.5     |
| <b>B Void fraction</b>              | 0.35    | 0.4     | 0.45    | 0.5     |
| <b>C Fibre length (m)</b>           | 1.2     | 1.6     | 2.0     | 2.4     |
| <b>D Imposed average flux (Lmh)</b> | 40      | 60      | 80      | 100     |

**Table 5.4:**  $L_{16}$  ( $4^5$ ) Orthogonal Array of Taguchi research control parameters.

| Row | Run | A | B | C | D | Inside radius (mm) | Void fraction | Fibre length (m) | Imposed average flux (L m h) |
|-----|-----|---|---|---|---|--------------------|---------------|------------------|------------------------------|
| 1   | 1   | 1 | 1 | 1 | 1 | 0.35               | 0.35          | 1.2              | 40                           |
| 2   | 5   | 1 | 2 | 2 | 2 | 0.35               | 0.4           | 1.6              | 60                           |
| 3   | 9   | 1 | 3 | 3 | 3 | 0.35               | 0.45          | 2.0              | 80                           |
| 4   | 13  | 1 | 4 | 4 | 4 | 0.35               | 0.5           | 2.4              | 100                          |
| 5   | 10  | 2 | 1 | 2 | 3 | 0.4                | 0.35          | 1.6              | 80                           |
| 6   | 14  | 2 | 2 | 1 | 4 | 0.4                | 0.4           | 1.2              | 100                          |
| 7   | 2   | 2 | 3 | 4 | 1 | 0.4                | 0.45          | 2.4              | 40                           |
| 8   | 6   | 2 | 4 | 3 | 2 | 0.4                | 0.5           | 2                | 60                           |
| 9   | 15  | 3 | 1 | 3 | 4 | 0.45               | 0.35          | 2                | 100                          |
| 10  | 11  | 3 | 2 | 4 | 3 | 0.45               | 0.4           | 2.4              | 80                           |
| 11  | 7   | 3 | 3 | 1 | 2 | 0.45               | 0.45          | 1.2              | 60                           |
| 12  | 3   | 3 | 4 | 2 | 1 | 0.45               | 0.5           | 1.6              | 40                           |
| 13  | 8   | 4 | 1 | 4 | 2 | 0.5                | 0.35          | 2.4              | 60                           |
| 14  | 4   | 4 | 2 | 3 | 1 | 0.5                | 0.4           | 2                | 40                           |
| 15  | 16  | 4 | 3 | 2 | 4 | 0.5                | 0.45          | 1.6              | 100                          |
| 16  | 12  | 4 | 4 | 1 | 3 | 0.5                | 0.5           | 1.2              | 80                           |

The multiple linear regression equations of  $\beta$  and  $\varphi$  developed from the Taguchi method (Taguchi and Taguchi, 1987) are as follows:

$$\beta = 0.52287a - 0.46973\epsilon - 0.09169L - 0.00010J_{av} + 0.59117, \quad (5.36)$$

$$\varphi = 0.21615a - 0.12675\epsilon - 0.01548L - 0.00003J_{av} + 0.96884, \quad (5.37)$$

where  $a$ ,  $\epsilon$ ,  $L$ , and  $J_{av}$  are the inside fibre radius, module void fraction, fibre length and the imposed average flux respectively. The ranges are as follows fibre inner radius (0.35 – 0.5 mm), void fraction (0.35 – 0.5), fibre length (1.2 – 2.4 m) and imposed average flux (40 – 100 L m h).

In Eq. (5.36), the normalised module productivity coefficient is positive for fibre radius,  $a$  but negative for void fraction,  $\epsilon$  and the fibre length,  $L$ . It is essentially zero for the imposed average flux,  $J_{av}$ . The pattern for the filtration uniformity (Eq. (5.37)) is similar. Given the units used for  $a$ ,  $\epsilon$ , and  $L$  and their typical values, the influence of  $a$ ,  $\epsilon$ , and  $L$  upon  $\beta$  are approximately the same (see Table 5.5). However with regard to uniformity the influence is in the order: fibre radius > void fraction > fibre length.

**Table 5.5:** Control parameters and their linear regression of  $\beta$  and  $\varphi$  developed from Taguchi method.

| Parameter | $a$                           | $\epsilon$                    | $L$                         | $J_{av}$                     |
|-----------|-------------------------------|-------------------------------|-----------------------------|------------------------------|
| $\beta$   | $0.52287 \times 0.425 = 0.22$ | $0.46973 \times 0.425 = 0.20$ | $0.09169 \times 1.8 = 0.17$ | $0.0001 \times 80 = 0.008$   |
| $\varphi$ | $0.21615 \times 0.425 = 0.09$ | $0.12675 \times 0.425 = 0.05$ | $0.01548 \times 1.8 = 0.03$ | $0.00003 \times 80 = 0.0024$ |

## 5.5 Conclusion

In this chapter, a realistic PDI hollow fibre model with finite fibre wall thickness has been developed. The model helps one to understand the influence of the geometric parameters upon the hydrodynamics and the associated filtration process. The results suggest that a hollow fibre module made with moderate void fraction ( $0.35 - 0.45$ ), a reasonable inner radius ( $0.4 - 0.45\text{mm}$ ) and fibre lengths between  $1.2\text{m}$  and  $1.8\text{m}$  will ensure a balance trade-off between module productivity and overall filtration uniformity. These values accord with industrial practice that has evolved over the last 20 years and this validates the model developed in this chapter. In practice the fibre wall thickness is usually designed together with the fibre inner radius with the former being around  $2/3$  of the latter. Table 5.1 summarised the results.

## **Chapter 6**

# **Time Evolution of Fouling Development in Direct-Flow Filtration**

This chapter explores inside feed direct-flow filtration performance under fouling conditions. Practical models are developed in this chapter so that flux distribution and particle deposition profiles are obtained. Geometrical parameters and operating conditions studied in Chapter 4 and 5 will be re-evaluated under fouling conditions, especially the imposed average flux effect while incorporating periodic backwash cleaning processes. Additionally, the effect of backwash cleaning frequency is investigated.

## 6.1 Introduction

In Chapter 5, the influence of the geometric parameters (i.e. fibre radius, fibre wall thickness, fibre length, void fraction) upon the hydrodynamics and the associated filtration performances were studied and the results were summarised in Table 5.1. In practice, problems have been reported under high imposed average flux conditions (Pearce, 2011). However, the effects of imposed average flux,  $J_{av}$ , on the filtration performances are not clear from the study presented in Chapter 5. Remaining questions included:

- Why higher imposed average flux causes problem in reality?
- Why commercial operations go for 60 – 80 L m h?

This chapter therefore sets up to explore the time evolution of fouling with due allowance for fouling and backwash cleaning processes so that overall one can ensure long term operational stability of these plants. This chapter aims to further identify the operational triggers of fouling problems in direct-flow filtration.

## 6.2 Model development

### 6.2.1 Setup

To investigate the filtration performance with fouling and backwash cleaning process, practical models for direct-flow hollow fibre filtration were developed. Schematics of a single hollow fibre in PDI filtration mode is given in Figure 6.1. The solid yellow annulus area represents the

permeable hollow fibre membrane wall, the dotted black line indicates the imaginary zero-stress boundary of each fibre, and the solid black area is the capped-end in direct-flow filtration PDI mode. As with the previous model (Figure 5.2) each fibre has an internal radius,  $a$ , an outer radius,  $b$ , and a length  $L$ . The zero-stress boundary is centred around the fibre with a radius  $c$ .

**Figure 6.1:** 3D Schematic flow regimes (n sections) of direct-flow filtration in a single hollow fibre membrane in PDI filtration mode.

Generally, the lumen side flow of each single hollow fibre is given by the standard equations used for pipe flow. For the shell side, the equations for the flow in the annulus were adapted. For a Newtonian fluid, velocity at any radial position on the shell side is a function of velocity, the geometry and the pressure gradient. Apply the same radial coordinate system ( $r, z$ ) to each fibre as presented in Chapter 5, integrating the general equation:

$$\tau = \frac{r}{2} \frac{dp}{dz} + \frac{C}{r}, \quad (6.1)$$

with the same boundary conditions applied in model developed in Chapter 5, the constant  $C$  can be solved and the velocity expression in the lumen side is obtained as,

$$u = \frac{1}{4\mu} \frac{dp}{dz} (r^2 - a^2), \quad (6.2)$$

and the velocity in the shell side is obtained as,

$$u = \frac{1}{\mu} \frac{dp}{dz} \left( \frac{b^2 - r^2}{4} + \frac{c^2}{2} \ln \frac{c}{b} \right). \quad (6.3)$$

Integrating the velocity profile in both side, the general flow rate of direct-flow filtration can be obtained as

$$\dot{Q} = \frac{\pi}{8\mu} \frac{dp}{dz} a^4, \quad (6.4)$$

$$\dot{V} = \frac{\pi}{8\mu} \frac{dq}{dz} \left( -3c^4 + 4c^4 \ln \frac{c}{b} + 4b^2 c^2 - b^4 \right). \quad (6.5)$$

It is noted that Eq. (6.5) is inaccurate at very high packing densities (i.e. very low void fraction) and consideration is given later to other expressions linking flowrate and shell-side pressure drop.

The effect of void fractions (packing densities) upon performance will also be considered. Apply Eqs. (6.4) and (6.5) to a single section  $n$ , the following equations can be followed:

$$\frac{\dot{Q}_n + \dot{Q}_{n+1}}{2} = -\frac{\pi}{8\mu} \frac{p_{n+1} - p_n}{L_s} a^4, \quad (6.6)$$

$$\frac{\dot{V}_n + \dot{V}_{n+1}}{2} = \frac{\pi}{8\mu} \frac{q_{n+1} - q_n}{L_s} \left( 3c^4 - 4c^4 \ln \frac{c}{b} - 4b^2 c^2 + b^4 \right). \quad (6.7)$$

Together with general filtration equations in PDI mode

$$\dot{Q}_n - \dot{Q}_{n+1} = 2\pi a L_s J_n = \dot{V}_{n+1} - \dot{V}_n, \quad (6.8)$$

$$J_n = \frac{\frac{p_n + p_{n+1}}{2} - \frac{q_n + q_{n+1}}{2}}{\mu R_m}, \quad (6.9)$$

the practical model can be successfully built. The outlet of the shell side is taken to be at atmospheric, therefore  $q_{N+1} = 101325$  kPa. Based on the initial conditions  $\dot{Q}_1 = \dot{V}_{N+1}$ ,  $\dot{V}_1 = 0$ ,  $\dot{Q}_{N+1} = 0$  and  $q_{N+1} = 101325$  kPa the model can be solved.

Firstly this practical model was verified by reproducing the results in Chapter 5 for zero fouling so that the model can be subsequently applied with fouling development. Although the results are not shown in this thesis, the practical model indeed recreates the profiles shown from Figure 5.3 to Figure 5.12, and thus reproduces the results summarised in Table 5.1.

## 6.2.2 Cake resistance of fouling development

As discussed in Section 2.4.3, in the presence of fouling, membrane resistance during filtration process increases due to particle accumulation on the membrane surface and/or in the membrane pores (see Eq. (2.6)). Theoretically, the solid phases (i.e. suspended solids) existing in the liquid phase would have an impact on the hydrodynamic profiles of the fluid, however, the effects are negligible due to the extremely low fraction of suspended solids. According to Carman-Kozeny equation as used in literature (Hamachi and Mietton-Peuchot, 1999, Marselina et al., 2008), membrane resistance can be correlated as a function of cake resistance,  $R_c$ , and the deposited particle mass,  $M_c$ , whereas porosity is a function of the Kozeny constant, Sauter diameter, fouling height and cake resistance:

$$\frac{h_c}{R_c} = \frac{1}{36h_k} \frac{d_g^2 \Phi^3}{(1 - \Phi)^2}, \quad (6.10)$$

$$\frac{R_c A_m}{M_c} = \frac{R_c 2\pi aL}{\pi h_c L (h_c + 2a)(1 - \Phi)\rho_c}, \quad (6.11)$$

where  $h_c$  is the height of fouling,  $R_c$  is the resistance caused by fouling,  $h_k$  is the Kozeny constant (5 used in this thesis),  $d_g$  is the Sauter diameter which is the mean value of particle size (1.63  $\mu\text{m}$  used in this thesis),  $\Phi$  is the porosity of the cake accumulated (0.49 used in this thesis),  $M_c$  is the mass of particle deposited,  $A_m$  is the surface area of membrane used ( $2\pi aL$  in PDI format), and  $\rho_c$  is the cake density (2966  $\text{kg/m}^3$  used in this thesis). Concentration of particles in the feed fluid used in this thesis is 200mg/L in order to parallel the results in existing literature (Marselina et al., 2008). At the start of this investigation, both the crossflow and the flux

contribute 50% of the particle deposition effect, and later on the particle deposition effects resulted from the crossflow and the flux will be varied. As discussed, the backwash cleaning frequency varies from 1 – 4 times per hour, therefore the difference of backwash cleaning frequencies will also be evaluated. Backwash cleaning cycle time,  $t_c$ , will therefore be varied at 15 mins, 30 mins, 45 mins, and 60 mins.

### 6.3 Filtration parameters

In this chapter, the direct-flow filtration fouling model will be analysed under various particle deposition and cleaning rules. In contrast to Chapter 5, this approach examines the filtration profiles established by the fluid mechanics in the presence of fouling effects.

The two performance parameters are re-defined and used as measures of merit. The relevant normalized module productivity,  $\beta$ , is therefore updated. Noting that  $k_m = 1/\mu R$ , Eq. (5.1) is rewritten as:

$$\beta = \frac{\mu R J_{av}}{P_p} (1 - \epsilon), \quad (6.12)$$

where  $R$  is the overall membrane resistance including the clean membrane resistance  $R_m$  and the resistance caused by fouling accumulation  $R_f$  (i.e.  $R = R_m + R_f$ ). The term  $P_p$  is the pump driving pressure (with respect to the ambient pressure as reference) observed during the filtration process. It also indicates the difference in pressure between feed and filtrate and thus quantifies the driving force through the membrane.

The filtration uniformity factor,  $\varphi$ , used in Chapter 4 and 5 (see Figure 4.6), is also modified:

$$\begin{aligned}
\varphi &= 1 - \frac{2}{L \dot{V}_L} \int_0^L \Delta \dot{V}(z) dz \\
&= 1 - \frac{2}{L \dot{V}_L} \sum_{n=1}^N \frac{1}{2} L_s \left( \left| \frac{\dot{V}_L}{L} (n-1) L_s - \dot{V}_n \right| + \left| \frac{\dot{V}_L}{L} n L_s - \dot{V}_{n+1} \right| \right) \quad (6.13) \\
&= 1 - \frac{1}{N \dot{V}_L} \sum_{n=1}^N \left( \left| \frac{n-1}{N} \dot{V}_L - \dot{V}_n \right| + \left| \frac{n}{N} \dot{V}_L - \dot{V}_{n+1} \right| \right),
\end{aligned}$$

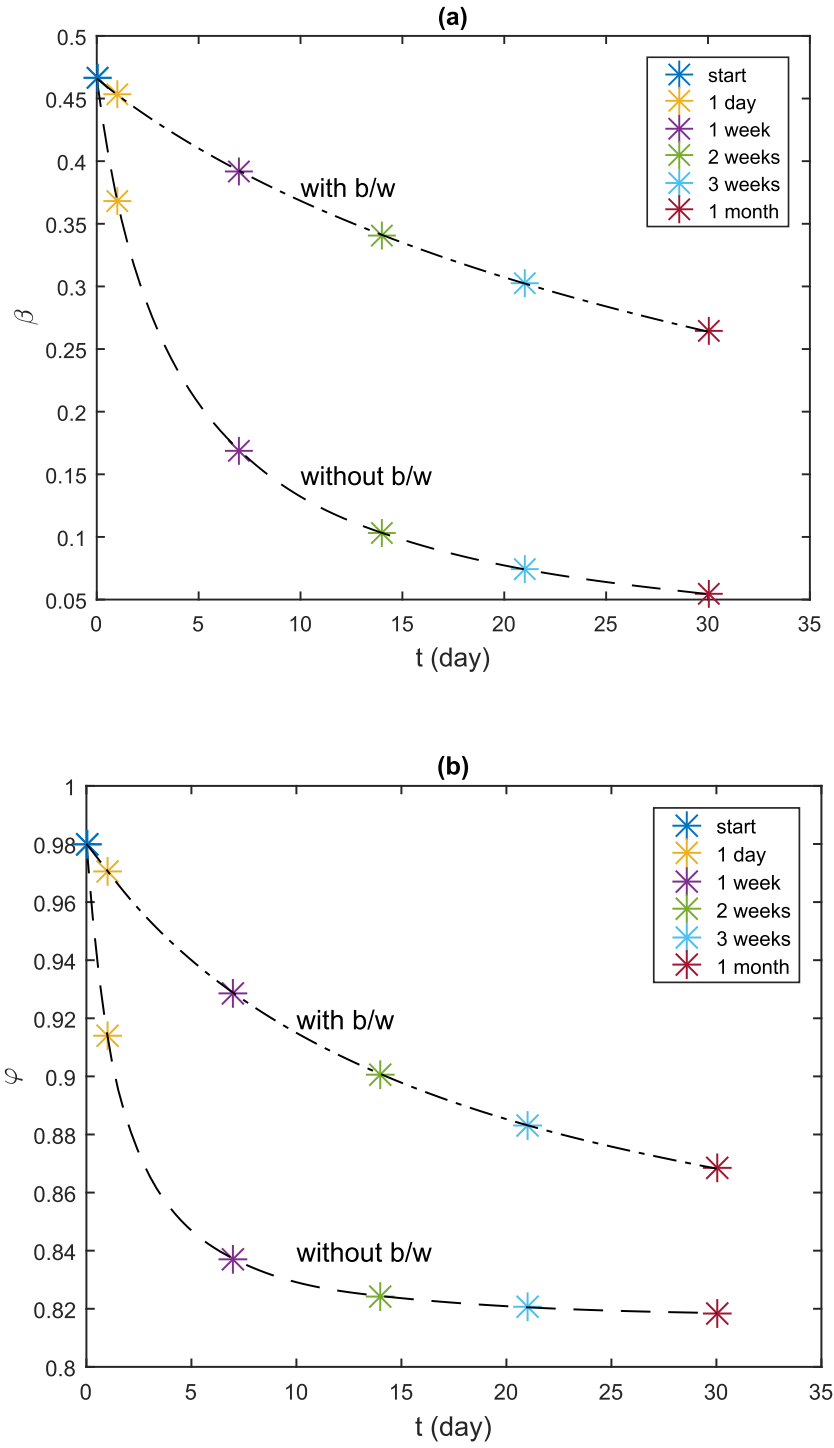
where  $L_s$  is the length of the sub-sections shown in Figure 6.1,  $N$  is the number of sections,  $\dot{V}_n$  and  $\dot{V}_{n+1}$  represent the cumulative flowrate at entrance and exit of specific section  $n$ ,  $\dot{V}_L$  is the overall cumulative flowrate exiting at the capped-end (i.e.  $\dot{V}_L = \dot{V}_{N+1}$ ). The term  $\frac{\dot{V}_L}{L} n L_s$  indicates the cumulative flowrate of permeate at the entrance of section  $n$  that would be filtered through the membrane under the idealised condition of uniform filtration.

## 6.4 Results and discussion

### 6.4.1 Influence of backwash cleaning

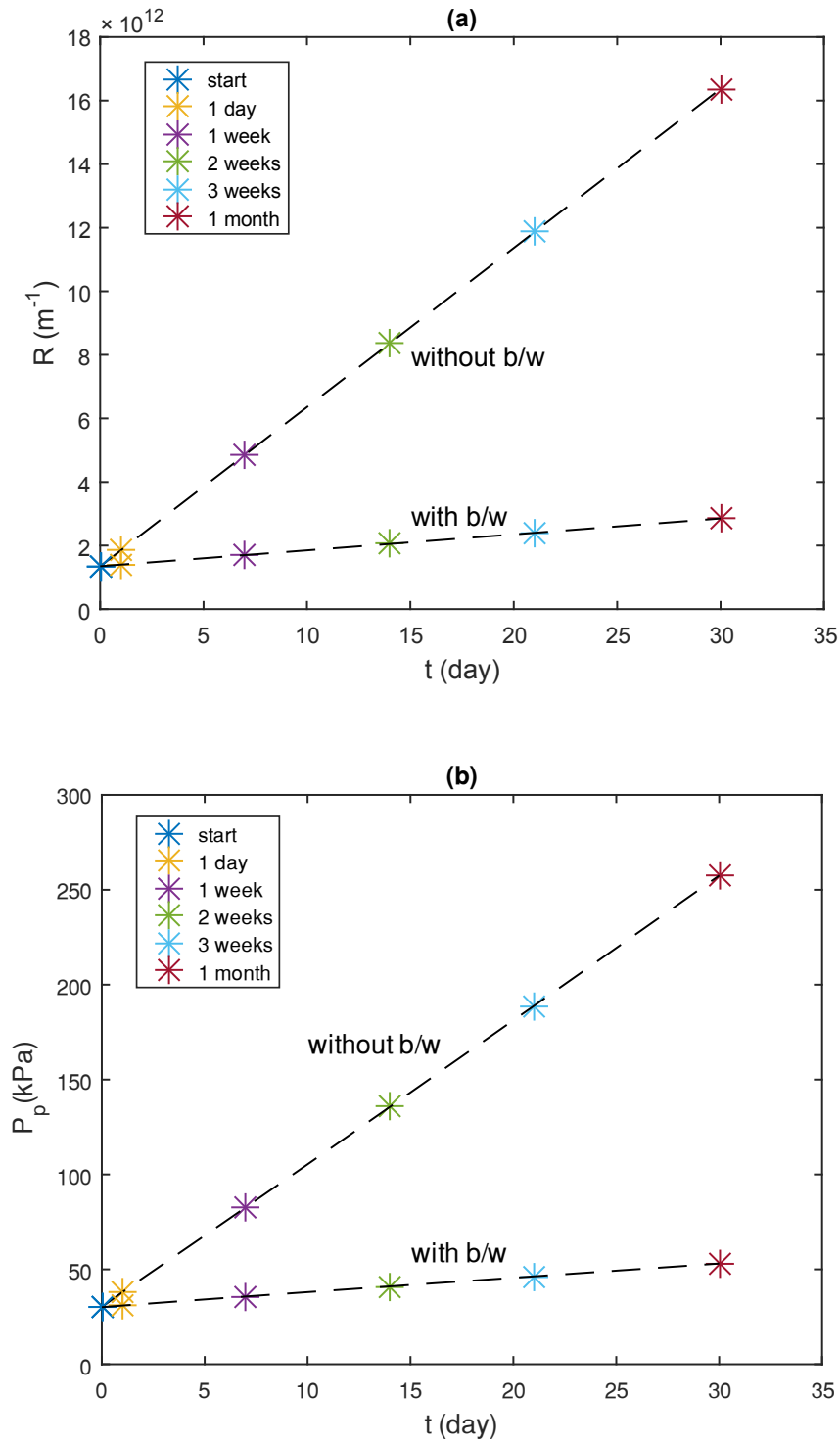
Figure 6.2 shows the normalised productivity and uniformity factor under the conditions with or without backwash cleaning processes. Under fouling conditions both the productivity and the uniformity of the filtration decreases with time. The filtration became even worse without backwash cleaning. After one-month operation (shown in Figure 6.2a), the normalized module productivity,  $\beta$ , reduces from 0.466 to 0.0544 without backwash cleaning, whereas  $\beta$  only reduces from 0.466 to 0.2639 with backwash cleaning incorporated at every 30 mins. This

indicates a reduction in productivity of 88% and 43% without and with backwash cleaning respectively. In Figure 6.2b, the uniformity factor,  $\varphi$ , reduces from 0.9801 to 0.8185 while  $\varphi$  only reduces from 0.9801 to 0.8682 with backwash cleaning every 30 mins. With backwash cleaning process, the filtration system is able to regain its membrane permeability by removing particles attached to the membrane surface. The higher the frequency of the backwash, the less the fibre integrity loss will be found in the system. Also, the variation in the filtration performances between non-cleaning and cleaning operations will be greater with time processes.



**Figure 6.2:** (a) The normalized module productivity,  $\beta$ , and (b) the uniformity of the filtration behaviour,  $\phi$ , of the filtration process versus time,  $t$ , under fouling conditions with or without backwash cleaning. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65$  mm,  $a = \frac{3}{5}b$ ,  $L = 1.6$  m,  $J_{av} = 70$  L m h,  $k_m = 300$  L m h/bar, and  $t_c = 30$  mins.

As shown in Figure 6.3a, without backwash cleaning for a month, the total resistance in the filtration module would be more than 10 times the clean membrane resistance. With backwash cleaning operates at every 30 mins, the resistance only doubles after a month. In Figure 6.3b, the pump driving pressure increases from 30 kPa to 258 kPa without backwash cleaning, while incorporating a backwash cleaning at every half an hour the pressure only increased to 53 kPa after a month. In industry, a typical pump driving pressure in membrane filtration usually lies between 20 kPa and 100 kPa (though some modules can withstand up to 300 kPa) (Pearce, 2011). With a  $P_p$  value  $< 60$  kPa, it is possible for a membrane system to maintain stable performance just by use of a maintenance cleaning strategy based on chemical enhanced backwash. Therefore, the direct-flow filtration module with the dimensions detailed in Figure 6.3 would be safe to operate for a month. Thus the fouling conditions chosen represent the scenario of PDI operation in which  $t_c = 30$  mins ensures 6 weeks of operations before the need for a CIP clean. The maximum  $P_p$  value to ensure stable low fouling operation in practice is 70 kPa, above which there will be a progressive need for recovery cleaning with CIP or even more frequent CEB operations. This is due to the sharply raised pressure in the backflushing process. When the pump driving pressure increases to 140 – 300 kPa, there will be permeability loss in the filtration and in an even worse situation fibre damage could happen. Thus the simulation model does reflect typical industrial practice as reported by (Pearce, 2011).



**Figure 6.3: (a)** The overall resistance,  $R$ , and **(b)** the pump driving pressure,  $P_p$ , of the filtration process versus time,  $t$ , under fouling conditions with or without backwash cleaning. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65$  mm,  $a = \frac{3}{5}b$ ,  $L = 1.6$  m,  $J_{av} = 70$  L m h,  $k_m = 300$  L m h/bar, and  $t_c = 30$  mins.

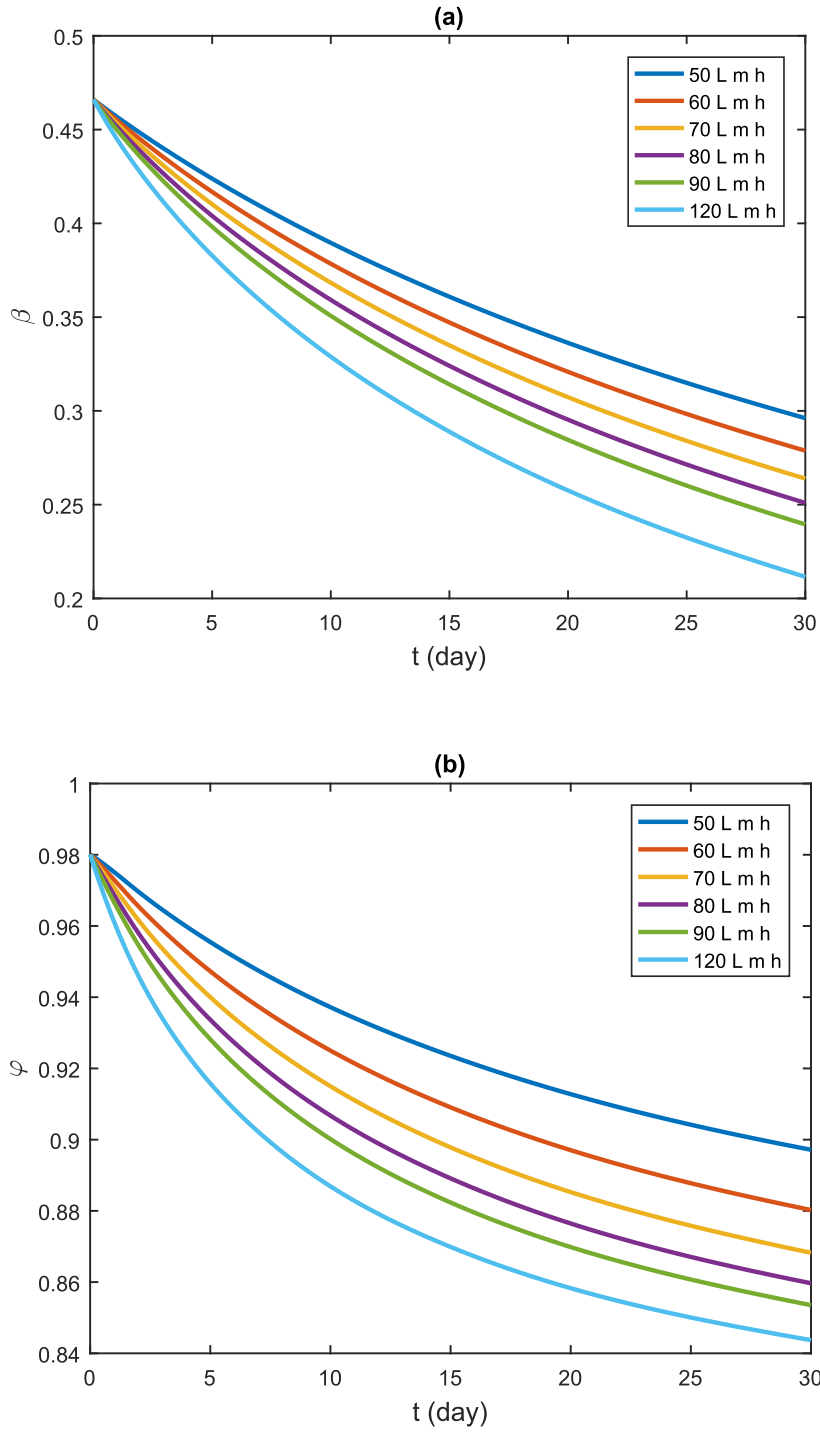
### 6.4.2 Influence of imposed average flux

In Chapter 5, the effect of imposed average flux,  $J_{av}$ , (based on inside fibre membrane area  $2\pi aL$ ) on the filtration performance is not obvious under non-fouling conditions (see Figure 5.12). Figure 6.4 presents the filtration profiles at various  $J_{av}$  values with fouling and backwash cleaning incorporated in the models. At high imposed average flux values, more productivity and uniformity loss are found as the filtration processes with time. For instance, operating at 120 L m h for a month, the productivity loss is 55% (light blue line in Figure 6.4a) and the uniformity loss is 14% (light blue line in Figure 6.4b). In contrast, the relevant productivity and uniformity loss is 36% and 8% when operating at 50 L m h (dark blue lines in Figure 6.4a and b). Even operating at 90 L m h, the productivity and uniformity loss is 49% and 13% respectively, which are still much higher than the loss at 50 L m h. The profiles shown in Figure 6.4 clearly indicate that too high a starting flux will lead to over fouling thus is not recommended in commercial operations. Also, increase the imposed average flux will generate higher cross-flow velocities and higher pressure drops across the module; as a consequence higher power consumption is required (Scott and Hughes, 2012).

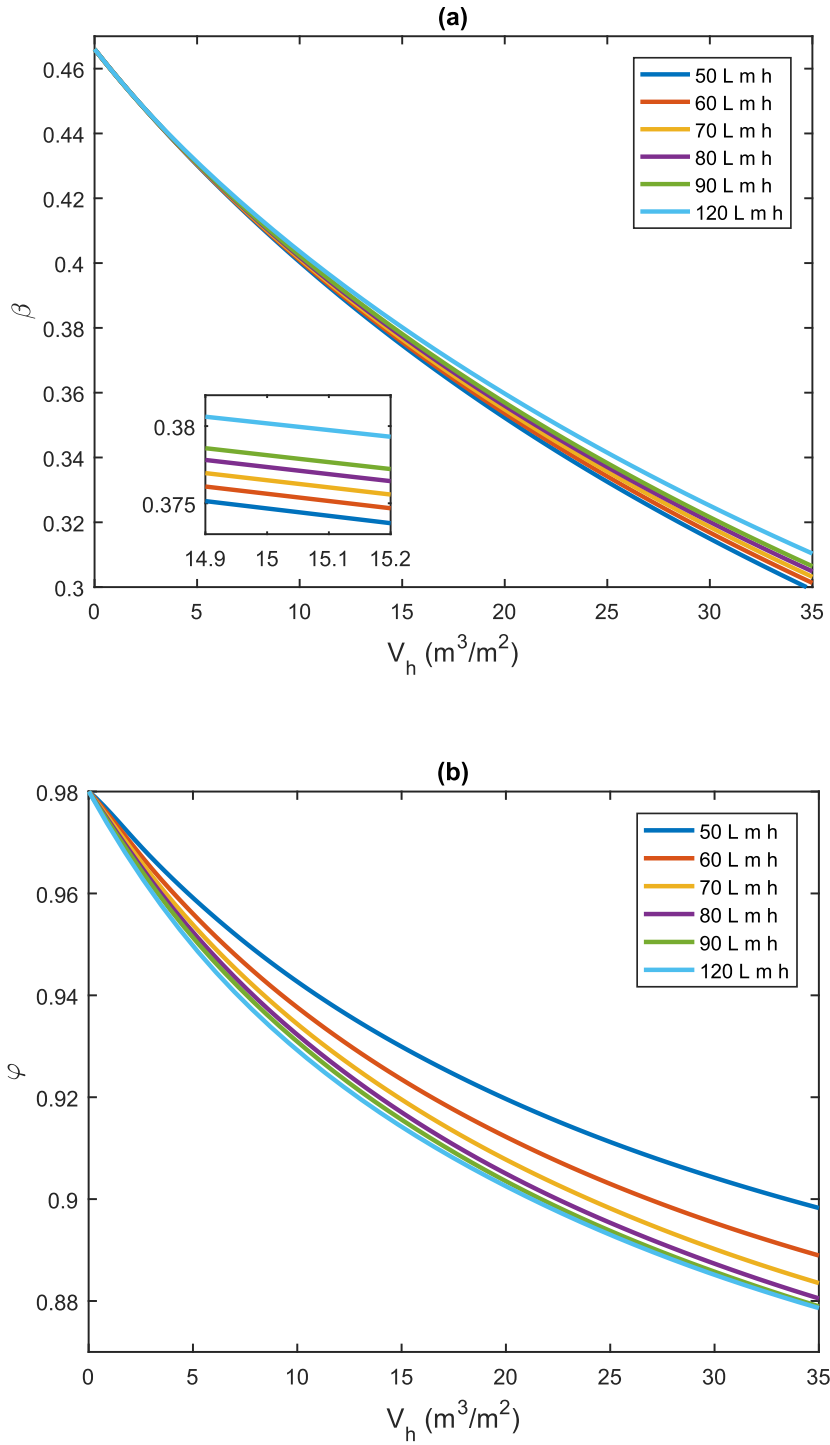
On the other hand, a higher imposed average flux is expected since more output can be achieved through the filtration process over the same time period. e.g. process same amount of fluid feed, operating at 50 L m h requires more than twice the time needed for 120 L m h operation. To quantify the overall filtration performance by considering both the process time and the fouling effects,  $V_h = J_{av} t$  is defined so that  $V_h$  is in units  $\text{m}^3/\text{m}^2$ , which represents the amount of feed fluid has been processed. Figure 6.5 shows the productivity and uniformity profiles at various

imposed average flux values for the processing of the same amount of fluid feed. It was found that higher  $\beta$  values are observed at high  $J_{av}$  operations (Figure 6.5a), but such values would result in a lost in filtration uniformity (Figure 6.5b). Therefore both very high and very low  $J_{av}$  values should be avoided to ensure both high productivity and high uniformity in the filtration.

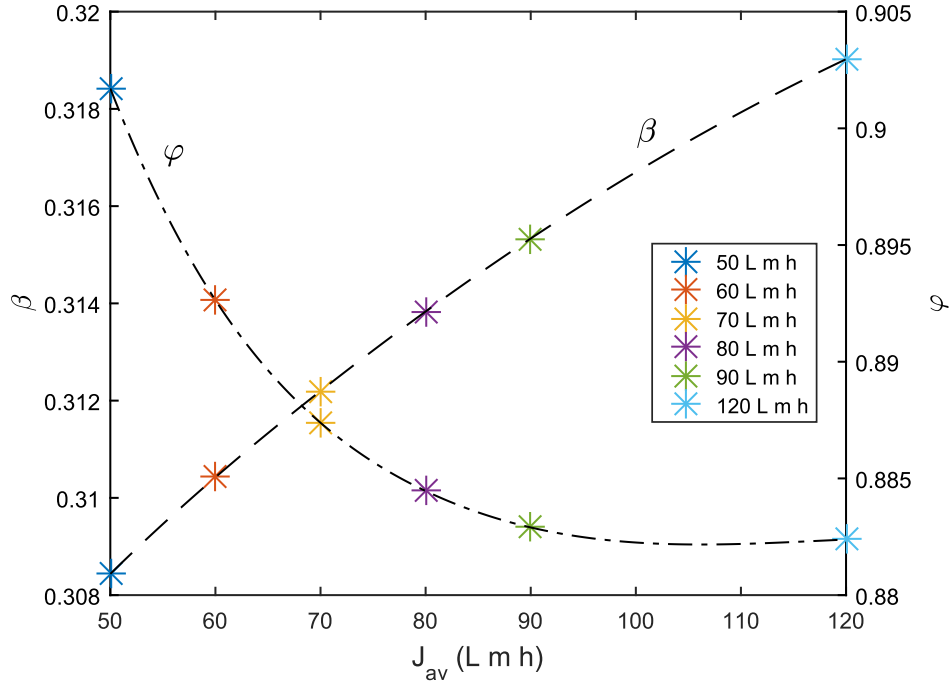
Figure 6.6 shows the profiles of  $\beta$  and  $\varphi$  at various imposed average fluxes when process  $32 \text{ m}^3/\text{m}^2$  fluid feed. Although not shown in this thesis, the plots in Figure 6.6 are reproduced at other  $V_h$  values and the profiles are comparable. The results indicate an optimum operating imposed average flux around  $65 - 70 \text{ L m h}$ . Overall, a recommendation of  $60 - 80 \text{ L m h}$  is made from the investigation in this study. The observation is indeed in line with current commercial operations to approach fouling controls.



**Figure 6.4:** (a) The normalized module productivity,  $\beta$ , and (b) the uniformity of the filtration behaviour,  $\phi$ , of the filtration process versus time,  $t$ , under fouling conditions with backwash cleaning incorporated. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65$  mm,  $a = \frac{3}{5}b$ ,  $L = 1.6$  m,  $k_m = 300$  L m h/bar, and  $t_c = 30$  mins.



**Figure 6.5:** (a) The normalized module productivity,  $\beta$ , and (b) the uniformity of the filtration behaviour,  $\phi$ , of the filtration process versus processed fluid feed,  $V_h$ , at various imposed average values under fouling conditions with backwash cleaning incorporated. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65$  mm,  $a = \frac{3}{5}b$ ,  $L = 1.6$  m,  $k_m = 300$  L m h/bar, and  $t_c = 30$  mins.

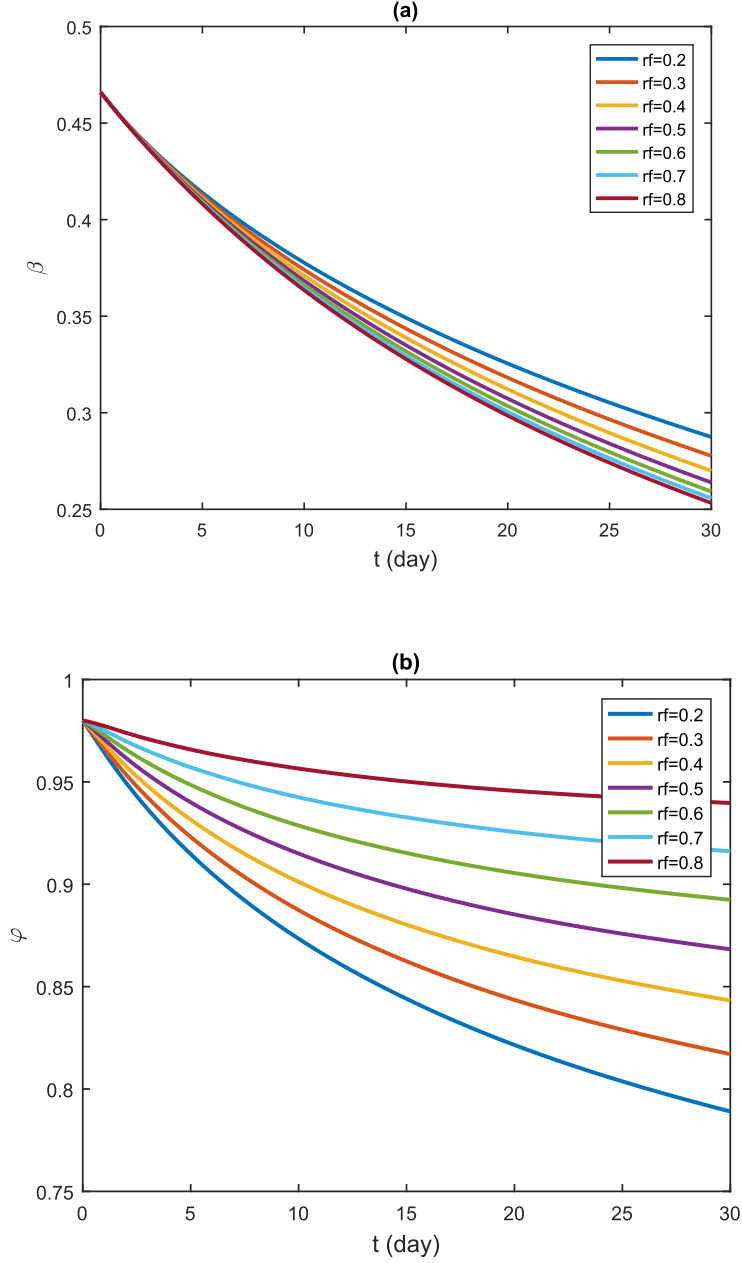


**Figure 6.6:** The normalized module productivity,  $\beta$ , and the uniformity of the filtration behaviour,  $\phi$ , of the filtration process versus imposed average flux,  $J_{av}$ , under fouling conditions with backwash cleaning incorporated. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65$  mm,  $a = \frac{3}{5}b$ ,  $L = 1.6$  m,  $k_m = 300$  L m h/bar,  $t_c = 30$  mins, and  $V_h = 32$  m<sup>3</sup>/m<sup>2</sup>.

### 6.4.3 Influence of fraction of flux and crossflow effects

In literature, both the flux and the crossflow in the filtration system have an impact on the fouling development (Pearce, 2011). The impact was apportioned between the two by defining the fraction of the flux effect to the overall effects as  $rf$ . The fraction of the crossflow effect therefore can be written as,  $1 - rf$ . The practical model developed in this chapter initially assumed equal contribution, i.e.  $rf = 0.5$ . Figure 6.7 shows how the ratio of the flux effect will influence the filtration performance. As shown in the profiles, a higher  $rf$  value will result in a lower productivity factor,  $\beta$ , and a higher uniformity factor,  $\phi$ . Although the percentage of flux and

crossflow effects are not clear in reality, Figure 6.7 is presented in this thesis so that one can use the profiles as a reference for future investigations.

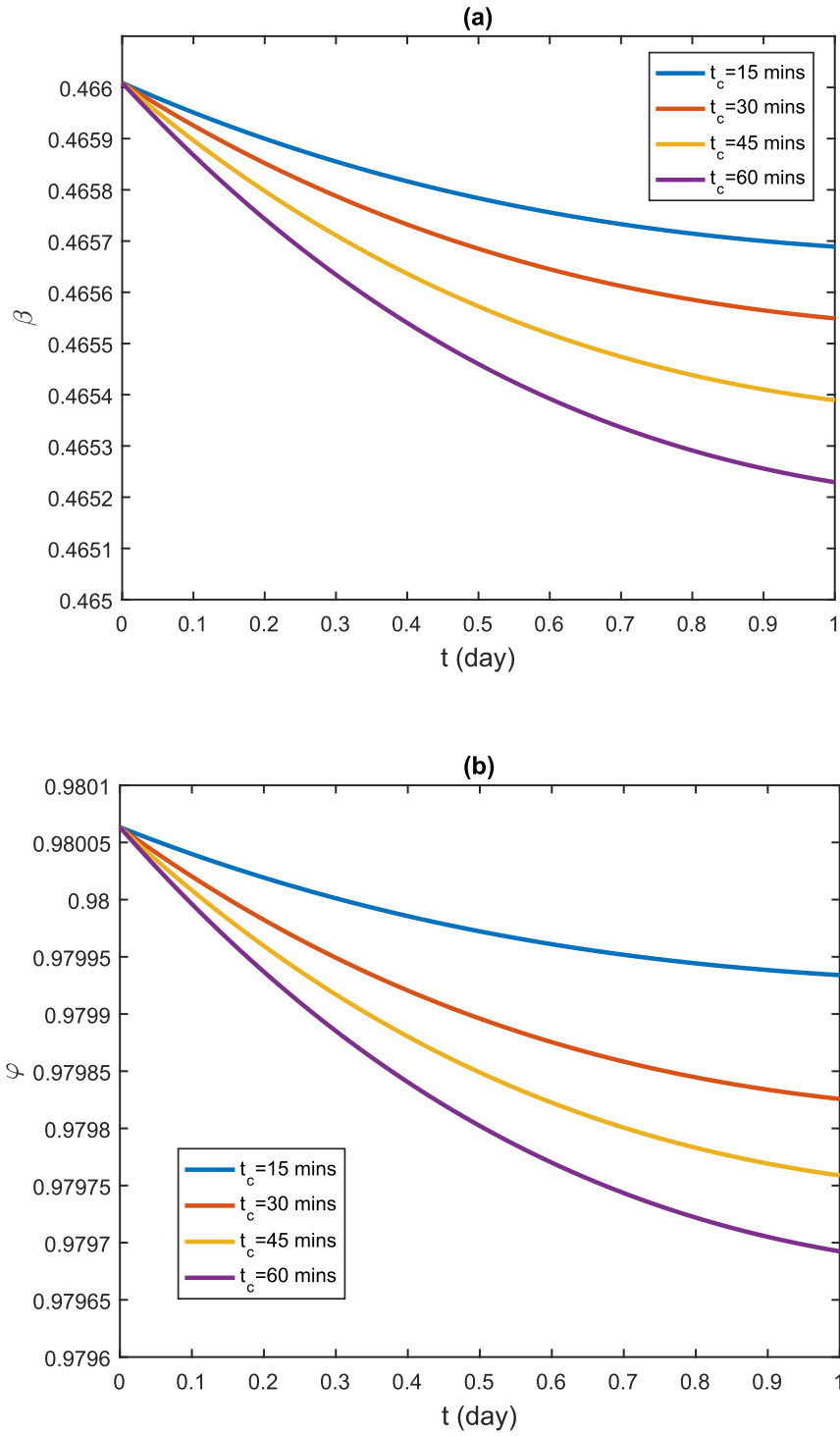


**Figure 6.7:** (a) The normalized module productivity,  $\beta$ , and (b) the uniformity of the filtration behaviour,  $\phi$ , of the filtration process versus time at various  $rf$  ratios. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65$  mm,  $a = \frac{3}{5}b$ ,  $L = 1.6$  m,  $J_{av} = 70$  L m h,  $k_m = 300$  L m h/bar, and  $t_c = 30$  mins.

#### 6.4.4 Influence of backwash cleaning frequency

In direct-flow filtration, the backwash cleaning process is an important procedure to maintain long term operational stability. As discussed in Section 3.2.2.1, the procedures are optimised for different applications. The backwash frequency, which is a vital parameter for successful long-term operation of the system (Smith et al., 2006, Xie et al., 2004), also varies between different module designs. The backwash frequency is normally 1 – 4 times per hour. The model developed in this chapter was firstly designed to have a backwash frequency at every half an hour, i.e.  $t_c = 30$  mins. Figure 6.8 shows how the backwash frequency influences the filtration productivity and the uniformity. The profiles indicate that the more frequent the backwash, then there is both higher productivity and higher uniformity.

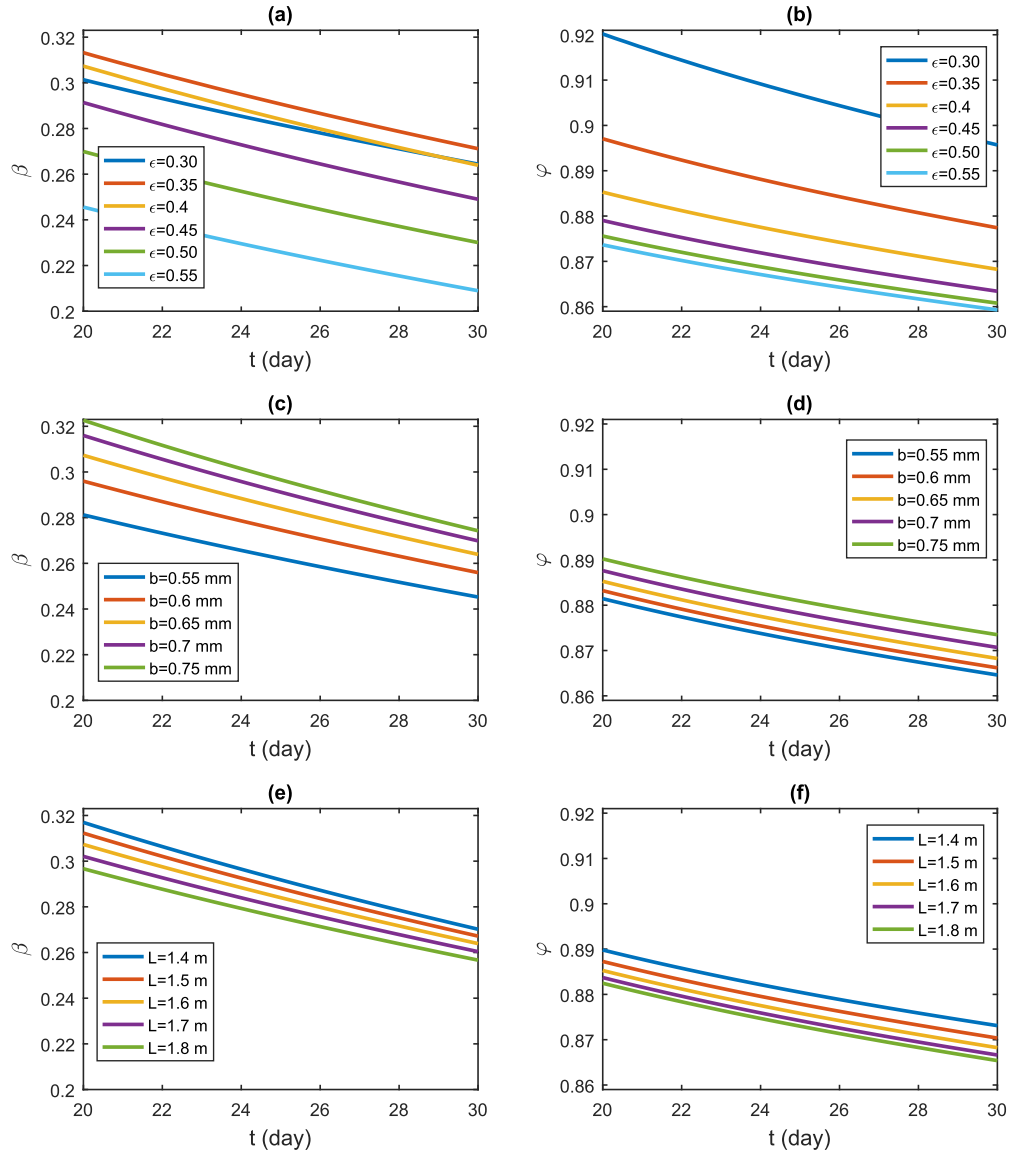
On the other hand, the backwash cleaning process does take time and the normal filtration (production) cycle is interrupted, e.g. interruption of 400 s in every hour with backwash cleaning at every 15 mins while 100 s interruption at every 60 mins cleaning. In literature, productivity losses were reported for increased backwash frequency and predetermination of the exact optimal backwash frequency is impossible (Smith et al., 2006). Although a higher backwash cleaning frequency is recommended from this study, the selection of backwash frequency in reality should always take the foulant concentration, permeate flux and the operational conditions into account. The downtime associated with frequent cleaning has a negative impact but fouling accumulation from cycle to cycle will be minimised by frequent b/w. The balance will be application specific.



**Figure 6.8:** (a) The normalized module productivity,  $\beta$ , and (b) the uniformity of the filtration behaviour,  $\phi$ , of the filtration process versus time at various backwash cleaning frequencies. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65$  mm,  $a = \frac{3}{5}b$ ,  $L = 1.6$  m,  $J_{av} = 70$  L m h, and  $k_m = 300$  L m h/bar.

### **6.4.5 Influence of fibre geometry (void fraction, fibre length, fibre radius)**

In Figure 6.9, the profiles of the filtration productivity and the uniformity are compared at various void fractions,  $\epsilon$ , fibre outer diameters,  $b$ , fibre lengths,  $L$ , so that the results are comparable to previous non-fouling results (as seen in Figure 5.10 and Figure 5.12). The plots reveal that increasing the module void fraction,  $\epsilon$ , from 0.3 to 0.4 can increase both the productivity and the uniformity, however, when increasing  $\epsilon$  from 0.4 to 0.55 both these parameters decrease (Figure 6.9a and b). As a result, an optimum void fraction design is recommended at around 0.4. As shown in Figure 6.9c, d, e and f, increasing the fibre outer radius or decreasing the fibre length will both contribute to a better filtration performance (higher  $\beta$  and  $\varphi$  values). The plots presented in Figure 6.9 are therefore in agreement with non-fouling results found previously.



**Figure 6.9:** (a) (c) (e) The normalized module productivity,  $\beta$ , and (b) (d) (f) the uniformity of the filtration behaviour,  $\varphi$ , of the filtration process versus time at various void fraction, fibre diameter and fibre lengths. Standard parameters:  $\epsilon = 0.4$ ,  $b = 0.65$  mm,  $a = \frac{3}{5}b$ ,  $L = 1.6$  m,  $J_{av} = 70$  L m h,  $k_m = 300$  L m h/bar, and  $t_c = 30$  mins.

In combination with the results summarised in previous chapters, the research in this chapter suggests that a direct-flow hollow fibre module should be made with void fraction of 0.35 – 0.45, reasonable inner radius larger than 0.4 mm, fibre lengths between 1.2 – 1.8 m and operated at 60 – 80 L m h. Imposed average fluxes in this range will ensure a balanced trade-off between module productivity and filtration uniformity. In practice the fibre wall thickness is usually designed together with the fibre inner radius with the former being around 2/3rds of the latter. Overall, in the absence of any detailed information on fouling of hollow fibres used in direct-flow mode (e.g. spatial info) a flexible generic approach to the modelling of fouling was taken in this chapter. Table 6.1 summaries the recommendations for the geometrical parameters and the operating conditions against commercial ranges.

**Table 6.1:** Summary of investigation results in this thesis.

| Sources                                 | Void fraction                  | Inner radius (mm)      | Wall thickness (mm)           | Fibre length (m)      | Imposed average flux (L m h) | Backwash frequency (per hour) |
|-----------------------------------------|--------------------------------|------------------------|-------------------------------|-----------------------|------------------------------|-------------------------------|
| <b>This thesis</b>                      | $0.35 \leq \epsilon \leq 0.45$ | $a \geq 0.4$           | $0.15 \leq \Gamma \leq 0.3$   | $1.2 \leq L \leq 1.8$ | $60 \leq J_{av} \leq 80$     | 3-4 times                     |
| <b>Commercial ranges (Pearce, 2011)</b> | $0.4 \leq \epsilon \leq 0.45$  | $0.4 \leq a \leq 0.45$ | $\Gamma \approx \frac{2}{3}a$ | $1.5 \leq L \leq 1.8$ | $60 \leq J_{av} \leq 80$     | 1-4 times                     |
| <b>Concerns</b>                         | Overall module size            | Mechanical strength    | Mechanical strength           | Overall module size   | Membrane fouling             | Interruption to filtration    |

## 6.5 Conclusion

In this chapter, a practical membrane fouling model is developed for inside feed direct-flow hollow fibre filtration. With this practical model, particle deposition and backwash cleaning processes were investigated under various operating conditions. The results in this chapter reviews the results summarised under non-fouling conditions in Chapter 4 and 5. More importantly, the results revealed previously undiscovered effects of imposed average fluxes on the filtration performances and showed the importance of backwash cleaning processes. Overall, the research suggests that a direct-flow hollow fibre module made with void fraction at 0.35 – 0.45, reasonable inner radius larger than 0.4 mm, fibre lengths between 1.2 – 1.8 m and operating at 60 – 80 L m h will ensure a balanced trade-off between module productivity and filtration uniformity (see Table 6.1). This chapter not only double confirms the empirical observation of module void fraction, fibre length and radius but also proves the empirical range of imposed average fluxes.

# Chapter 7

## Concluding Remarks and Future Work

### 7.1 Concluding Remarks

The objective of this study was to gain a better understanding of direct-flow filtration by addressing its operation and design guidelines. The research started with an overview of membrane filtration technologies (Chapter 2) and was then followed by a comprehensive literature survey on direct-flow filtration (Chapter 3). After the background knowledge review, a dimensionless mathematical model was firstly developed for the fluid flow in an axisymmetric direct-flow pipe (Chapter 4). The model was derived without considering the effect of permeable wall thickness. The results predicted the operating regimes that optimized both the productivity and the uniformity of the filtration process. To maximize the uniformity of use of the filtration area, direct-flow filtration modules should be operated at low fluxes, be composed of a filter material with a relatively low permeability and have neighbouring module spacing such that the ratio of the volumes on the feed and permeable side are approximately equal. The results also highlighted that for inside feed direct-flow filtration a large portion of the available membrane area will not be used when the fibres are packed too closely together; in this case the majority of the filtration becomes localized near the impermeable capped-end.

After identifying the basic hydrodynamic phenomena of direct-flow filtration through the mathematical model, a realistic hollow fibre model was derived which included the fibre wall thickness (Chapter 5). The realistic direct-flow hollow fibre model was then used to explore a

broad range of realistic module dimensions encompassing the values applied in today's water industry, including module void fraction, fibre radius, fibre wall thickness, and fibre length. The modelling results showed that at small void fraction values, the fluid hardly goes through the membrane near the entrance and thus the majority of the filtration process takes place near the capped-end. Increasing void fraction generates a more uniform filtration process along the axial length. However, continuingly increase its value beyond 0.5 will hardly change the uniformity profiles but there will be fewer fibres per module. The trade-off between the uniformity and the productivity was also considered with regard to optimizing the fibre geometries. Increasing fibre diameter or reducing fibre length both promote uniform filtration. Apart from the influence of imposed average fluxes, a thorough understanding has been achieved by the combination of productivity and uniformity profiles. The study revealed that the optimized ranges for inside feed direct-flow filtrations are as follows void fraction (0.35 – 0.45), fibre inner radius larger than (0.4 mm), fibre wall thickness (0.15 – 0.3 mm), and fibre length (1.2 – 1.8 m).

Although the study of the imposed average flux effects was also carried out by using the models under non-fouling conditions, the effects of flux were not clear. After considering the importance of particle deposition and fouling effects, a practical direct-flow filtration model was developed so that fouling phenomena and backwash cleaning effects were both incorporated (Chapter 6). With this practical model, particle deposition and the backwash cleaning processes were investigated under various operating conditions. The study emphasised the backwash cleaning and its frequencies are curial to fouling controls in direct-flow filtration. The investigation also re-investigated the optimized results summarised under non-fouling conditions. Especially, the results indicated the problems raised in filtration uniformity and productivity when operating under high imposed average fluxes. The investigation revealed the optimized range of imposed

average fluxes at 60 – 80 L m h , which is indeed in accord with current commercial operations.

Overall, this study provided a comprehensive approach to optimize direct-flow hollow fibre filtration in the inside feed format. The work presented confirms the empirical observation of module void fraction, fibre radius, fibre length and imposed average fluxes over the last 20 years in real industry. The discovered evidences behind the ranges currently used will aid future design of commercial membrane products when new membrane materials become available.

## **7.2 Future Work**

There are many possibilities to extend the current study on direct-flow filtration in the future. Firstly, only pressure driven inside feed direct-flow filtration under constant flux operations were investigated in the present work. In industry, direct-flow filtration can be operated with different feed formats (inside or outside feed) under various operating modes (constant flux or constant pressure). The inside feed models developed in Chapter 4, 5, and 6 together with the outside feed models developed in Appendix-A provide the possibility to conduct a comparison of direct-flow filtration of different formats.

Secondly, due to time and MATLAB modelling limitations, the work presented in this thesis only investigated the fouling and backwash cleaning cycles within a month's operation at each operating condition. In future studies, there exists the possibility to conduct a longer time fouling study of direct-flow filtration. Especially, over time the module productivity will largely be offset by the increased resistance from the irreversible fouling layer. Therefore, chemical enhanced backwash cleaning and cleaning-in-place should also be involved in future investigations.

Thirdly, in addition to module void fraction (packing density) itself the uniformity of packing, fibre lengths and the polydispersity of fibre diameters will also have an effect on the filtration behaviours even in the absence of fouling development. Therefore, possible studies can be conducted to investigate the effects of the packing uniformity upon filtration behaviours of direct-flow hollow fibre filtration. The optimum ranges summarised in this thesis may be refined once a thorough study on the effect of spatial variations in the fibre packing and particle deposition has been undertaken.

Additionally, the models presented in this thesis ignored the radial differences in the hollow fibre module. A more complicated direct-flow filtration model including the spatial variations along the radial axis would help one better understand the filtration performance. Fouling in membrane filtration is complex and heavily depends on the properties of components in the feed stream. The current models herein have the limitation of being for feed streams with a low level of incompressible, chemically inert, particulate foulants. Future developments could involve adsorption of organics and potentially the development of a gel phase. The evolution of fouling resistance with foulant accumulation should also be assessed; currently a simple linear relationship is assumed. Apart from pore blocking arises from suspended solids, other fouling mechanisms may also be incorporated in future modelling works.

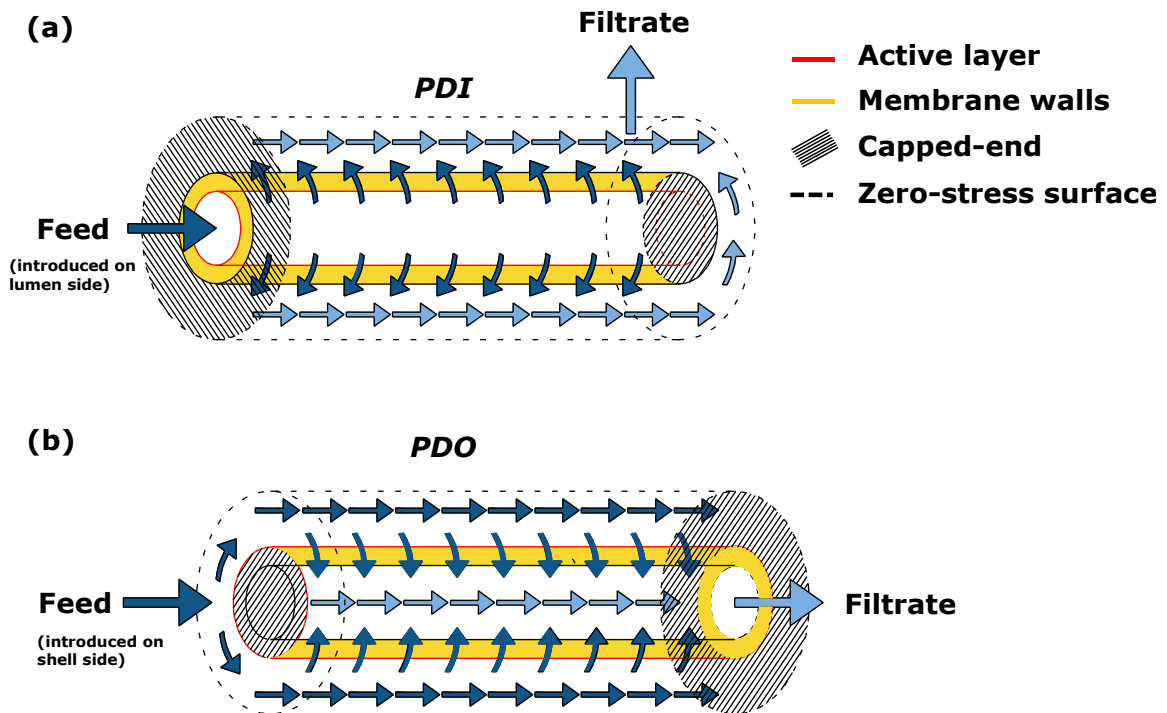
Finally, the work presented in this thesis are all based on mathematical and theoretical modelling evaluations. Although the modelling results were constantly reviewed by professionals and academic researchers working in the membrane field, some experimental work in the future such as a post mortem study of used modules (which would give a permeability profile  $k_m(z)$ ) could help one to extend our knowledge on direct-flow hollow fibre filtrations.

# **Appendix-A Direct-Flow Filtration in PDO format**

This appendix develops direct-flow hollow fibre filtration models in pressure driven outside feed format. PDO models are introduced so that a comparison of inside and outside feed filtration format can be made in the future.

## A.1 Introduction

As discussed in Section 3.1.1, direct-flow hollow fibre membrane filtrations can be configured to operate in either PDI or PDO format (shown in Figure A.1). The major difference between the configurations is where the feed flow is introduced. For PDI arrangement, the feed is introduced into the lumen of the fibre with filtrate being withdrawn/collected from the outside or shell-side of the module, while the opposite flow configuration operates in PDO modules.



**Figure A.1:** Schematic of process flows of the two formats of direct-flow filtration illustrated for a single fibre. **(a)** Pressure driven inside feed (PDI) format; **(b)** Pressure driven outside feed (PDO) format.

## A.2 PDO model development

Similar to PDI model developed in Chapter 5, a corresponding model based on bundles of small porous hollow fibres is developed for PDO mode. The feed flow is introduced in the shell side and is filtrated into the lumen side of fibres. For simplicity, single isolated fibre as illustrated in Figure A.1b is focused. Each fibre has an internal radius  $a$ , an outer radius  $b$  and a length  $L$ . A symmetry condition is applied from the fibre centrally so that a radial coordinate system  $(r, z)$  can be used. In the axisymmetric coordinate,  $r$  denotes the radial coordinate and  $z$  represents the distance along the fibre length  $L$ . Each fibre has an imaginary boundary, known as zero-stress outer surface, which is centred around the fibre with a radius  $c$ . The flow rate in the shell side is denoted by  $\dot{V}$  and flow rate in the lumen side is  $\dot{Q}$ .

In this PDO operation, a constant shell side flow rate  $\dot{V}_1 = 2\pi bLJ_{av}$  is applied to each isolated fibre based on the outside fibre area. The fluid velocity in  $r$  direction is  $u$ , and velocity along the fibre length direction is  $w$ . The pressure in the shell side is  $q$ , and in the lumen side is  $p$ . The transmembrane pressure difference TMP, now becomes  $q - p$  in PDO mode. As explained in previous chapters, a HFM used in commercial practice normally has an aspect ratio (diameter/length) of the module  $\delta = a/L \ll 1$  and the reduced Reynolds number  $\delta Re = O(10^{-2}) \ll 1$ . Therefore, lubrication equations can also be applied in PDO filtration mode.

In shell side (for  $b \leq r \leq c$  and  $0 \leq z \leq L$ ), the lubrication equations are

$$\frac{1}{r} \frac{\partial}{\partial r} (ru) + \frac{\partial w}{\partial z} = 0, \quad (\text{A.1})$$

$$\mu \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial w}{\partial r} \right) = \frac{dq}{dz}. \quad (\text{A.2})$$

The boundary conditions for shell side can be defined as

$$\text{Zero stress surface:} \quad \frac{\partial w}{\partial r} = 0; u = 0, \quad \text{when } r = c, \quad (\text{A.3})$$

$$\text{No axial wall slip:} \quad w = 0, \quad \text{when } r = b, \quad (\text{A.4})$$

$$\text{Permeate flow:} \quad u = \frac{k(p - q)}{\mu b \ln \frac{b}{a}}, \quad \text{when } r = b, \quad (\text{A.5})$$

$$\text{Fluid inflow:} \quad 2\pi \int_b^c wr \, dr = \dot{V}_1, \quad \text{when } z = 0, \quad (\text{A.6})$$

$$\text{No fluid outflow:} \quad w = 0, \quad \text{when } z = L. \quad (\text{A.7})$$

Lumen side region (for  $0 \leq r \leq a$  and  $0 \leq z \leq L$ ) the lubrication equations are

$$\frac{1}{r} \frac{\partial}{\partial r} (ru) + \frac{\partial w}{\partial z} = 0, \quad (\text{A.8})$$

$$\mu \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial w}{\partial r} \right) = \frac{dp}{dz}. \quad (\text{A.9})$$

The boundary conditions for lumen side can be defined as

$$\text{Lumen side symmetry:} \quad \frac{\partial w}{\partial r} = 0; u = 0, \quad \text{when } r = 0, \quad (\text{A.10})$$

$$\text{No axial wall slip:} \quad w = 0, \quad \text{when } r = a, \quad (\text{A.11})$$

$$\text{Permeate flow:} \quad u = \frac{k(p - q)}{\mu a \ln \frac{b}{a}}, \quad \text{when } r = a, \quad (\text{A.12})$$

$$\text{No fluid inflow:} \quad w = 0, \quad \text{when } z = 0, \quad (\text{A.13})$$

$$\text{Fixed outlet pressure:} \quad p = p_L = 1 \text{ atm} \quad \text{when } z = L. \quad (\text{A.14})$$

The equations can be solved analytically to get the transverse velocity expressions, in shell side region we find,

$$w = \frac{1}{4\mu} \frac{dq}{dz} \left( r^2 - 2c^2 \ln \frac{r}{b} - b^2 \right), \quad (\text{A.15})$$

$$u = \frac{1}{16\mu} \frac{d^2q}{dz^2} \left\{ 2b^2r - r^3 \right. \\ \left. + c^2 \left[ -2r + 4r \ln \frac{r}{b} - \frac{2b^2}{r} + c^2 \left( \frac{3}{r} - \frac{4}{r} \ln \frac{c}{b} \right) \right] \right\}, \quad (\text{A.16})$$

while in lumen side region

$$w = \frac{1}{4\mu} \frac{dp}{dz} (r^2 - a^2), \quad (\text{A.17})$$

$$u = \frac{1}{16\mu} \frac{d^2p}{dz^2} (2a^2r - r^3). \quad (\text{A.18})$$

Substituting the boundary equations into the velocity expressions, a system of ODEs can be read

$$\frac{d^2p}{dz^2} = \frac{16k(p - q)}{\Lambda_1}, \quad \frac{dp}{dz} \Big|_{z=0} = 0, \quad p|_{z=L} = p_L = 1 \text{ atm}, \quad (\text{A.19})$$

$$\frac{d^2q}{dz^2} = -\frac{16k(p - q)}{\Lambda_2}, \quad \frac{dq}{dz} \Big|_{z=0} = \Lambda_3, \quad \frac{dq}{dz} \Big|_{z=L} = 0, \quad (\text{A.20})$$

where

$$\Lambda_1 = a^4 \ln \frac{b}{a}, \quad (\text{A.21})$$

$$\Lambda_2 = -b^4 \ln \frac{b}{a} + c^2 \ln \frac{b}{a} \left[ 4b^2 - c^2 \left( 3 - 4 \ln \frac{c}{b} \right) \right], \quad (\text{A.22})$$

$$\Lambda_3 = \frac{8\mu\dot{V}_1}{\pi (3c^4 - 4c^4 \ln \frac{c}{b} - 4b^2c^2 + b^4)}. \quad (\text{A.23})$$

The ODEs can be solved analytically to get

$$p(z) = \frac{1}{\Lambda_1 + \Lambda_2} [-\Lambda_2 \mathcal{N}(z) - \Lambda_2 \Lambda_3 (L - z) + \mathcal{N}_0] + p_L, \quad (\text{A.24})$$

$$q(z) = \frac{1}{\Lambda_1 + \Lambda_2} [\Lambda_1 \mathcal{N}(z) - \Lambda_2 \Lambda_3 (L - z) + \mathcal{N}_0] + p_L, \quad (\text{A.25})$$

where

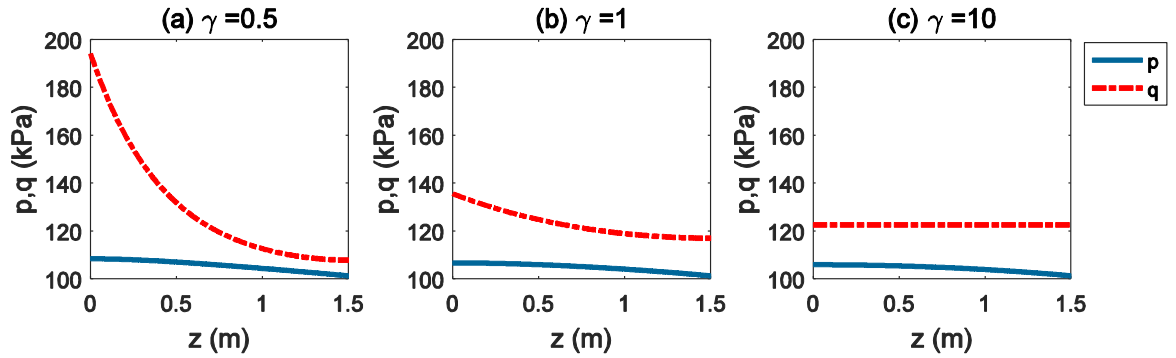
$$\mathcal{N}(z) = -\frac{\Lambda_3}{C_1} \text{Csch}(C_1 L) \left[ \frac{\Lambda_2}{\Lambda_1} \text{Cosh}(C_1 z) + \text{Cosh}[C_1 (L - z)] \right], \quad (\text{A.26})$$

$$\mathcal{N}_0 = -\frac{\Lambda_2 \Lambda_3}{C_1} \left[ \frac{\Lambda_2}{\Lambda_1} \text{Coth}(C_1 L) + \text{Csch}(C_1 L) \right], \quad (\text{A.27})$$

$$C_1 = \sqrt{16k \left( \frac{1}{\Lambda_1} + \frac{1}{\Lambda_2} \right)}. \quad (\text{A.28})$$

### A.3 Flow in PDO direct-flow filtration

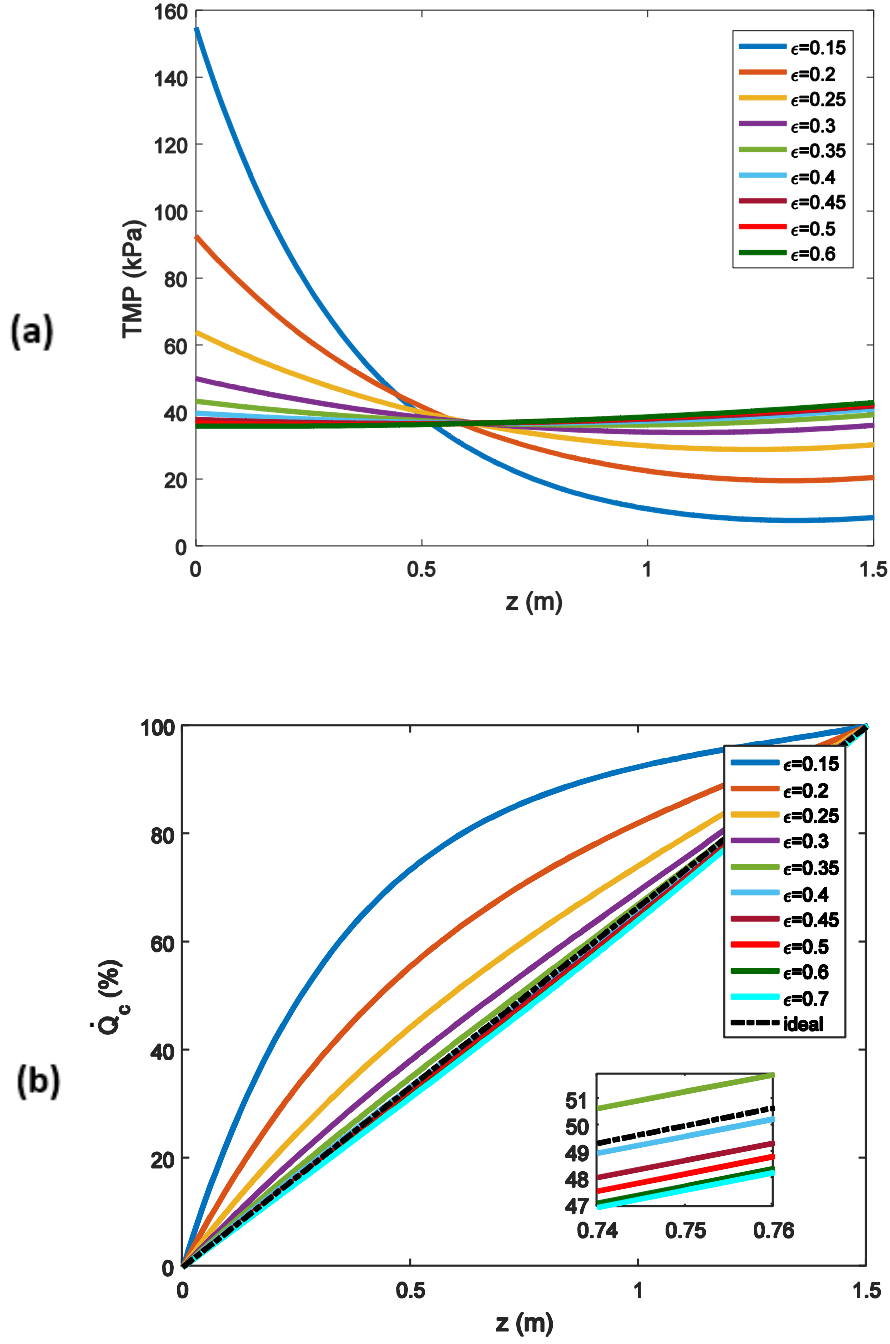
Figure A.2 shows three patterns of pressure profiles at constant flux PDO filtration mode, in parallel with corresponding profiles in PDI mode in Chapter 5 (see Section 5.4.1). When the spacing on the shell side is large as in Figure A.2c the shell side behaves as a quiescent bath that is essentially at the inlet pressure over the whole length. With equal areas on the two sides (Figure A.2b) the difference of  $q - p$  (i.e. TMP) varies modestly along the fibre length, which show analogous in PDI format. Too smaller a  $\gamma$  value (Figure A.2a) introduces a hydrodynamic resistance on the shell side and results in a major maldistribution of TMP and hence of fluxes, i.e. majority of the filtration processes near the entrance.



**Figure A.2:** Variation of PDO mode fluid pressure in the lumen side,  $p$ , and in the shell side,  $q$ . Fixed parameters:  $a = 0.4\text{mm}$ ,  $\Gamma = 0.25\text{mm}$ ,  $L = 1.5\text{m}$ ,  $J_{av} = 70\text{ Lmh}$  and  $k_m = 300\text{ Lmh/bar}$  with spacing parameters **(a)**  $\gamma = 0.5$ , **(b)**  $\gamma = 1$  and **(c)**  $\gamma = 10$ .

In Figure A.3, TMP and cumulative fluid volume profiles are compared at different void fraction,  $\epsilon$ , with other parameters set at the standard values as used in PDI model in Chapter 5. Compare to inside feed filtration, outside feed format provides a higher filtration area therefore the overall volumetric flow is higher at the same imposed average flux. With 70 L m h imposed, PDO filtration show optimized TMP profiles at 40 kPa (Figure A.3a), whereas PDI shows optimized TMP profiles around 25 kPa.

At small  $\epsilon$  values, both PDI and PDO show large variation of the filtration performance along the axial distance. Specifically, the fluid hardly goes through the permeate wall near the entrance in PDI mode while the majority of the filtration process takes place near the entrance in PDO mode. Despite this difference, increasing  $\epsilon$  value promotes uniform filtration in both modes and the profiles suggest the same void fraction range for optimal operation. This will be more closely revealed when the overall filtration performance is quantified both in terms of its productivity and uniformity.



**Figure A.3:** The effect of void fraction,  $\epsilon$ , upon (a) the transmembrane pressure difference (TMP) profiles and (b) the cumulative volume transported through the fibre walls,  $\dot{Q}_c$  (%), versus axial distance,  $z$  in PDO mode. Fixed parameters:  $a = 0.4$ mm,  $\Gamma = 0.25$ mm,  $L = 1.5$ m,  $J_{av} = 70$  Lmh and  $k_m = 300$  Lmh/bar.

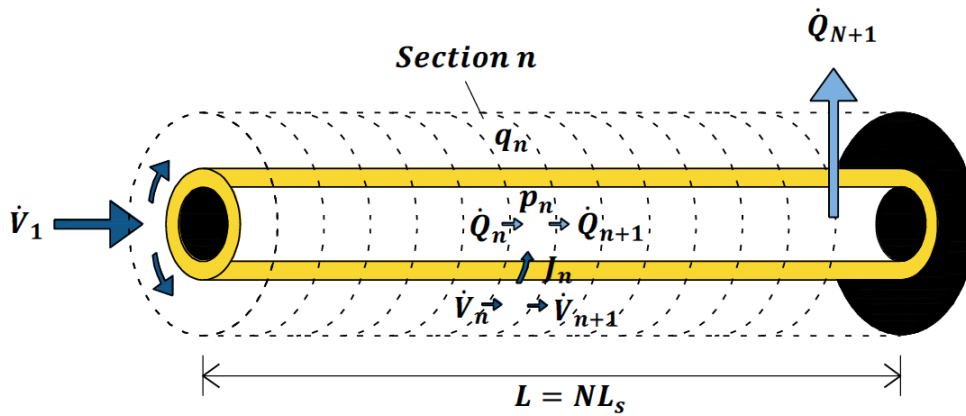
## A.4 Practical PDO model development

Schematics of a corresponding single hollow fibre in PDO filtration mode is illustrated in Figure A.4. As with previous model, the hollow fibre length  $L$  contains  $N$  sections with each section having a small length  $L_s$ . In section  $n$ ,  $p_n$  is the pressure in lumen side and  $q_n$  is the pressure in shell.  $\dot{Q}_n$  and  $\dot{Q}_{n+1}$  are the lumen side volumetric flow rate at the section inlet and outlet,  $\dot{V}_n$  and  $\dot{V}_{n+1}$  are the volumetric flow rate in the shell side respectively. In this practical PDO model,  $J_n$  now denotes the filtration flux from the shell side to the lumen side. The model can be solved by cooperating previous results of general flow Eqs. (6.4) (6.5) (see Section 6.2.1) together with the general filtration equations in PDO mode

$$\dot{V}_n - \dot{V}_{n+1} = 2\pi a L_s J_n = \dot{Q}_{n+1} - \dot{Q}_n, \quad (\text{A.29})$$

$$J_n = \frac{\frac{q_n + q_{n+1}}{2} - \frac{p_n + p_{n+1}}{2}}{\mu R_m}, \quad (\text{A.30})$$

with initial conditions  $\dot{V}_1 = \dot{Q}_{N+1}$ ,  $\dot{Q}_1 = 0$ ,  $\dot{V}_{N+1} = 0$  and  $p_{N+1} = 101325$  kPa.



**Figure A.4:** 3D Schematic flow regimes (n sections) of direct-flow filtration in a single hollow fibre membrane in PDO filtration mode.

## Appendix-B Supplementary of Productivity Parameter ( $\beta$ )

One can define a module productivity factor,  $\beta_M$ , with respect to the hypothetical case of a fully packed module (touching equilateral triangular packing) which assumes the TMP is uniformly distributed at  $P_p$  (the pump driving pressure with respect to the ambient pressure as reference), i.e.  $J_{av} = k_m P_p$ . Now number of fibres,  $N_{fibre}$ , is given by:

$$N_{fibre} = \frac{\text{module crossection area} \times (1 - \epsilon)}{\text{fibre cross section area}} = \frac{\pi D_M^2 (1 - \epsilon)}{4\pi b^2} \quad (\text{B.1})$$

$$= \frac{D_M^2 (1 - \epsilon)}{4b^2}$$

where  $D_M$  is the module diameter,  $\epsilon$  is the module void fraction, and  $b$  is the fibre outside radius.

Therefore the actual volumetric throughput, based on the inside fibre area, is:

$$\dot{V} = \frac{D_M^2}{4b^2} (1 - \epsilon) \cdot 2\pi a L J_{av}. \quad (\text{B.2})$$

Now consider the hypothetical maximum number of fibres inside the module (at touching equilateral triangular packing,  $\epsilon = 0.0931$ ):

$$N_{fibre-max} = \frac{D_M^2}{4b^2} (1 - 0.0931) = 0.9069 \frac{D_m^2}{4b^2}. \quad (\text{B.3})$$

The hypothetical maximum volumetric throughput (which would be approached in a very short module where TMP approaches  $P_p$ ) is:

$$\dot{V}_{max} = 0.9069 \frac{D_M^2}{4b^2} \cdot 2\pi aL J_{av} = 0.9069 \frac{D_M^2}{4b^2} \cdot 2\pi aL \cdot k_m P_p. \quad (B.4)$$

Therefore the module productivity can be defined with respect to this maximum value as:

$$\beta_M = \frac{\dot{V}}{\dot{V}_{max}} = \frac{\frac{D_M^2}{4b^2} (1 - \epsilon) \cdot 2\pi aL J_{av}}{0.9069 \frac{D_M^2}{4b^2} \cdot 2\pi aL \cdot k_m P_p} = \frac{J_{av}}{k_m P_p} \frac{(1 - \epsilon)}{0.9069}, \quad (B.5)$$

where  $\frac{(1-\epsilon)}{0.9069}$  refers to the actual area available for filtration to the maximum theoretically available (in a touching equilateral triangular packing). To simplify the productivity term, a normalised module productivity factor,  $\beta$ , was defined as follows:

$$\beta = \frac{J_{av}}{k_m P_p} (1 - \epsilon). \quad (B.6)$$

For a given value of  $J_{av}$ , as the number of fibres increases so does  $(1 - \epsilon)$ . However the required inlet pressure  $P_p$  also increases. Eq. (B.6) represents the influence of these competing effects.

# Appendix-C Publications and Presentations

## A. Publications

1. Herterich, J.G., **Xu, Q.** (Co-first author), Field, R.W., Vella, D. and Griffiths, I.M., 2017. **Optimizing the operation of a direct-flow filtration device.** *Journal of Engineering Mathematics*, 104(1), pp.195-211. (Based on Chapter 4)
2. **Xu, Q.**, Pearce, G.K. and Field, R.W., 2017. **Pressure driven inside feed (PDI) hollow fibre filtration: Optimizing the geometry and operating parameters.** *Journal of Membrane Science*, 537, pp.323-336. (Based on Chapter 5)
3. **Xu, Q.**, Pearce, G.K. and Field, R.W., 2018. **Time evolution of fouling development in pressure driven inside feed (PDI) hollow fibre filtration.** *Journal of Membrane Science* (to be submitted in July 2018). (Based on Chapter 6)

## **B. Conference Presentations**

1. **Xu, Q.**, Field, R.W., 2016. **Optimizing the operation of a direct-flow filtration device.** *Membrane Modelling and Simulation, Separation Division*, 2016 AIChE Annual Meeting, San Francisco, CA, USA
2. Field, R. W., **Xu, Q.**, 2017. **Operational considerations and design aspects of direct-flow filtration.** 2017 International Congress on Membranes and Membrane Processes, San Francisco, CA, USA
3. **Xu, Q.**, Field, R.W., 2017. **The time evolution of fouling development upon design aspects of direct-flow filtration.** *Water Treatment, Desalination, and Reuse II, Separation Division*, 2017 AIChE Annual Meeting, Minneapolis, MN, USA

# Nomenclature

| Letters  |                                                                                            |
|----------|--------------------------------------------------------------------------------------------|
| $a$      | fibre inside radius (mm)                                                                   |
| $A_m$    | surface area of membrane used (m <sup>2</sup> )                                            |
| $b$      | fibre outside radius (mm)                                                                  |
| $c$      | radius of imaginary circular boundary of each fibre (mm)                                   |
| $CP$     | concentration polarization                                                                 |
| $D$      | radial position of zero-stress surface in 3D pipe model (dimensionless)                    |
| $d_g$    | Sauter diameter                                                                            |
| $d_i$    | fibre inside diameter (mm)                                                                 |
| $d_o$    | fibre outside diameter (mm)                                                                |
| $H$      | width in 2D direct-flow channel (dimensionless)                                            |
| $h_c$    | height of fouling (m)                                                                      |
| $h_k$    | Kozeny constant                                                                            |
| $J$      | permeate flux across the membrane walls (L m h)                                            |
| $J_{av}$ | imposed average flux (L m h)                                                               |
| $J_n$    | flux flow from the lumen side to the shell side across the membrane in section $n$ (L m h) |
| $k$      | permeability (m <sup>2</sup> )                                                             |

|           |                                                                                                            |
|-----------|------------------------------------------------------------------------------------------------------------|
| $k_d$     | dimensionless permeability as defined in 3D pipe model study                                               |
| $k_{eff}$ | effective mass transfer coefficient that combines the mass transfer in the unfouled section of the channel |
| $k_f$     | the mass transfer coefficient in a clean membrane system                                                   |
| $k_{fc}$  | the mass transfer in the cake layer which is a function of the cake layer thickness                        |
| $k_m$     | permeance (L m h/bar)                                                                                      |
| $L$       | fibre length (m)                                                                                           |
| $L_s$     | section length that one can ignore the differential changes along each section                             |
| $\ell$    | module spacing parameter (dimensionless)                                                                   |
| $M_c$     | mass of particle deposited ( $kg$ )                                                                        |
| $N$       | number of sections that a fibre has been divided                                                           |
| $p$       | pressure in lumen side (kPa)                                                                               |
| $P_m$     | maximum TMP for a chosen set of conditions (kPa)                                                           |
| $p_n$     | pressure in the lumen side at section $n$ (kPa)                                                            |
| $P_0$     | maximum operating pressure (kPa)                                                                           |
| $P_p$     | pump driving (inlet) pressure with respect to the ambient pressure (kPa)                                   |
| $q$       | pressure in shell side (kPa)                                                                               |
| $q_n$     | pressure in the shell side at section $n$ (kPa)                                                            |
| $Q$       | flux in 2D direct-flow channel (dimensionless)                                                             |

|                 |                                                                                                                                 |
|-----------------|---------------------------------------------------------------------------------------------------------------------------------|
| $\dot{Q}$       | inlet flow: $\dot{Q} = 2\pi a L J_{av} = \pi a^2 w_0$ (m <sup>3</sup> /s)                                                       |
| $\dot{Q}_n$     | volumetric flowrate at the lumen inlet of section $n$ (m <sup>3</sup> /s)                                                       |
| $\dot{Q}_{n+1}$ | volumetric flowrate at the lumen outlet of section $n$ (m <sup>3</sup> /s)                                                      |
| $r$             | distance along radial direction in each hollow fibre                                                                            |
| $rf$            | ratio of the flux effect contributes to the overall effects                                                                     |
| $R$             | filtration resistance including membrane original resistance and the resistance accumulated during operation (m <sup>-1</sup> ) |
| $R_{ads}$       | resistance due to surface or pore adsorption (m <sup>-1</sup> )                                                                 |
| $R_c$           | resistance caused by fouling (m <sup>-1</sup> )                                                                                 |
| $Re$            | Reynolds number                                                                                                                 |
| $R_f$           | $= R_{ads} + R_{rev} + R_{irrev}$ (m <sup>-1</sup> )                                                                            |
| $R_{irrev}$     | resistance added by irreversible component (m <sup>-1</sup> )                                                                   |
| $R_m$           | membrane resistance (m <sup>-1</sup> )                                                                                          |
| $R_{rev}$       | resistance added due to reversible component (m <sup>-1</sup> )                                                                 |
| $S_p$           | module spacing parameter ( $\ell$ in 2D and $\gamma$ in 3D) (dimensionless)                                                     |
| $t$             | time (day)                                                                                                                      |
| $t_c$           | backwash cleaning cycle time (mins)                                                                                             |
| TMP             | transmembrane pressure difference (kPa)                                                                                         |
| $u$             | velocity in $r$ direction (m/s)                                                                                                 |
| $\dot{V}_c$ (%) | cumulated filtration volume percentage                                                                                          |

|                      |                                                                             |
|----------------------|-----------------------------------------------------------------------------|
| $\Delta\dot{V}_c(z)$ | cumulative flowrate of permeate at axial position $z$ (m <sup>3</sup> /s)   |
| $\dot{V}_n$          | volumetric flowrate at the shell inlet of section $n$ (m <sup>3</sup> /s)   |
| $\dot{V}_{n+1}$      | volumetric flowrate at the shell outlet of section $n$ (m <sup>3</sup> /s)  |
| $V_h$                | amount of feed fluid been processed per unit area (L/m <sup>2</sup> )       |
| $w$                  | velocity in $z$ direction (m/s)                                             |
| $z$                  | distance along fibre length direction (m)                                   |
|                      |                                                                             |
| <b>Greek letters</b> |                                                                             |
| $\alpha$             | normalized module productivity in dimensionless 3D pipe model               |
| $\beta$              | normalized module productivity in realistic hollow fibre modules            |
| $\gamma$             | shell side area to lumen side area ratio: $\gamma = (c^2 - b^2)/a^2$        |
| $\Gamma$             | fibre wall thickness (mm)                                                   |
| $\delta$             | fibre inside radius to fibre length ratio: $\delta = a/L$                   |
| $\epsilon$           | module void fraction: $\epsilon = (c^2 - b^2)/c^2$                          |
| $\mu$                | fluid viscosity (Pa s)                                                      |
| $\Delta\Pi$          | osmotic pressure difference (kPa)                                           |
| $\Pi_m$              | the osmotic pressure at the interface between membrane and suspension (kPa) |
| $\rho$               | fluid density (kg/m <sup>3</sup> )                                          |

|           |                                                               |
|-----------|---------------------------------------------------------------|
| $\rho_c$  | cake density (kg/m <sup>3</sup> )                             |
| $\varphi$ | filtration uniformity parameter                               |
| $\phi$    | module packing density: $\phi = \frac{b^2}{c^2} \times 100\%$ |
| $\Phi$    | porosity of the cake accumulated                              |

# Abbreviations

|             |                                |
|-------------|--------------------------------|
| <b>CEB</b>  | chemical enhanced backwash     |
| <b>CEOP</b> | cake enhanced osmotic pressure |
| <b>CF</b>   | constant flux                  |
| <b>CIP</b>  | cleaning in place              |
| <b>CP</b>   | constant pressure              |
| <b>FO</b>   | forward osmosis                |
| <b>HFM</b>  | hollow fibre membrane          |
| <b>MBRs</b> | membrane reactors              |
| <b>MF</b>   | microfiltration                |
| <b>NF</b>   | nanofiltration                 |
| <b>PA</b>   | polyamide                      |
| <b>PC</b>   | polycarbonate                  |
| <b>PDI</b>  | pressure driven inside feed    |
| <b>PDO</b>  | pressure driven outside feed   |
| <b>PE</b>   | polyethylene                   |
| <b>PEEK</b> | polyetheretherketone           |
| <b>PEI</b>  | poly (ether imide)             |
| <b>PES</b>  | polyethersulfone               |

|             |                                   |
|-------------|-----------------------------------|
| <b>PI</b>   | polyimide                         |
| <b>PP</b>   | polypropylene                     |
| <b>PSF</b>  | polysulfone                       |
| <b>PTFE</b> | polytetrafluoroethylene           |
| <b>PVDF</b> | polyvinylidene difluoride         |
| <b>RO</b>   | reverse osmosis                   |
| <b>TMP</b>  | transmembrane pressure difference |
| <b>UF</b>   | ultrafiltration                   |

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