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# Zinc complexes for PLA formation and chemical recycling

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#### Zinc Complexes for PLA Formation and Chemical Recycling: Towards a Circular Economy

Paul McKeown,<sup>[a], §</sup> Luis A. Román-Ramírez,<sup>[b], §</sup> Samuel Bates,<sup>[a]</sup> Joseph Wood,<sup>\*, [b]</sup> and Matthew D. Jones<sup>\*,[a]</sup>

Abstract: A series of Zn(II) complexes, based on propylenediamine Schiff bases, have been prepared and fully characterized. X-ray crystallography and NMR spectroscopy identified significant differences in the solid- and solution- state for the Zn(II) species. All complexes have been applied to the ring opening polymerization of Llactide with emphasis on industrial conditions. High conversion and good molecular weight control was generally achievable for Zn(A-D)2, high molecular weight PLA was prepared in 1 min at a 10,000:1:33 [LA]:[Zn]:[BnOH] loading. The more active Zn(II) catalysts were also applied to PLA degradation to alkyl lactate under mild conditions. Zn(A-B)<sub>2</sub> demonstrate high activity and selectivity in this process with PLA being consumed within 1 h at 50 °C. Zn(C-D)<sub>2</sub> were shown to be less active and these observations can be related to the catalyst's structure and the degradation mechanism. Initial results for the degradation of poly(ethylene terephthalate) and mixed feeds are also presented, highlighting the broader applicability of the systems presented.

Due to the inevitable depletion of fossil fuel resources, and inherent carbon emissions, alternatives to petrochemical plastics are desperately needed.<sup>[1]</sup> Poly(lactic acid) (PLA) is a potential replacement for fossil fuel derived plastics used for packaging applications.<sup>[2,3]</sup> PLA has the added advantage of being biocompatible, therefore suitable for biomedical applications.<sup>[4,5]</sup> As it is derived from annually harvested crops, PLA is biorenewable and has promising green credentials in terms of  $CO_2$  emissions and Life-cycle assessment.<sup>[6,7]</sup> Preparation of high molecular weight PLA is preferentially from the cyclic dimer of lactic acid, lactide (LA), *via* ring opening polymerization (ROP).<sup>[8]</sup> Current PLA research seeks to reduce energy/material input of LA monomer synthesis,<sup>[9–12]</sup> demonstrate and elucidate stereoselective initiation,<sup>[13-29]</sup> and prepare robust initiators to compete with Sn(Oct)<sub>2</sub> under industrial conditions.<sup>[30–34]</sup>

Plastic waste and pollution is a further 21<sup>st</sup> century challenge for both academia and industry. While PLA is biodegradable under high temperature industrial conditions, it will not readily degrade

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	end of the document.

in the natural environment and therefore will contribute to the millions of tonnes of waste in landfill and in oceans.[35-37] End of life plastic waste management is key to tackling this issue and it is imperative this is addressed for all aspiring materials such as PLA. For PLA, chemical recycling is a particularly attractive route as it can produce value-added products such as alkyl lactates, lactic acid and acrylic acid.<sup>[38,39]</sup> These can be useful in their own right or used to reform LA, and therefore PLA, to facilitate a circular economy approach. Lactic acid, for example, is regarded as a platform chemical and alkyl lactates are considered green solvents.<sup>[40-44]</sup> The conversion of PLA back to lactic acid is highly desirable as the traditional fermentation route is costly and wasteful, producing gypsum as a by-product.<sup>[42]</sup> Compared to other waste management strategies such as incineration and biodegradation, chemical recycling offers the greatest value with potential to reduce the cost of PLA and increase market uptake.[45] Previous examples of PLA recycling processes include hydrolysis,<sup>[46–53]</sup> alcoholysis,<sup>[54–63]</sup> hydrogenation,<sup>[64,65]</sup> and hydrosilylation.[66]

Commercially available metal salts and precursors have been shown to facilitate the transesterification of PLA to alkyl lactates, typically methyl lactate (Me-La).[55,58,67] Sobota and co-workers have also shown the use of metallic precursors at relatively high temperatures, demonstrating a range of alcohols for transesterification.[55] There are also examples of organocatalyzed degradation with TBD and DMAP.<sup>[59,63]</sup> The use of metal complexes for this purpose is rare despite the amount of complexes reported for LA polymerization.[60,68,69] Recently we have reported the use of a discrete homoleptic Zn(II) complex {Zn(A<sup>ethyl</sup>)<sub>2</sub>} (where ethyl denotes the length of the N-N backbone with the same substituents as in Scheme 1) for PLA degradation to Me-La. To demonstrate the versatile nature of the process, a range of PLA grades were degraded in a controlled manner; experimental design and kinetic modelling was used to further elucidate key experimental factors and reaction mechanism.<sup>[62]</sup>

Herein, we report the rapid polymerization of LA, under industrially relevant conditions, competitive with Sn(Oct)<sub>2</sub>, and the fast and selective chemical degradation of PLA. The effect of ligand and metal on these processes is highlighted and discussed.

$$R^{1} \xrightarrow{\text{OH}} R^{2} \xrightarrow{\text{Zn}(\text{Et})_{2}} \text{Zn}(\textbf{A-D})_{2}$$

**A**H,  $R^1 = {}^{t}Bu$ ,  $R^2 = N(H)Me$  **B**H,  $R^1 = {}^{t}Bu$ ,  $R^2 = NMe_2$  **C**H,  $R^1 = CI$ ,  $R^2 = NMe_2$ **D**H,  $R^1 = {}^{t}Bu$ ,  $R^2 = H$ 

Scheme 1. Synthesis of metal complexes.



Figure 1. Solid-state structure of, from left to right, [Zn(A)<sub>2</sub>]<sub>2</sub>, Zn(B)<sub>2</sub>, Zn(C)<sub>2</sub> (top), Zn(D)<sub>2</sub> (bottom) ellipsoids are shown at 30% probability level and all hydrogen atoms have been omitted for clarity.

Ligands, **A-DH**, were prepared *via* a simple Schiff base condensation in methanol (MeOH). All ligands were dried prior to complexation and characterized by multinuclear NMR spectroscopy and high-resolution mass spectrometry. **A**H exists as two isomers in solution, having two distinct series of resonances. This results from ligand cyclisation due to intramolecular attack of the secondary amine (see ESI). Complexes were prepared using diethylzinc, with the homoleptic complex,  $Zn(A-D)_2$ , targeted (Scheme 1, Figure 1). Successful formation of  $Zn(A-D)_2$  was confirmed by multinuclear NMR spectroscopy, X-ray crystallography and elemental analysis for all complexes.

	Solid-State			Solution		
Complex	CN	τ(4/5)	(Pseudo) Structure	D / × 10 <sup>-10</sup> m <sup>2</sup> s <sup>-1</sup>	Predicted Coordination	
Zn( <b>A</b> ) <sub>2</sub>	5 (Dimer)	0.54	t.b.p	5.89	4/5	
Zn( <b>B</b> ) <sub>2</sub>	4	0.80	Tet.	5.94	4	
Zn( <b>C</b> ) <sub>2</sub>	6	-	Oct.	8.12	6	
Zn( <b>D</b> ) <sub>2</sub>	4	0.87	Tet.	6.70	4	
Zn( <b>A</b> <sup>ethyl</sup> ) <sub>2</sub>	6	-	Oct.	7.38	6	
$Zn(\mathbf{B^{ethyl}})_2$	5	0.01	s.b.p	7.11	5/6	
Zn( <b>C</b> <sup>ethyl</sup> ) <sub>2</sub>	-		-	8.16	6	

Diffusion coefficients calculated from DOSY NMR spectroscopy (CDCl<sub>3</sub>). t.b.p = trigonal bipyramidal, s.b.p = square based pyramidal.

A range of different coordination numbers and motifs were observed in the solid-state (Figure 1, Table 1).  $Zn(A)_2$  tended towards a dimeric species while the remaining structures were monomeric. The <sup>1</sup>H NMR spectra of all complexes were consistent with a symmetrical monomeric structure in solution. DOSY NMR spectroscopy was used to further analyze the solution-state structure of these complexes (Table 1).The decrease in diffusion coefficients relative to the ethylenediamine analogues,  $Zn(A-C^{ethyl})_2$ , is suggested to be due a lower coordination number and unbound amine moieties.

Initial studies were carried out to demonstrate the activities of these complexes towards ROP of L-lactide (Table 2). Singly recrystallized lactide was used to align with industrial practices and demonstrate the robust nature of the complexes. Polymerization reactions were exclusively performed under solvent free conditions and high temperatures (180 °C) with low metal loading ([LA]:[Zn]:[BnOH] = 10,000:1:100) to increase industrial relevance. The ratio of metal complex to alcohol is consistent with an immortal polymerization. The complexes were tolerant to air/moisture in the solid-state allowing for storage under ambient conditions. However, the complexes had no polymerization activity in air, signifying complex decomposition under the reaction conditions. Therefore all reactions were performed under argon. Under these conditions, Zn(A/B)<sub>2</sub> demonstrated the highest activity achieving high conversion within 3 min. Excellent molecular weight control was achieved despite the challenging conditions. There is good agreement between the theoretical molecular weight, based on conversion, and that measured via GPC {for  $Zn(A)_2$ ,  $M_n = 14,400 \text{ mol}^{-1}$ ,  $M_{n,theo}$ = 14,100 g mol<sup>-1</sup>}. In addition, a relatively narrow dispersity is observed (D = 1.24 - 1.45). The relative activities of  $Zn(A/B)_2$ agree with the previous ethylene diamine based system,<sup>[31]</sup> but the complexes herein are twice as active. The activities of Zn(A/B)<sub>2</sub> are comparable to Sn(Oct)<sub>2</sub> under identical conditions {for Sn(Oct)<sub>2</sub>, 3 minutes, 95%,  $M_n = 16,100$  Da, D = 1.54}.<sup>[31]</sup> Zn(C/D)<sub>2</sub> required considerably

#### Table 2. Polymerisation of L-lactide

Cat.	Time / min	X <sub>L-LA</sub> . % <sup>[d]</sup>	M <sub>n,theo</sub> <sup>[e]</sup>	<i>M</i> <sup>[f]</sup>	${\cal D}^{[{ m f}]}$
Zn( <b>A</b> ) <sub>2</sub> <sup>[a]</sup>	1	98	14,100	14,400	1.27
$Zn(\mathbf{A})_{2^{[b]}}$	1	97	42,000	53,750	1.95
$Zn(\mathbf{A})_{2^{[c]}}$	30	77	11,200	9,400	1.54
$Zn(\mathbf{B})_{2}^{[a]}$	3	94	13,650	11,500	1.45
$Zn(\boldsymbol{C})_2^{[a]}$	30	69	10,050	10,150	1.11
$Zn(\mathbf{D})_2^{[a]}$	12	89	12,900	13,350	1.21

Conditions: *L*-LA, 180 °C, solvent free; [a] [LA]:[I]:[BnOH] = 10,000:1:100; [b] [LA]:[Zn]:[BnOH] = 10,000:1:33; [c] [LA]:[I]:[BnOH] = 20,000:1:200. [d] Determined by <sup>1</sup>H NMR spectroscopy; [e] Theoretical molecular weight calculated from conversion and [LA]:[BnOH]; [f] Measured by GPC (RI), correction factor of 0.58 has been applied to  $M_n$  and  $M_w$ .

more time (15-30 min) to achieve a conversion similar to that of  $Zn(A/B)_2$ . Despite the extended reaction time, good control over molecular weight was maintained. All catalysts furnished white polymer under these conditions (see ESI).  $Zn(C)_2$  The slower reaction for Zn(C)<sub>2</sub> is likely related to the more sterically hindered octahedral metal centre and this is likely a reason for increased molecular weight control (D = 1.11). MALDI-ToF analysis of polymer derived from  $Zn(C)_2$  revealed a symmetrical distribution with the expected BnO-/-H end groups. A peak spacing of 144 g mol<sup>-1</sup> demonstrates the absence of undesired transesterification reactions. Similar to  $Zn(A/B)_2$ ,  $Zn(C)_2$  is more active than the analogous ethylene diamine complex, Zn(Cethyl)2, attaining a similar conversion in half the time.<sup>[31]</sup> The reduction in activity for  $Zn(D)_2$  could be related to the mechanistic pathway. It is suggested that all initiators follow an activated monomer mechanism,<sup>[70]</sup> with molecular weight control suggesting immortal characteristics. However, the pendant amine groups of Zn(A-C)<sub>2</sub> are suggested to participate in the reaction, potentially assisting in the proton transfer steps, leading to a ligand-assisted activated monomer mechanism.<sup>[71]</sup> In the case of  $Zn(\mathbf{D})_2$ , there are no amine groups to facilitate this step leading to the difference in activity when compared to Zn(A-C)<sub>2</sub>. It is also possible that these amine moieties allow the formation of dimeric zinc structures in which there is a cooperative effect between metal centres.

DSC analysis of the PLA produced by  $Zn(A-D)_2$  revealed melting temperatures of 160 – 167 °C (see ESI). This indicates high polymer crystallinity and a lack of stereocentre epimerization.

The potential to control molecular weight by varying the ratio of benzyl alcohol has also been demonstrated with  $Zn(A)_2$ . Higher molecular weight PLA was achieved through the reduction of alcohol amount ([LA]:[I]:[BnOH] = 10,000:1:33). The rapidity of the polymerization remains unchanged, with high conversion achieved within 1 min. However, molecular weight control is slightly reduced ( $M_n = 53,750 \text{ g mol}^{-1}$ ,  $M_{n,theo} = 42,000 \text{ mol}^{-1}$ ;  $\mathcal{D} = 1.95$ ). This catalyst was further tested by reducing the catalyst loading ([LA]:[I]:[BnOH] = 20,000:1:200). Reaction time is greatly lengthened to 30 min, however reasonable conversion is still achieved. There is also a decrease in molecular weight control ( $\mathcal{D} = 1.54$ ) and this is likely related to the extended reaction time.

Table 3. PLA degradation at 50 °C

Cat.	Time / h	X <sub>Int</sub>	SA-La	Y <sub>A-La</sub>	<i>k<sub>app</sub>∕</i> min⁻¹
$Zn(\mathbf{A})_2$	0.5	100	81	81	0.20
$Zn(\mathbf{B})_2$	1	100	84	84	0.094
Zn( <b>C</b> ) <sub>2</sub>	3	51	20	10	3.8 × 10⁻³
Zn( <b>D</b> ) <sub>2</sub>	3	29	17	5	1.7 × 10 <sup>-3</sup>
$Zn(\mathbf{A}^{ethyl})_{2}^{[a]}$	3	66	29	19	5.9 × 10 <sup>-3</sup>
$Zn(\mathbf{A})_2^{[b]}$	0.5	100	89	89	0.14
$Zn(\mathbf{A})_2^{[c]}$	0.5	100	63	63	0.12
$Zn(\mathbf{B})_2^{[b]}$	1	99	81	80	0.066
$Zn(\mathbf{B})_2^{[c]}$	1	99	71	70	0.064
Zn( <b>A</b> ) <sub>2</sub> <sup>[d]</sup>	1.5	93	55	53	0.029
$Zn(\mathbf{B})_2^{[d]}$	1.5	95	60	57	0.028
Zn( <b>B</b> ) <sub>2</sub> <sup>[e]</sup>	1	100	87	87	0.087
$Zn(\mathbf{B})_2^{[f]}$	1	100	80	80	0.094

Conditions: 50 °C, 4 wt % catalyst loading, 0.42 – 0.48 mol % relative to ester linkages, THF/MeOH = 4:1,  $n_{MeOH}$ :  $n_{ester}$  = 7:1; Internal methine group conversion (*X<sub>m</sub>*), A-La selectivity (S<sub>A-La</sub>) and A-La yield (Y<sub>A-La</sub>) by <sup>1</sup>H NMR spectroscopy. [a] Ethylene diamine complex; [b] Toluene; [c] Acetonitrile; [d] EtOH;  $n_{EtOH}$ :  $n_{ester}$  = 7:1; [e] PET (100 mg) added; [f] PET (250 mg) added.

Initial degradation studies were carried out using commercially available PLA (0.25 g, VegWare<sup>TM</sup>, PLA cup,  $M_n$  = 45,150 g mol<sup>-1</sup>). Degradation progress was monitored by regular sampling and <sup>1</sup>H NMR spectroscopic analysis. This technique quantified the relative concentrations of methine groups which are defined as internal (Int), chain end (CE) and alkyl lactate (A-La). We have previously shown the production of Me-La is a two step process *via* the intermediate formation of CE groups (see ESI).<sup>[62]</sup> For comparison, internal methine conversion (*X*<sub>Int</sub>), alkyl lactate selectivity (*S*<sub>A-La</sub>), and alkyl lactate yield (*Y*<sub>A-La</sub>) are provided for each catalyst.

Our previous studies demonstrated the use of THF/MeOH at 50 °C to be reasonable conditions for the transesterification reaction and these conditions were used to perform a catalyst screen for PLA degradation activity. The amount of catalyst was reduced from 8 to 4 wt % in an attempt to improve the viability of the process. Under these conditions,  $Zn(A)_2$  was shown to be highly active, with PLA being completely consumed within 30 min, with 20% of CE methine groups remaining (Table 3). For Zn(B)<sub>2</sub>, complete PLA consumption was generally achieved after 1 h. In contrast, Zn(C/D)2 were poorer towards PLA degradation under the same conditions, with PLA still being present after 20 h with a low Me-La selectivity.  $Zn(C)_2$  and  $Zn(A^{ethyl})_2$ , which has an ethylenediamine backbone, had rates and selectivities on the same order of magnitude. The activity of these complexes can be related to the geometry, where those with a higher coordination demonstrated a lower activity. For Zn(D)2, the reduced activity

may suggest the presence of the pendant amine groups to be important, perhaps assisting in the degradation mechanism through hydrogen bonding interactions as suggested for the polymerization studies. In comparison, commercially available  $ZnCl_2$  was found to be inactive for degradation under these conditions and we have previously shown  $Zn(OAc)_2$  to be much less active than  $Zn(\mathbf{A}^{\text{ethyl}})_2$ . <sup>[62]</sup>

The active catalyst species was investigated by removal of volatile components, with the residue being analysed by <sup>1</sup>H NMR spectroscopy. For Zn(**A**,**B**,**D**)<sub>2</sub>, there is evidence of ligand dissociation, as well as the formation of a new species. This new species is suggested to be a heteroleptic complex, Zn**X**L, formed with concomitant loss of one ligand. The ancillary ligand could be methoxy, lactyl or higher oligomers. For Zn(**C**)<sub>2</sub>, and our previous Zn(**A**<sup>ethyl</sup>)<sub>2</sub> catalyst, there is no evidence of ligand dissociation, despite the extended reaction time. Instead, there is only broadening of the Zn(II) complex NMR resonances suggesting the coordination of reaction components only.

These complexes were six coordinate in the solid-state and it is likely that the propensity for a higher coordination prevents dissociation. Equally, the dissociation of ligand for  $Zn(A/B)_2$  (5 and 4 coordinate respectively), likely leads to the enhanced activity. Based on this result, a control experiment was performed with ligand BH (12 wt %). No degradation was observed after 24 h indicating dissociated ligand was not the active species.

For  $Zn(\mathbf{C})_2$ , time-dependent GPC kinetics was performed (see ESI). An exponential decrease in molecular weight is observed, with  $M_n$  being less than 10% of the original value after 30 min. This suggests that the chain scission is random and not localized at chain ends. The rapid decrease in molecular weight precludes similar analysis for other catalysts.

The scope of solvents was expanded to include toluene and acetonitrile with  $Zn(A/B)_2$ . For both complexes there was a slight reduction in the rate of degradation but the selectivity and yield remained high after 30 min. The ability to produce ethyl lactate is arguably more desirable, with this compound having wider applications than Me-La.<sup>[43,44]</sup> Using ethanol (EtOH), instead of MeOH, reduced the rate of degradation for both catalysts, however, high conversion of [Int] units was still achievable within 90 min showing the viability of producing other alkyl lactates.



Conditions: 130 °C, 1 h, 1 wt % catalyst loading, 0.105 - 0.110 mol % relative to ester linkages, MeOH (2 mL),  $n_{MeOH}$ :  $n_{ester} = 14:1$ ;  $n_{MeOH}$ :  $n_{ester} = 14:1$ ; Internal methine group conversion ( $X_{Int}$ ), A-La selectivity ( $S_{A-La}$ ) and A-La yield ( $Y_{A-La}$ ) by <sup>1</sup>H NMR spectroscopy.

 $Zn(\mathbf{A}/\mathbf{B})_2$  were also tested in the absence of solvent at 130 °C (Table 4). Catalyst loading was decreased further to 1 wt %. Under these conditions, full PLA consumption is generally achieved after 1 h. As for the solution degradation reactions,  $Zn(\mathbf{A})_2$  was more efficient, giving a higher selectivity towards Me-La.

Initial investigations were made into the degradation of other polyesters. With  $Zn(B)_2$ , the degradation of poly(ethylene terephthalate) (Coca-Cola<sup>®</sup> bottle) was demonstrated at 150 °C with benzyl alcohol (5 wt %, 30 min). Complete solubilisation and subsequent analysis by <sup>1</sup>H NMR spectroscopy demonstrated the complete degradation of PET under these conditions. On cooling, the benzyl alcohol di-ester crystallised from solution the structure was confirmed by single crystal X-ray diffraction (see ESI).

The contamination of plastic waste streams with different polymers can severely impact the recycling procedure and resultant material quality.<sup>[67,72]</sup> For Zn(**B**)<sub>2</sub>, the rate of degradation of PLA, in solution with a PLA/PET mixed feed, was not impaired by addition of PET (100 - 250 mg). Subsequent removal of Me-La *via* distillation and addition of benzyl alcohol then allowed for the selective degradation of PET at 150 °C. Crystallization of the terephthalic di-ester allowed for the removal of trace amounts of benzyl lactate.

The scalability of the PLA degradation process was also tested on the  $Zn(\mathbf{B})_2$  catalyst, with reactions being performed at 50 °C in a 300 mL SS316 bench top stirred reactor (See ESI). In a typical experiment, 12.5 g of commercial PLA (NatureWorks Ingeo TM 6202D,  $M_n = 44,350$  g mol<sup>-1</sup>) reacted with 50 mL of MeOH in the presence of 250 mL of THF as solvent, and 4 wt % catalyst loading. The progress of the reactions was followed by measuring the A-La concentration by GC-FID with further analysis by <sup>1</sup>H NMR spectroscopy to identify the CE intermediates. For GC analysis, 0.07 g mL<sup>-1</sup> is the maximum Me-La concentration achieved (maximum theoretical is 0.072 g mL<sup>-1</sup>), suggesting a similar reaction mechanism with both catalysts (Figure 2, S31 and S39).  $Zn(B)_2$  clearly showed to be higher in activity than  $Zn(A^{ethyl})_2$ (Figure 2). The calculated X<sub>Int</sub>, S<sub>A-La</sub> and Y<sub>A-La</sub> and rate coefficient for the degradation step  $(k_1, \text{ Table 5})$  are in good agreement with the values obtained at the small scale studies (Table 3) demonstrating the scale-up potential of the process. Rate coefficients  $k_2$  and  $k_2$  correspond to the reversible reaction for A-La formation from CE groups (Figures S31 and S39), calculated from the kinetic model presented in [62].

Table 5.	Scale up	studies of	PLA	degradation	at 50	°C.
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Cat.	Time / h	X <sub>Int</sub>	S <sub>A-La</sub>	Y <sub>A-La</sub>	$k_1/\min^{-1}$	$k_2 / \min^{-1}$	<i>k</i> -2/min <sup>-1</sup>
Zn( <b>B</b> ) <sub>2</sub>	1	97	79	77	6.9 × 10 <sup>-2</sup>	5.5 × 10 <sup>-2</sup>	6.3 × 10 <sup>-3</sup>
Zn( <b>A</b> <sup>ethyl</sup> ) <sub>2</sub>	1	30	17	5	0.4 × 10 <sup>-2</sup>	0.2 × 10 <sup>-2</sup>	0.5 × 10 <sup>-3</sup>

Conditions: 50 °C, 1 h, PLA = 12.5 g, 4 wt % catalyst loading, 0.40 – 0.43 mol % relative to ester linkages, THF/MeOH = 4:1,  $n_{MeOH}$ :  $n_{ester}$  = 7:1; Internal methine group conversion ( $X_{Int}$ ), A-La selectivity ( $S_{A-La}$ ) and A-La yield ( $Y_{A-La}$ ) by <sup>1</sup>H NMR spectroscopy.



**Figure 2**. A-La concentration profiles at 50 °C with  $Zn(B)_2$  and  $Zn(A^{\text{ethyl}})_2$  (maximum theoretical Me-La concentration is 0.072 g mL<sup>-1</sup>).

In conclusion, four new and fully characterized zinc complexes have been applied to the catalytic ROP of *L*-lactide and the transesterification of PLA into alkyl lactates under industrially relevant conditions. Various degradation parameters (solvent, solvent-free, temperature, alcohol) were assessed and the difference in the activities of the complexes were found to be related to the differences in their structure. The degradation process has also been scaled up for the more active catalyst and kinetic parameters have been calculated demonstrating the fast degradation at mild conditions. As a proof-of-concept, the selective degradation of PET has also been demonstrated.

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**Keywords:** Poly(lactic) acid • alkyl lactate • degradation • polymerisation • Poly(ethylene terephthalate) • homogeneous catalysis.

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