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Better Material Properties and Faster Catalyzed Chemical Recycling for Poly(*L*-Lactide) Using a Simple Commercial Glycerol Ethoxylate Additive

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ABSTRACT

Poly(*L*-lactide) (PLLA) is the largest volume commercial bio-derived plastic, but its brittleness and end-of-life recycling remain challenges. Here, glycerol ethoxylate (GEO), a branched ethylene glycol derivative, both toughens commercial PLLA and accelerates its catalyzed chemical recycling to *L*-lactide. A series of GEO–PLLA blends, containing 2–20 wt.% GEO, show significantly improved ductility and toughening compared to pure PLLA. The lead 10 wt.% GEO–PLLA sample achieves 9x higher elongation at break (191% ± 4%) and 6x higher tensile toughness (57.9 ± 1.9 MJ m^{-3}), while retaining desirable tensile strength (36.3 ± 1.5 MPa), thermal properties ($T_g = 39°C$ and $T_m = 149°C$) and crystallinity (25%). The GEO–PLLA samples are efficiently chemically recycled to *L*-lactide, showing both high recycling activity (TOF = 2240 ± 73 h^{-1}) and quantitative selectivity for *L*-lactide (> 99%). The recycling is performed neat, at 180°C, using low loadings of commercial Sn(II)Oct_2 catalyst. The 10 wt.% GEO–PLLA sample shows significantly faster chemical recycling than PLLA, with $k_{obs} = 22.9 \pm 0.8\ h^{-1}$ versus $k_{obs} = 1.8 \pm 0.2\ h^{-1}$ for PLLA. This recycling process is successful even with contamination from other commercial plastics, demonstrating its applicability to future postconsumer waste streams.

1 | Introduction

Poly(*L*-lactide) (PLLA) is a semi-crystalline, bio-based, and compostable polymer which is sourced from high-starch content biomass, for example, corn or sugar cane [1]. It is already commercialized, with > 900,000 tons (*t*) produced in 2024, and production volumes expected to exceed 2 Mt/annum by 2029 [2]. It shows useful properties including high optical transparency, high tensile strength, moderate/high melting temperature, and sufficient thermal stability; after use it can be industrially composted [2–4]. Its properties enable it to replace current plastics, like polystyrene (PS) or polyolefins, in applications such as food packaging, textiles, and agriculture [4, 5]. Nonetheless, improvements to its material properties and recyclability are still needed [6].

PLLA has a high strength and stiffness but a low ductility, hence it is brittle and requires toughening [5]. Many promising approaches and additives have been developed to toughen and plasticize PLLA and, among the commercial additives, oligo- or polyethers are particularly promising [3, 7–11]. Early reports described PLLA blended with low quantities (1–20 wt.%) polyethylene glycol (PEG) which resulted in better ductility but compromised the glass transition temperature, T_g, and crystallinity, particularly on sample ageing, and severely reduced the overall tensile strength [12–15]. Bates, Ellison, and coworkers developed PLLA blends toughened with just 5 wt.% poly(ethene oxide-*b*-butene oxide) (PEO-*b*-PBO), which showed outstanding ductility without significant compromise to thermal properties or tensile strengths; very recent reports suggest these

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additives may accelerate aqueous hydrolysis, which could be relevant to environmental degradation [5, 7–10]. Given these excellent material property improvements, other low-cost, low-toxicity, and commercial oligoethers should be explored for PLLA toughening. Another challenge is that it is unclear how such additives would affect any future PLLA recycling, particularly its chemical recycling to monomer, which could be a very useful route to preserve both economic and material value.

PLLA is a thermoplastic, so it is mechanically recyclable, but operating such recycling processes repeatedly is very challenging due to the narrow gap between its melting temperature (T_m = 160°C–200°C) and the high temperature onset of thermal degradation (200°C–250°C) [16]. Under typical high-temperature mechanical recycling conditions, PLLA undergoes accelerated hydrolysis, a process which is autocatalytic, which is a further complication [17]. Efficient PLLA chemical recycling may solve some of these difficulties and several different recycling approaches are proposed in the academic literature, mostly focused on its hydrolysis to lactic acid or its alcoholysis to alkyl lactates [18]. One detraction of these reactions is that they do not directly produce the monomer, that is, further chemical transformations and energy input are required to reproduce *L*-lactide which is needed for commercial PLLA production [18]. It has been known for some time that PLLA can be depolymerized to *L*-lactide, either at elevated temperatures or in dilute solutions, but fast, selective, and industrially applicable processes for chemical recycling to monomer are still needed [6, 19–22]. One advantage of developing chemical recycling processes directly to *L*-lactide is that remanufacturing delivers PLLA with equivalent properties to virgin PLLA [21, 23–29]. Previously, we reported a commercial Sn(II)Oct_2 catalyst that efficiently depolymerizes commercial PLLA, in the neat polymer melt at 160°C and at low catalyst loadings (1:10,000), resulting in both high *L*-lactide conversions (> 90%) and very high enantio-selectivity (> 99%) [21]. Also, we discovered that when excess glycerol ethoxylate (GEO) is added to the catalysis (vs. Sn(II)), the recycling occurred significantly faster, and equivalently selectively. It is proposed that GEO reacts with PLLA by transesterification to increase the concentration of active hydroxyl chain end groups, and then chains undergo chemical recycling to extrude *L*-lactide by a chain-end-catalyzed mechanism [21]. GEO is already commercially produced and widely used in formulations; it is a tri-hydroxyl end-capped oligo-ether, that is, a 3-arm star analogue of the well-known oligo-/poly(ethylene glycols) (PEG). GEO is manufactured from commodity chemicals ethene oxide and glycerol, and has very low toxicity (GEO LD_{50} > 5000 mg/kg) [30]. In addition, it is highly soluble in solvents compatible with PLLA, for example, THF and DCM, which allows for the formation of homogenous GEO–PLLA films; many other common multifunctional alcohols, for example, pentaerythritol, show poor solubility in these solvents. As such, GEO could be an attractive PLLA additive delivering both tougher samples and materials that are pre-primed for faster catalyzed chemical recycling to monomer. This latter point is important, since the addition of the Sn(II) catalyst is essential for any PLLA chemical recycling to *L*-lactide. Thus, GEO addition to PLLA should not compromise its thermal stability or processability.

2 | Results and Discussion

To test our hypothesis, a series of GEO–PLLA blend samples were prepared comprising commercial, high molar mass PLLA mixed with low weight percentages (2–20 wt.%) of GEO. The samples thermal and mechanical properties are compared to neat PLLA that was processed and tested equivalently. The commercial PLLA should have sufficiently high molecular weight to match the majority of its applications as a plastic; here, PLLA was supplied by Goodfellows (ME34-GL-000110) and has M_n = 60 kg mol^{-1}, $Đ$ = 1.81. The GEO was supplied by Sigma Aldrich and has M_n ~1000 g mol^{-1}. A further control sample was prepared by mixing the same PLLA with 10 wt.% PEG (M_n ~ 1000 g mol^{-1}) so as to probe influences of oligoether architecture on toughening. The series of PLLA samples feature systematically increasing GEO contents: 2, 5, 7.5, 10, 15, and 20 wt.%. The blended samples were prepared by mixing GEO and PLLA in solution (minimum amount of methylene chloride), followed by film formation and drying at 60°C, under vacuum, for 2 days to produce colourless films of around 0.5 mm thickness. All films look homogenous without any air bubbles and the complete removal of the processing solvent was indicated by NMR spectroscopy and thermal gravimetric analysis (TGA).

Differential scanning calorimetry (DSC) was used to characterise the PLLA samples; in all cases materials are semi-crystalline showing both glass transitions (T_g) and melting transitions (T_m). The 2–15 wt.% GEO–PLLLA samples all show a single glass transition temperature, T_g at 65°C–34°C, respectively; for 20 wt.% GEO–PLLA, a second T_g at −14°C was observed (Figures 1a and S15, Tables 1 and S1). This finding indicates that PLLA and GEO are miscible from 2–15 wt.%, which is further supported by the close agreement of the measured and theoretical T_g values, the latter determined using the Fox equation (Figure 1b). As any additives should be miscible with PLLA to improve its toughness, the miscible 2–15 wt.% GEO–PLLA samples were selected for further investigations. For these samples, the PLLA T_g value decreases with increasing GEO content, from 65°C for pure PLLA to 34°C for the 15 wt.% GEO–PLLA. All samples are semi-crystalline, with PLLA T_m = 148°C–150°C and all show moderate/good high temperature stability, with the onset of high temperature decomposition ($T_{d,5\%}$) between 240°C–300°C (Table 1 and Figure S17). The high temperature stability is slightly lower at higher GEO contents, consistent with prior reports on PEG–PLLA blends and likely due to increased chain flexibility [31]. This hypothesis supports the notion that GEO may serve as both an impact modifier and recycling accelerator. Comparing the 10 wt.% GEO–PLLA sample with a 10 wt.% PEG–PLLA sample, at equivalent oligoether molar mass, shows both samples have equivalent T_g = 40°C. Based upon the thermal properties, PLLA samples containing 2–10 wt.% GEO are selected as the lead candidates, all samples show T_g values of ≥ 39°C and thermal decomposition onset temperatures of ≥ 249°C.

Next, uniaxial tensile extension experiments were performed using these 2–10 wt.% GEO–PLLA samples at a strain rate of 10 mm min^{-1}. Dumbbell-shaped specimens were prepared from the polymer films according to ISO 527–2, specimen type 5B, using a cutting press. The tensile strength, elongation at break,

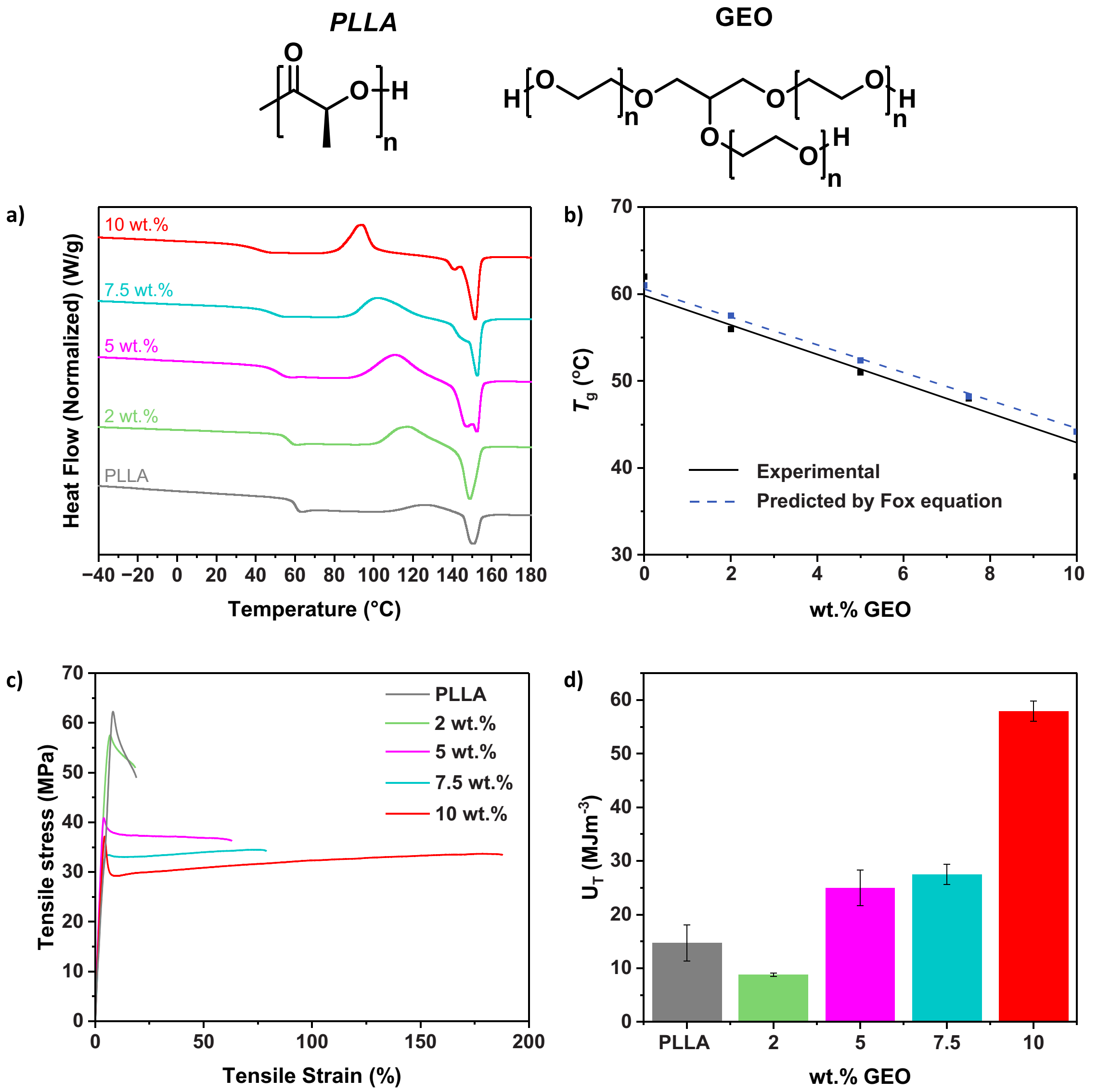

FIGURE 1 | Thermal and tensile mechanical data for the 2–10 wt.% GEO–PLLA samples. (a) DSC thermograms for 2-10 wt.% GEO–PLLA samples (second heating cycle). (b) Plot of sample T_g values (DSC) vs. the GEO weight fraction. The experimental data (black squares) are compared with $T_{g,\text{theo}}$ values (blue squares, dashed line) determined using the Fox equation. (c) Bar chart of tensile toughness values vs. wt.% GEO in the PLLA. (d) Representative stress–strain data for the samples (10 mm min^{-1}).

and toughness measurements were repeated five times for each sample, using separate specimens, with the mean value reported, along with the standard deviation representing the error (Table 1). All the GEO–PLLA blends displayed significantly higher ductility and toughness than PLLA. The lead sample 10 wt.% GEO–PLLA showed > 100% elongation at break and toughness values of up to 57.9 ± 1.9 MJ m^{-3} (Figure 1c,d). These mechanical properties are comparable to other widely used plastics, for example, polypropylene (σ = 9–80 MPa, average = 30 MPa, and ε = 2%–900%, average = 160%) [33]. PLLA on its own is very brittle, showing a low toughness of 9.2 ± 1.8 MJ m^{-3}. Even at lower GEO loadings, significant improvements to toughening were feasible, for example, 5 wt.% GEO–PLLA shows an elongation at break of 64% ± 10% and a toughness of 25 ± 3.3 MJ m^{-3}. The 10 wt.% GEO–PLLA shows 9x higher ductility (elongation at break) and 6x greater toughness than pure PLLA. It shows slightly lower tensile strength than PLLA, with σ = 36.3 ± 1.5 MPa, but, importantly, the tensile strength matches or exceeds values for many other commodity plastics [34]. The structure of the oligo-ether additive significantly influenced overall sample mechanical performance, with 10 wt.% GEO–PLLA showing much greater elongation at break (ε = 191% ± 4%) than 10 wt.% PEG–PLLA (ε = 20% ± 2%) (Figure S19). As both the GEO and PEG additives are the same molar mass (M_n ~ 1000 g mol^{-1}), the branched GEO structure is likely responsible for the better PLLA toughening. Previous research has suggested that branched additives may be more effective plasticizers than linear analogues, perhaps because they are more plastic-matrix miscible, due to smaller radii of gyration and increased numbers of chain end groups [35].

As discussed in the introduction, there are quite a few studies of oligoether PLLA plasticizers and significant prior literature of PEG–PLLA blends, but surprisingly no prior investigations into its branched analogue, GEO [36, 37]. The GEO–PLLA

TABLE 1 | GEO–PLLA sample characterization data.

Entry	Blend	T_g (°C)[a]	T_{cc} (°C)[b]	T_m (°C)[c]	χ (%)[d]	$T_{d,5\%}$ (°C)[e]	σ (MPa)[f]	ε_B (%)[g]	U_T (MJ m^{-3})[h]	E' (GPa)[i]
1	2 wt.% GEO	56	116	150	28	297	52.4 ± 3.6	20 ± 1	8.8 ± 0.3	3.20
2	5 wt.% GEO	51	110	149	35	287	43.7 ± 3.8	64 ± 10	25.0 ± 3.3	2.29
3	7.5 wt.% GEO	48	101	150	29	267	33.7 ± 1.0	85 ± 7	27.5 ± 1.9	2.59
4	10 wt.% GEO	39	93	149	25	249	36.3 ± 1.5	191 ± 4	57.9 ± 1.9	2.40
7	PLLA	61	125	149	22	325	60.8 ± 3.0	21 ± 3	9.2 ± 1.8	2.84
8	10 wt.% PEG	40	96	149	31	268	31.4 ± 1.7	20 ± 2	5.4 ± 0.7	2.60

[a] Glass transition temperature determined from the second DSC heating cycle.

[b] Cold crystallization temperature determined from the second DSC heating cycle.

[c] Melt temperature determined from the first DSC heating cycle.

[d] Crystallinity calculated by $(\Delta H_m - \Delta H_{cc})/\Delta H_{m,100 \times 100}$, where $\Delta H_{m,100}$ is the reported melting enthalpy for crystalline PLLA = 93.7 J/g [32]. ΔH_m obtained by integration of the endothermic peak in the first DSC heating cycle. No cold crystallization peak was observed in the first heating cycle, so $\Delta H_{cc} = 0$.

[e] Thermal degradation temperature corresponds to the temperature at 5% mass loss, determined by TGA.

[f] Maximum tensile strength.

[g] Strain at break.

[h] Tensile toughness (area under the stress–strain curve).

[i] Storage modulus, at 25°C, determined by DMA with temperature control.

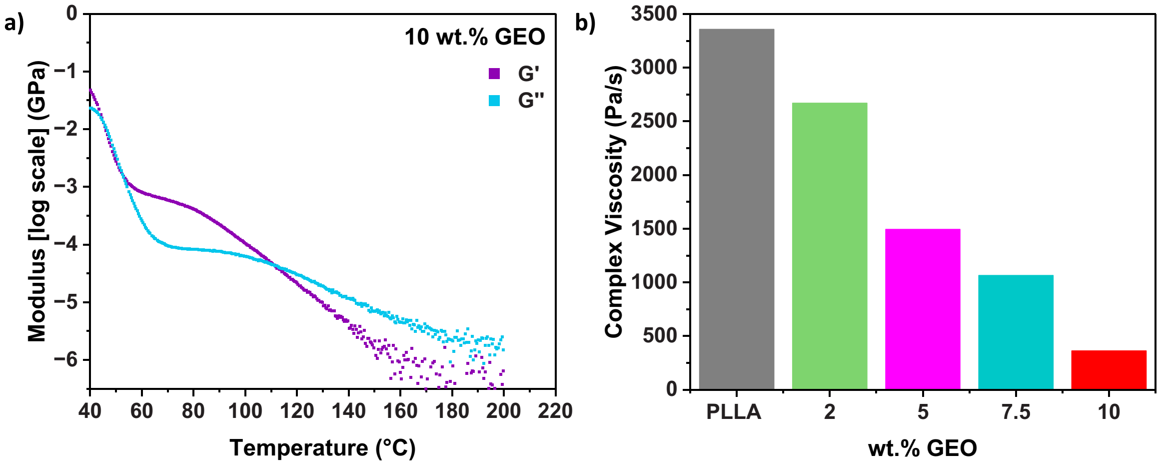

FIGURE 2 | Rheology and DMTA data GEO–PLLA Samples. (a) Rheological storage and loss modulus profiles against temperature for 10 wt.% GEO–PLLA blend. (b) Bar chart showing complex viscosity, measured at 180°C, vs. GEO wt.% in the samples.

samples show better properties than prior PEG–PLA blends, reported by Scandola et al., where the 12.5 wt.% PEG–PLA (PEG M_W = 1500 g mol^{-1}) showed an elongation at break of 194%, but significantly lower tensile strength, σ = 18.5 MPa [38]. Thus, GEO seems to be a more effective toughener than PEG, particularly as it is more effective at lower loadings, helping to retain the desirable high PLLA tensile strength. The mechanical properties of the GEO–PLLA blends are comparable with some of the most effective toughened PLLA samples [12, 39]. For example, PLLA with 10 wt.% poly(butylene adipate terephthalate) (PBAT) achieved an elongation at break of ~150% and maximum tensile stress of up to 55 MPa, while PLLA with 1.8 wt.% PEO-*b*-PBO exhibited an elongation at break of 221% and a maximum tensile stress of 40 MPa [40, 41].

To investigate the high-temperature mechanical and processing properties of GEO–PLLA blends, dynamic mechanical temperature analyses (DMTA) and rheology were used to determine the storage and loss moduli and damping factors (Figure 2). In melt rheology, all samples showed somewhat similar storage and loss moduli from 50°C to 200°C (Figure S20). As the wt.% of GEO, in the PLLA samples, increases, the storage and loss modulus cross-over temperature, signaling the transition to the liquid state, and the sample's complex viscosity at 180°C, decrease. The lead sample featuring 10 wt.% GEO–PLLA shows a cross-over transition temperature of 111°C and a complex viscosity of 363 Pa s (Figure 2). These values are substantially lower than for pure PLLA, where the transition temperature is 136°C and complex viscosity is 3358 Pa s. Thus, lower energy PLLA processing may be feasible for the samples containing GEO, which may benefit some applications. In the DMTA measurements, all the GEO–PLLA samples show high storage moduli, with E′ > 2 GPa at 25°C, and good mechanical stability from −80°C to 50°C (Figure S21). As the GEO content in the PLLA samples increases, so the sample storage modulus falls, which is consistent with the tensile mechanical measurements (Figure 2). The lead 10 wt.%

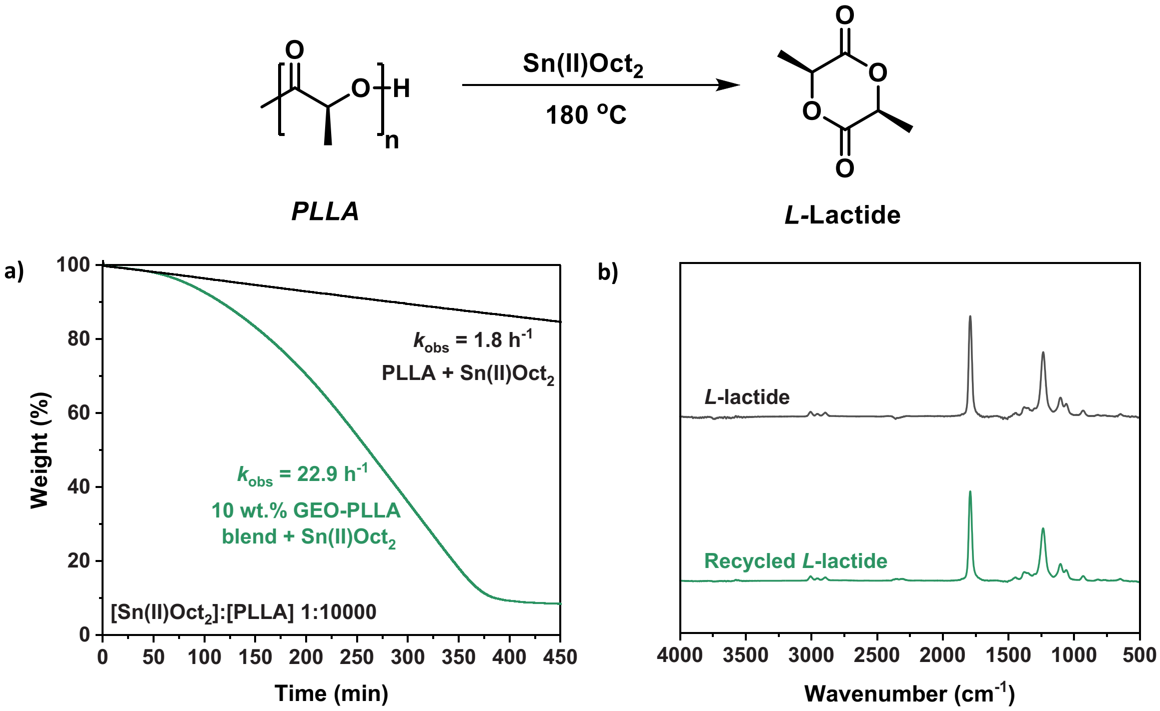

FIGURE 3 | Data showing fast and selective catalyzed chemical recycling of 10 wt.% GEO–PLLA. (a) Plot of PLLA mass loss vs. time for the catalysis using the 10 wt.% GEO–PLLA (green) and PLLA (black) samples. Catalysis conditions: [Sn(II)Oct$_2$]:[PLLA] 1:10,000, neat film, 180°C using a TGA–FTIR instrument. The rate coefficients, k_{obs}, are the gradients of linear fits. (b) The FTIR spectra of the product of the catalyzed chemical recycling, *L*-lactide (green), and the standard spectrum for pure *L*-lactide (black).

GEO–PLLA sample shows E′ = 2.40 GPa, which is sufficiently high for many plastics applications (Table 1).

As mentioned, we recently reported the catalytic chemical recycling of commercial PLLA, at 160°C in the melt, using a commercial Sn(II)Oct$_2$ catalyst, to selectively form *L*-lactide with high conversion and enantioselectivity [21]. We also showed that adding excess GEO to the Sn(II) catalyst, significantly enhances the depolymerization rates, and the recycling can tolerate up to 200 equivalents of GEO (vs. catalyst, i.e., [Sn(II)Oct$_2$]:[GEO]:[PLLA] 1:200:10,000). The depolymerization was proposed to follow a chain-end mechanism, whereby the catalyst active site is a Sn(II) alkoxide species (Scheme S1 illustrates the proposed recycling mechanism). GEO is proposed to accelerate recycling rates by fast, initial PLLA transesterification reactions to form both shorter chains and more hydroxyl chain end-groups, prior to chain end catalyzed depolymerization to *L*-lactide. The GEO, therefore, serves to reduce the overall sample viscosity and increase the hydroxyl end-group concentration; it may also coordinate to the Sn(II) alkoxide active site, thereby increasing its nucleophilicity and reducing the barrier to chain-end backbiting. Here, we discovered that 10 wt.% GEO–PLLA samples showed the best thermal and mechanical performances, so understanding performances in the catalyzed chemical recycling is a priority.

The chemical recycling of the lead 10 wt.% GEO–PLLA was first tested on a small scale, using a methodology previously reported by our team [21]. In brief, samples are analyzed using thermal gravimetric analysis to quantify PLLA mass loss over time (Figures 3 and S28 and S29) [21]. The 10 wt.% GEO–PLLA sample was mixed with the Sn(II)Oct$_2$ catalyst, in minimum amounts of methylene chloride, such that the molar ratios were [Sn(II)Oct$_2$]:[PLLA] 1:10,000. A known mass of the mixture was then added to the TGA crucibles, and the solvent was completely removed under vacuum. In the recycling experiments, the samples were heated to 180°C, and mass loss was analyzed under continuous nitrogen gas flows (25 mL min^{-1}) with the effluent gases passing through an in-line FTIR spectrometer to characterize the recycled product. All samples show a steady decrease in PLLA mass (weight) over time and, in parallel, increasing concentrations of *L*-lactide were detected by FTIR spectroscopy. *L*-Lactide is the only detected product consistent with the very high selectivity reported previously under these recycling conditions. The pseudo-zero-order chemical recycling rate coefficient, k_{obs}, is the gradient of the linear fit to the mass loss versus time data (Figure 3a). After 6 h, under these low Sn(II) catalyst loading conditions, samples showed > 90% mass loss, which corresponds to the complete PLLA depolymerization and k_{obs} = 22.9 ± 0.8 h^{-1}; TOF = 2240 ± 70 h^{-1}. The residual mass content (10 wt.%) corresponds almost exactly to the original amount of GEO added to the PLLA. The chemical recycling rate of the GEO–PLLA blend is > 12x faster than that for pure PLLA, under equivalent conditions, where k_{obs} = 1.8 ± 0.2 h^{-1} and TOF = 180 ± 20 h^{-1}.

To evaluate the chemical recycling at the laboratory scale, the 20 wt.% GEO–PLLA sample was used since it facilitated GEO recovery (isolation of the additive is more challenging at lower loadings). In these experiments, the mixture of [Sn(II)Oct$_2$]:[PLLA] 1:1000 was prepared as before, and the sample was heated to 180°C under vacuum (0.2 mbar).

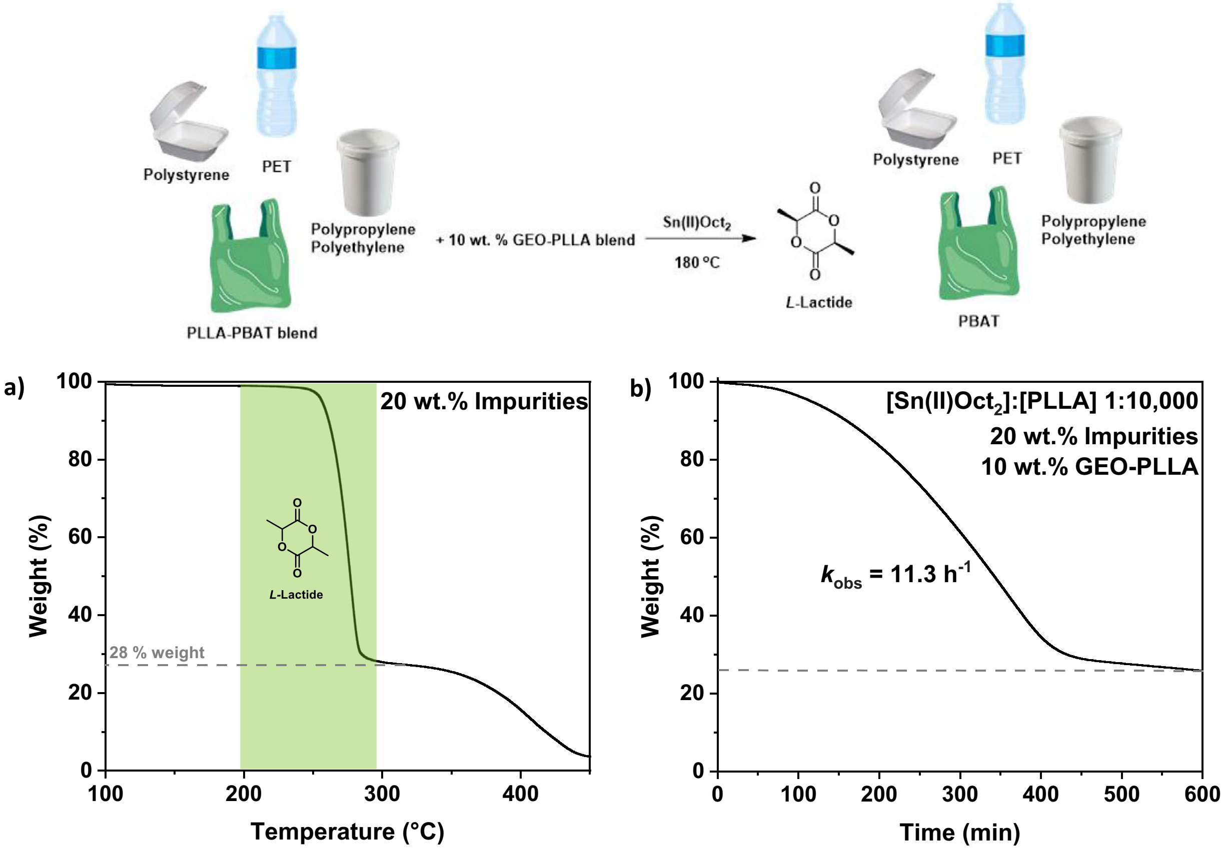

FIGURE 4 | Catalyzed chemical recycling of 10 wt.% GEO–PLLA with a model plastic waste mixture (containing PE, PP, PS, PET, and PBAT). (a) Plot of sample mass loss (wt.%) vs. temperature for sample containing 80:20 wt.% of 10 wt.% GEO–PLLA:waste plastic impurities (overall, the sample contains 1:1:2:3:3 wt.% PE:PP:PS:PET:PBAT-PLLA). The highlighted (green) temperature range corresponds to the range where to only *L*-lactide is detected in the FTIR spectrum of all the decomposition products. The residual weight %, after recycling, corresponds well to the initially added wt.% of other plastic impurities (20 wt.%) + GEO content (8 wt.% for overall sample). (b) Plot of sample mass loss (wt.%) against time for the same sample (80:20 wt.% of 10 wt.% GEO–PLLA:waste plastic impurities) at 180°C. Recycling conditions: [Sn(II)Oct$_2$]:[PLLA] 1:10,000. Final weight % = 26%.

The *L*-lactide product was collected using a sublimation apparatus. Over 6 h, the PLLA underwent nearly complete (93%) conversion to *L*-lactide as the sole product ($> 99\%$); the monomer was characterized by ^{1}H NMR spectroscopy and GC-MS (Figures S24–S26). There was no evidence of any *L*-lactide epimerization, and the GEO was recovered from the recycling residue in 74% yield.

When planning a future chemical recycling process, it would be economically and environmentally preferable to mix the PLLA and catalyst samples without any organic solvents (small-scale experiments use solvents). To demonstrate the feasibility for total solvent-free recycling, the 10 wt.% GEO–PLLA sample was melt mixed with the Sn(II)Oct$_2$ catalyst ([Sn(II)Oct$_2$]:[PLLA] 1:740). As such, the PLLA and catalyst were heated in the polymer melt at 160°C, under nitrogen, with stirring for 15 min. After cooling the mixture to room temperature, the sample was subjected to the same chemical recycling conditions described previously (TGA–FTIR apparatus, 180°C, N_2 = 25 mL min^{-1}). Over 4 h complete PLLA depolymerization occurred, with $k_{obs} = 32.9 \pm 3$ h^{-1} and TOF = 241 ± 30 h^{-1} (Figure S29). The recycling rates are slower than those obtained using solvent-mixed samples, but future larger-scale optimization of the melt mixing process should improve rates.

Another key aspect for a new chemical recycling process is the tolerance of the catalysis to other plastic contaminants. Future post-consumer PLLA waste streams are likely to have small amounts of contamination by other common plastics and their additives (plasticizers, colorants, etc). Here, proof-of-concept contaminant effects were investigated by blending the GEO–PLLA samples with a model plastic waste mixture; the mixture comprised known quantities of plastic wastes including a poly(ethylene terephthalate) (PET) bottle, expanded PS, polypropylene (PP) and polyethylene (PE) packaging, and a compostable PBAT–PLLA plastic shopping bag (commonly used in UK supermarkets). To test the catalysis under demanding conditions, the waste plastics were combined and mixed with the GEO–PLLA so as to produce samples containing 5, 20, or 50 wt.% of the contaminant mixture (i.e., of impurities). Due to differences in solubility between the various plastics, and to better mimic mixed post-consumer plastic waste treatments, the samples were prepared by physically mixing shredded plastic flakes with the GEO–PLLA blend. Accordingly, the PET bottle, expanded PS, PP and PE packaging, and PBAT–PLLA bag were first shredded individually into small flakes. The flakes of the different plastics were then combined to make up the model impurity sample comprising of 10:10:20:30:30 wt.% of

PE:PP:PS:PET:PBAT–PLLA. Meanwhile, to ensure an accurate catalyst loading, a film containing the Sn(II)Oct_2 and 10 wt.% GEO–PLLA blend was prepared ([Sn(II)Oct_2]:[PLLA] 1:10,000), shredded and combined with known amounts of the model impurity mixture. The combined samples, each with known wt.% of plastic contamination, were then chemically recycled, at 180°C, and the recycling was monitored using TGA–FTIR spectroscopy. In all cases, including with a sample containing 50 wt.% plastic impurities, the PLLA content of the mixture was completely depolymerized within 10 h. Any residual weight, after the recycling, corresponded to the original quantity of waste plastic impurites and GEO added to the samples (Figures 4b and S34). The depolymerization rates for these contaminated PLLA samples are slower than those for the pure GEO–PLLA samples, with $k_{obs} = 11.3\ h^{-1}$ for 20 wt.% impurities versus $k_{obs} = 22.9\ h^{-1}$ for the pure GEO–PLLA sample. The rate reductions likely arise from both catalyst dilution and mass transfer limitations. To understand the catalytic temperature range and selectivity, samples containing 10 wt.% GEO–PLLA with different amounts of impurities, at [Sn(II)Oct_2]:[PLLA] 1:10,000, were subjected to dynamic heating experiments and the depolymerization products analyzed by FTIR spectroscopy. From 150°C to 300°C, only the PLLA was recycled and the sole product was *L*-lactide. At > 300°C, other plastic decomposition processes begin. Thus, it is quite feasible to selectively depolymerize PLLA from the mixture; this finding might even be developed into a waste plastic mixture reactive separation process in future (Figures 4a and S30–S32).

Most PLLA used in packaging is part of a formulation (blend) with PBAT, as such the chemical recycling of 10 wt.% GEO–PLLA mixed with PBAT–PLLA was investigated. A 20:80 wt.% mixture of PBAT–PLLA:10 wt.% GEO–PLLA was tested ([Sn(II)Oct_2]:[PLLA] 1:740, 180°C). Importantly, the PLLA recycling rate and selectivity were maintained. Comparing the PLLA recycling rates shows $k_{obs} = 31.7\ h^{-1}$ for the sample contaminated by PBAT compared with $k_{obs} = 32.9\ h^{-1}$ for GEO–PLLA, under equivalent conditions. The sole recycling product was *L*-lactide and ^1H NMR spectroscopy of the reaction residue showed both PBAT and GEO (Figure S36). This result demonstrates that complete PLLA conversion, including the PLLA in the original PBAT–PLLA blend, was achieved. It also indicates that any transesterification of the PBAT was either much slower than that of the PLLA, and/or the transesterification resulted in non-volatile products. Overall, these proof-of-concept recycling studies show that toughened GEO–PLLA samples undergo fast and selective chemical recycling to *L*-lactide, even when the starting plastic is contaminated, as would be expected in real waste streams.

3 | Conclusions

A simple, commercial glycerol ethoxylate (GEO) blended with poly(*L*-lactide) (PLLA), at 2–10 wt.% loadings, simultaneously improved material ductility and toughness, as well as significantly accelerating chemical recycling to the true monomer, *L*-lactide. The GEO–PLLA blend samples are suitable for conventional high–temperature plastic processing and show similar crystallinity and thermal properties to pure PLLA. The samples have greater ductility and tensile toughness, thereby overcoming the well-known brittleness of PLLA. The lead 10 wt.% GEO–PLLA sample showed 9x greater elongation at break and 6x higher tensile toughness than PLLA; it has a T_g value of 39°C and melting temperature was 149°C, which are only slightly lower than for PLLA. The same GEO–PLLA blends showed significantly (> 12x) faster rates of catalyzed chemical recycling than PLLA on its own and quantitative selectivity for *L*-lactide (>99%). The recycling process, conducted at low catalyst loading and in the neat PLLA melt, tolerated contamination by other common waste plastics and even other polyesters. Using contaminated waste plastic samples, the selective recycling of PLLA to *L*-Lactide occurred from 150°C–300°C. The GEO additive is already a commercial product, synthesized from commodity chemicals, glycerol, and ethylene oxide, both of which can also be bio-derived at large scale [42, 43]. It has very low toxicity and is already used in formulations. Therefore, it shows real potential as a beneficial additive to improve both PLLA performance and end-of-life recycling. In future, this new concept of a single additive to tackle both material property and recycling performances should be more broadly explored, including through larger-scale material engineering and application development.

4 | Experimental Section

All air-sensitive and moisture-sensitive experiments were carried out under N_2, using standard Schlenk/glovebox techniques unless otherwise stated. Tin(II) 2-ethylhexanoate (Sn(II)Oct_2) was purchased from Sigma Aldrich and used as received. PLLA was purchased from GoodFellows (ME346310, M_n = 60,000 g mol^{-1} in $CHCl_3$). GEO (GEO, M_n ~ 1000 g mol^{-1}) was purchased from Sigma–Aldrich, dried at 120°C under high vacuum (1×10^{-2} mbar), and stored over 3 Å sieves, for 2 days prior to use. PEG (M_n ~ 1000 g mol^{-1}) was purchased from Sigma–Aldrich and dried over P_2O_5 under high vacuum (1×10^{-2} mbar), prior to use. The poly(*L*-lactide)-poly(butylene adipate terephthalate) (PLLA–PBAT) blend sample used was a kitchen caddy liner from a U.K. supermarket. PET was obtained from a postconsumer plastic water bottle. PE, PP, and PS were obtained from postconsumer packaging. Anhydrous dichloromethane was purchased from Sigma–Aldrich and degassed, by N_2 purge, prior to use.

Size exclusion chromatography (SEC) was carried out on a Shimadzu LC-20AD instrument, using two PSS SDV linear M columns in series, with a $CHCl_3$ eluent. Measurements were conducted at 30°C, with a flow rate of 1 mL/min. Samples were detected with a differential refractive index (RI) detector. Number-average molar mass ($M_{n,SEC}$) and dispersities, ($Đ = M_w/M_n$)) were calculated by calibration against a series of well-defined PS standards showing molar masses from 500 to 1,000,000 g mol^{-1}. The polymer samples were dissolved in HPLC-grade $CHCl_3$, at a concentration of *ca* 10 mg/mL, and filtered through a 0.2 µm microfilter prior to analysis.

DSC was performed using a TA discovery 25 auto. Experiments were performed under N_2 flow (50 mL/min), using aluminum TZERO pans. Samples (2–5 mg) were equilibrated at 40°C, then heated at a rate of 10°C/min to 200°C, and held at 200°C, for 2 min. The sample was then cooled, at a rate of 10°C/min, to −80°C and held at −80°C for 2 min. The sample was then heated, at a rate of 10°C/min, to 200°C and cooled, at a rate 10°C/min, to

−80°C for three successive cycles. Thermal data is reported from the second heating cycle.

Tensile testing was carried out using an Instron 8600 series universal testing system, using a 500 N load cell and 250 N pneumatic grips. Dumbbell-shaped specimens were cut using a Zwick ZCP020 cutting press, equipped with a cutting device for ISO 527-2 type 5B. Uniaxial extension experiments (10 mm min^{-1} cross-head speed) were run according to ISO 527.

Rheological experiments were conducted in the melt on an ARES-G2 (TA Instruments) using polymer samples between 8 mm stainless steel plates. The samples were subjected to a temperature sweep from 200°C to 40°C at a heating rate of 3°C min^{-1}, 1 Hz frequency. The shear storage (G') and loss (G'') moduli were monitored. The complex viscosity was measured at 180°C.

Dynamic mechanical thermal analysis (DMTA) was carried out using a DMA850 (TA Instruments), using an ACS III cooling system. Specimens of uniform width (5.3 mm) were cut using two parallel blades. Samples were heated from −80 to 250°C (or until the material deformed beyond the limits of the geometry employed), at a rate of 3°C min^{-1}, with a frequency of 1 Hz, 0.1 N preload force and 0.1% strain amplitude.

Thermal gravimetric analysis (TGA) data was collected on a TGA5500 System (TA Instruments), equipped with the TRIOS software package. Detailed procedures are given in the protocol section.

^1H and $^{13}C\{^1H\}$ NMR spectra were obtained using a Bruker AVIII HD nanobay NMR spectrometer. Coupling constants are given in Hertz.

GC–MS spectra were recorded on an Agilent 7820A equipped with a HP5-MS ultra inert column (30 m length, 0.25 mm internal diameter, 0.25 µm film thickness), a 5977B single quad mass spectrometer, a liquid injection autosampler, and He carrier gas. Data was processed using MassHunter software. Samples of 5 mg/mL were prepared in dichloromethane with 1 µL injected. Samples were loaded on to the column in 1:100 sample:solvent splitter ratio and injection port temperature of 300°C. The column was pressurized at 9.1 PSI, with a column flow of 1.2 mL/min and total flow of 22.12 mL/min. Following equilibration of the column oven at 40°C for 1 min, the temperature was ramped from 40°C to 300°C at a rate of 10°C/min and held at 300°C for 3 min. The MS source and quadrupole temperature was 230°C and 150°C, respectively.

Turnover frequency (TOF) calculations were performed using mass loss against time plots from 20%–80% mass loss of the PLLA over time.

4.1 | Preparation of Blends

Polymer films were prepared by solvent casting high molecular weight commercial PLLA (M_n = 60,000 g mol^{-1}) with various wt.% of GEO (M_n = 1000 kg mol^{-1}). The PLLA was dissolved in CH_2Cl_2, (1 g per 10 mL), and stirred at room temperature until dissolved. Various wt.% of GEO was added to the dissolved PLLA and the polymer and GEO solution were transferred into a PTFE mould. The PTFE mould was covered by a punctured aluminum foil and left to evaporate, at room temperature, overnight. The polymer film was then dried, in a vacuum oven, at 60°C for 2 days. The absence of any residual solvent was checked using DSC and TGA.

4.2 | Chemical Recycling of GEO–PLLA Blends by $Sn(II)Oct_2$

In the glovebox, stock solutions of the different GEO–PLLA blends (1.00 M, 72 mg of PLLA in 1.00 mL of CH_2Cl_2) and $Sn(II)Oct_2$ (1.00×10^{-3} M, CH_2Cl_2) were prepared. The GEO–PLLA blend stock solution (100 µL, 0.1 mmol, 10000.0 equiv) was added to a vial containing $Sn(II)Oct_2$ (10.0 µL, 1.00×10^{-5} mmol, 1.0 equiv). The $Sn(II)Oct_2$-GEO-PLLA solution was thoroughly mixed before being dropcast (*ca* 3 drops) onto Platinum TGA crucibles. The solvent was allowed to evaporate before the crucible was loaded into the TGA for monitored solid-state depolymerization. The following TGA method template was used for the reaction:

1. N_2 flow of 25.0 mL min^{-1}.
2. Heat from room temperature to 180°C.
3. Maintain the reaction isotherm at 180°C for 300 min (or until PLLA depolymerization is complete).
4. Heat, at 20°C min^{-1}, to 600°C.
5. Cool to 30°C.

The catalyst loading, GEO–PLLA blend, and length of the isotherm were varied as required. To account for residual solvent loss from the polymer films, which will also be detected as a mass loss in TGA, data from the first 2.0 min of the run were removed from the analysis. The mass at 2.0 min was then taken as the polymer/catalyst initial mass and used to calculate the change in weight % of the sample. The residual mass of the GEO was accounted for as follows:

1. Take the mass at time = 2.0 min as the initial PLLA, GEO, and catalyst mass.
2. Subtract the theoretical %mass of GEO in the run: for example, for the 10 wt.% GEO–PLLA blend this would be 10%. Therefore, the mass of initial polymer/catalyst film = 90% of mass at 2.0 min.
3. Take the mass in step (2) as the initial mass for calculation of the change in mass % of the sample.

4.3 | Lab-Scale Chemical Recycling of GEO–PLLA Blends by $Sn(II)Oct_2$

In the glovebox, a stock solution of $Sn(II)Oct_2$ (1.00×10^{-1} M, 40.7 mg of $Sn(II)Oct_2$ in 1.00 mL of CH_2Cl_2) was prepared. The $Sn(II)Oct_2$ (54.4 µL, 5.44×10^{-3} mmol, 1.0 equiv) was added to a Schlenk containing the 20 wt.% GEO–PLLA blend (0.49 g, 5.44 mmol, 1000.0 equiv). The GEO–PLLA blend and $Sn(II)Oct_2$ mixture was dissolved in a minimum amount of DCM (approx.

1 mL) before the solvent was removed in vacuo. A water-cooled cold-finger was connected to the Schlenk, and the reaction mixture was heated to 180°C with magnetic stirring under vacuum (0.1–1 mbar). After 6 h, a white crystalline powder was collected, which was determined to be lactide (0.34 g, 2.36 mmol, 87% yield, 95% *L*-LA as determined by ^{1}H NMR spectroscopy and GC-MS).

4.4 | Solvent-Free Chemical Recycling of PLLA

The 10 wt.% GEO–PLLA sample (1.22 g) was added to a 50 mL round-bottom flask and the total mass recorded. Several drops of $Sn(II)Oct_2$ catalyst were added to the flask, and the catalyst loading was calculated from the change in mass (8.29 mg, $[Sn(II)Oct_2]$:[PLLA] 1:743). A stirrer bar was added, and the round-bottom flask was heated to 180°C, above the PLLA melt temperature, with stirring for 15 min. After leaving the flask to cool to room temperature, a sample of the solidified mixture was loaded onto a TGA crucible. The depolymerization was performed at 180°C, under an N_2 flow of 25 mL min^{-1}, and monitored by TGA–IR spectroscopy.

4.5 | Chemical Recycling of GEO–PLLA Samples Contaminated by Impurities

In the glovebox, a stock solution of $Sn(II)Oct_2$ (1.00×10^{-2} M, 12.2 mg of $Sn(II)Oct_2$ in 3.00 mL of THF) was prepared. The $Sn(II)Oct_2$ stock solution (68.1 µL, 6.81×10^{-4} mmol ,1.0 equiv) was added to a vial containing 10 wt.% GEO–PLLA (0.55 g, 6.81 mmol, 10,000 equiv) and the mixture was dissolved in the minimal amount of CH_2Cl_2 (~5 mL). The solvent was removed in vacuo to obtain a film of the 10 wt.% GEO–PLLA and $Sn(II)Oct_2$ mixture. The film was removed from the glovebox and shredded into small flakes.

Samples of PE, PP, PS, PET, and a poly(lactide)-poly(butylene adipate terephthalate) (PLA–PBAT) blend were prepared by shredding each plastic into small flakes. A model plastic impurity mixture was prepared by combining the PE (1 mg, 10 wt.%), PP (1 mg, 10 wt.%), PS (2 mg, 20 wt.%), PET (3 mg, 30 wt.%), and PLA–PBAT blend (3 mg, 30 wt.%) flakes. Flakes of the 10 wt.% GEO–PLLA blend was then combined with varying amounts of the impurity mixture and loaded into TGA crucibles. The depolymerizations were performed at 180°C, under an N_2 flow of 25 mL min^{-1}, and monitored by TGA–IR spectroscopy.

4.6 | Chemical Recycling of GEO–PLLA Samples Contaminated by PLA–PBAT

The 10 wt.% GEO–PLLA and $Sn(II)Oct_2$ sample prepared in the solvent-free chemical recycling (4 mg, $[Sn(II)Oct_2]$:[PLLA] 1:743) was combined with flakes of the PLA–PBAT blend (1 mg) to make up a mixture containing 20 wt.% of the PLA–PBAT blend and 80 wt.% of the 10 wt.% GEO–PLLA blend. The mixture was loaded into a TGA crucible, and the depolymerization was performed at 180°C, under an N_2 flow of 25 mL min^{-1}. The TGA crucible residue post depolymerization was analyzed by ^{1}H NMR spectroscopy and SEC.

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Conflicts of Interest

The authors declare no conflicts of interest.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

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