

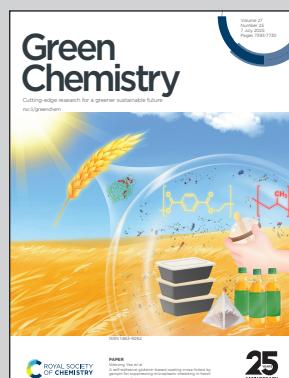
Showcasing research by Y. Kinbara, Professor H. Ishitani and Professor S. Kobayashi from The University of Tokyo, Japan.

Efficient low-temperature depolymerization of polycarbonate catalyzed by lanthanum  $\beta$ -diketonate complexes

Efficient catalysis of lanthanum diketonate-type complex for the depolymerization of polycarbonate was optimized, and a system capable of converting it into monomers at temperatures below 60°C was developed. Slurrying the polycarbonate sample with toluene prior to depolymerization was revealed to accelerate the methanolysis reaction.

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## Efficient low-temperature depolymerization of polycarbonate catalyzed by lanthanum $\beta$ -diketonate complexes<sup>†</sup>

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The chemical recycling of polycarbonate is crucial for addressing plastic waste and promoting sustainability. Conventional methods often require high temperatures and harsh conditions, leading to energy consumption and potential degradation of the recovered monomers. In this study, we present a high-yielding depolymerization of BPA-based powdered polycarbonate, prepared through a pretreatment with toluene as the substrate using a series of lanthanum  $\beta$ -diketonate complexes as catalysts. Among the complexes investigated, lanthanum 3,4-dimethoxybenzoylacetone exhibited the highest catalytic activity. This method enables depolymerization at significantly lower temperatures (e.g., 60 °C) compared to conventional approaches without using co-solvent. Under optimized conditions, we achieved a high depolymerization rate to give monomers in 96% and 93% yield within 8 h. This mild and high-yielding depolymerization process offers a promising approach to sustainable polycarbonate recycling and contributes to the development of a circular economy.

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### Green foundation

1. We have developed a catalytic system that enables the depolymerization of polycarbonate with methanol as a sole solvent into dimethyl carbonate and bisphenol A at 60 °C, achieving over 93% monomer yield. This was achieved using an optimized ligand and a simple pre-treatment method involving only mixing with toluene at room temperature.
2. We identified a highly active lanthanum benzoylacetone complex and conducted a simultaneous analysis of both liquid and solid phases, providing new insights into the reaction mechanism of solidstate polymers. The slurry obtained can be separated by filtration into powdered PC and toluene, allowing the recovered toluene to be reused.
3. This study highlights the strong impact of initial polymer characteristics on depolymerization reactivity. A more detailed investigation into changes in molecular weight distribution during pretreatment is expected to contribute to more advanced plastic recycling technologies.

## Introduction

For thermoplastics like polycarbonate, chemical recycling presents a promising alternative to traditional mechanical recycling, enabling the recovery of monomers and the production of virgin-quality materials.<sup>1</sup> However, despite this potential, the proportion of chemical recycling remains surprisingly low.<sup>2–4</sup> This limited adoption can be attributed to several factors, including polymer degradation due to environmental exposure, the complexity of mixed plastic waste

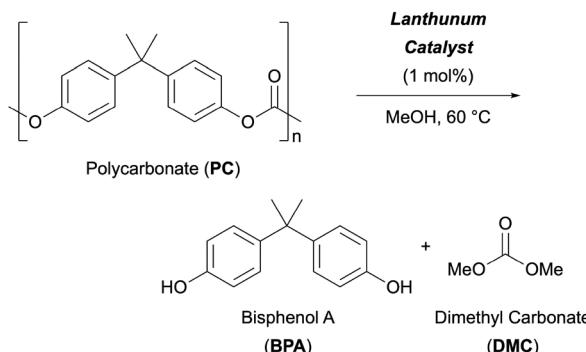
streams, and the economic challenges associated with sorting, depolymerization, and purification processes.<sup>5–8</sup> To achieve sustainable chemical manufacturing and reduce reliance on fossil resources, overcoming the technical challenges associated with chemical recycling is of paramount importance. In particular, reducing the energy requirements of the depolymerization process is a crucial and urgent issue. We focused on polycarbonate (PC), an ester-based polymer relatively amenable to depolymerization reactions (Scheme 1). Polycarbonate is an engineering plastic widely used in applications such as low-refractive-index eyeglass lenses and protective goggles due to its high transparency and impact resistance.<sup>9</sup> Various methods for PC depolymerization have been reported, including pyrolysis,<sup>10,11</sup> hydrolysis,<sup>12–14</sup> alcoholysis,<sup>15–20</sup> and aminolysis.<sup>21–23</sup> However, pyrolysis requires significant thermal energy and produces a mixture of low-molecular-weight compounds.<sup>10,11</sup> While hydrolysis can yield bisphenol A (BPA) monomer with high efficiency, it necessitates reactors

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Scheme 1 Methanolysis of PC with La complex catalyst.

capable of withstanding high-pressure steam, which is a disadvantage. Additionally, the generation of carbon dioxide as a by-product is problematic.<sup>13,14</sup>

Transesterification with alcohols, glycols, and amines enables depolymerization under relatively mild conditions. The nucleophile primarily influences this reaction. Among the various depolymerization methods available, this study focuses on methanolysis. Methanolysis simultaneously produces bisphenol A (BPA) and dimethyl carbonate (DMC). The use of DMC as an intermediate in polycarbonate synthesis is attracting attention because it eliminates the need for hazardous phosgene.<sup>24,25</sup> This depolymerization process offers advantages beyond raw material recycling, including the high industrial value of DMC. DMC finds wide application in areas such as semiconductor manufacturing, coatings, adhesives, and as an electrolyte in lithium-ion batteries. This underscores the potential of this process to effectively convert waste plastics into high-value products.<sup>26–30</sup>

Numerous studies on the methanolysis of PC have been conducted over the past decade. Analysis of these studies reveals two main approaches: those using co-solvents and those using methanol as the sole solvent. Co-solvents can lower the depolymerization temperature to below 80 °C,<sup>15</sup> while methanol-sole systems typically require temperatures of 100 °C<sup>16</sup> or higher, often 120–140 °C.<sup>17–20</sup> This trend suggests that PC solubility in the solvent significantly influences reactivity. When considering industrial-scale implementation of chemical recycling, the use of co-solvents should be minimized, and low-energy processes are preferable. From this perspective, achieving high-yielding depolymerization using methanol as the sole solvent, particularly below its boiling point, would be highly significant.

Moreover, to obtain high yields of both BPA and DMC, nearly neutral or mildly acidic conditions, typically achieved using Lewis acids, are required. While many metal Lewis acids are not well-suited for highly Lewis basic conditions, the high oxophilicity and rapid ligand-exchange capability of rare earth compounds might make this possible. Indeed, Hirano *et al.* have demonstrated the effectiveness of lanthanum acetyl-acetonate ( $\text{La}(\text{acac})_3$ , **1**) in the methanolysis of PET and PC.<sup>16,31</sup> Their research suggests the potential of rare earth Lewis acids,

and optimizing catalyst structure is considered an effective approach to enable depolymerization at lower temperatures.

In this study, we carefully explored catalysts, especially the structure of ligands, for the methanolysis of PC. We focused particularly on the influence of rare earth 1,3-diketone and ketoester ligands on activity, identifying the most active complex from a total of 20 complexes.<sup>32,33</sup> Furthermore, to deepen our understanding of PC methanolysis, we analyzed the molecular weight distribution in the initial stages of the reaction using GPC, investigating the macroscopic reaction process of polymer degradation. These findings provide interesting insights for catalyst and reaction design in the mild depolymerization of polycarbonate.

## Results and discussion

As mentioned above, the depolymerization of PC at lower temperatures is highly dependent on the choice of solvent. Our strategy involves converting the polymer into its more reactive state through a pre-treatment process,<sup>34</sup> enabling high-yielding depolymerization at lower temperatures and in a shorter reaction time. In this study, we incorporated a dispersion step in toluene as a pre-treatment method. While this pre-treatment step might appear inefficient, it should be noted that if the slurring of PC in toluene proceeds almost quantitatively at room temperature, the heating required for depolymerization is expected to decrease. Furthermore, as will be discussed later, this method allows for the selective slurring of PC from plastic products, making it advantageous from a separation and recycling perspective.

When PC powder, obtained through toluene treatment, was treated in methanol without any catalyst at 60 °C, only trace amounts of the decomposition products BPA and DMC were detected (entry 1, Table 1). In contrast, the addition of 1 mol% of  $\text{La}(\text{acac})_3$  (**1**) significantly accelerated the reaction, yielding BPA and DMC in 36% and 40% yields, respectively (entry 5).

Table 1 Catalytic screening for methanolysis of PC<sup>a</sup>

Entry	Catalyst	Temp. [°C]	Yield <sup>b</sup> [%]	
			BPA	DMC
1	No	60	3	3
2	$\text{LaCl}_3$	60	13	19
3	$\text{La}(\text{OTf})_3$	60	26	28
4	$\text{La}(\text{OAc})_3$	60	2	4
5	$\text{La}(\text{acac})_3$ ( <b>1</b> )	60	36	40
6	$\text{La}(\text{O}^{\prime}\text{Pr})_3$	60	9	8
7	$\text{La}(\text{OH})_3$	60	3	3
8	No	80	20	20
9	$\text{La}(\text{OTf})_3$	80	76	78
10	$\text{La}(\text{acac})_3$	80	87	92

<sup>a</sup> All reactions were performed using 1.02 g of powdered PC prepared through toluene pre-treatment (4 mmol, based on BPA-CO units) and 0.04 mmol of La complexes in 40 mL of methanol at 60 °C (for entries 1 to 7) or at 80 °C (for entries 8 to 10) for 2 h. <sup>b</sup> Determined by GC analysis.

Although  $\text{La}(\text{acac})_3$  is classified as a mild Lewis acid, its effectiveness in ester exchange reactions has been demonstrated not only by Hirano *et al.*<sup>16,28</sup> but also by Neverov and Brown,<sup>35,36</sup> making it noteworthy for this type of reaction.

Remarkably, it outperformed  $\text{LaCl}_3$  and  $\text{La}(\text{OTf})_3$ , which were expected to exhibit higher Lewis acidity.<sup>28,37</sup> Considering that  $\text{La}(\text{O}^i\text{Pr})_3$  and  $\text{La}(\text{OH})_3$ , which are predicted to have basic characteristics, showed almost no activity, it is plausible that La-dicarbonyl-type complexes act in a concerted manner as Lewis acids, not only activating the carbonyl group in PC but also generating alkoxides in an alcoholic solvent, methanol. In fact, when  $\text{La}(\text{acac})_3$  was used, BPA was obtained in an 87% yield at 80 °C (entry 10). This result represents one of the lowest temperature examples of PC depolymerization in methanol reported to date. Nevertheless, aiming to achieve even lower temperature conditions, we focused on optimizing the structure of 1,3-dicarbonyl-type ligands through systematic screening in this study.

To explore the effect of substituents on the 1,3-dicarbonyl scaffold, we prepared nine different diketones illustrated in Fig. 1, all based on the fundamental 1,3-diketone structure, and converted them into lanthanum complexes in basic methanol. Since each complex crystallized in methanol, we utilized the resulting solids as catalysts to investigate their activity in the depolymerization of PC at 60 °C in methanol (Table 2). This study primarily focused on examining the effects of substituent groups at the external carbon positions, using the acetylacetone **2a** complex, **1**, as the reference. The results showed that the introduction of an  $^i\text{Pr}$  group slightly improved the yield, while bulkier substituents such as the  $^t\text{Bu}$  group significantly decreased the yield. This trend is likely due to steric hindrance caused by the bulky substituents near the carbonyl group, which serves as the coordination site for the La atom, making it more difficult to activate the carbonyl group of PC. In contrast, complexes with electron-withdrawing trifluoromethyl or heterocyclic groups (**2d–g**) did not exhibit a notable effect. However, an improvement in yield was observed with diketone **2h**, leading to complex **La-2h**, which featured benzene rings at both sides of the diketone skeleton. Similarly, complex **La-2i** also showed positive results, although its performance

**Table 2** Screening of 1,3-diketone ligands for methanolysis of PC<sup>a</sup>

Entry	Ligand	Catalyst	Yield <sup>b</sup> [%]	
			BPA	DMC
1	<b>2a</b>	<b>1</b>	36	40
2	<b>2b</b>	<b>La-2b</b>	46	47
3	<b>2c</b>	<b>La-2c</b>	11	11
4	<b>2d</b>	<b>La-2d</b>	28	28
5	<b>2e</b>	<b>La-2e</b>	13	12
6	<b>2f</b>	<b>La-2f</b>	12	13
7	<b>2g</b>	<b>La-2g</b>	26	26
8	<b>2h</b>	<b>La-2h</b>	69	67
9	<b>2i</b>	<b>La-2i</b>	45	45

<sup>a</sup> All reactions were performed using 1.02 g of powdered PC prepared through toluene pre-treatment (4 mmol, based on BPA-CO units) and 0.04 mmol of La complexes in 40 mL of methanol at 60 °C for 2 h.

<sup>b</sup> Determined by GC analysis.

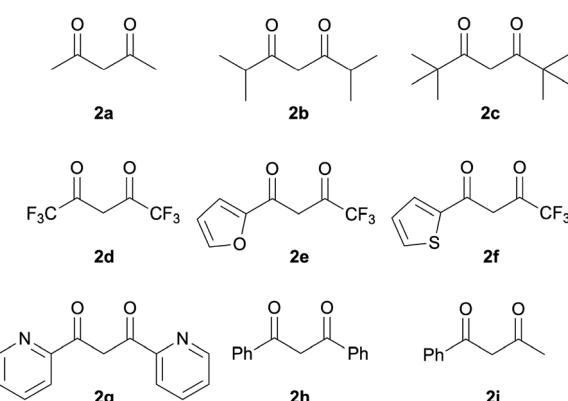
was comparable to that of the isopropyl-substituted complex **La-2b**.

Based on the above studies, it was determined that an ideal catalyst for the depolymerization of PC should have a La atom surrounded by a moderately spacious environment, coupled with electron-rich ligands. The above initial screening identified bis-aryl type ligands as the most effective. However, to prioritize ease of structural modification, we focused on benzoylacetone-type diketone ligands and examined the substituent effects on the aromatic ring of these ligands. Additionally, to evaluate the influence of electron-donating functional groups, we synthesized similar complexes using 1,3-ketoester ligands and assessed their catalytic activities (Fig. 2).

When using benzoylacetone (**2i**) as a standard, no clear trend was observed upon introducing either electron-withdrawing or electron-donating groups onto the aromatic ring. Notably, the use of ligand **3f** resulted in the highest yield among complexes synthesized under identical temperature conditions, slightly surpassing the activity of **La-2g**. Comparing the benzoylacetone derivatives, ligands **4a** and **4b** exhibited higher yields than their corresponding **2a** and **2i**-ligated lanthanum. However, the **La-4c** complex, which was specifically designed to enhance performance, unfortunately resulted in a lower yield (Table 3).

Additionally, the reaction profile of **2a** in combination with **2h** and **3f** were monitored. The results indicated that using **La-2h** and **La-3f**, the reactions reached nearly quantitative yields of BPA within 8 h, whereas the yield was only 65% when using **1** under the same conditions (Fig. 3). These findings demonstrate that the synthesized complex exhibits superior catalytic activity.

To gain insight into the methanolysis process of PC, the reaction solution with catalyst **La-2i** was analyzed using Gel Permeation Chromatography (GPC) at various reaction times (Fig. 4). As the reaction progressed, we observed a gradual increase in peaks corresponding to degradation products with molecular weights ranging from approximately 200 to 300. In contrast, the polymer components with molecular weights between  $1.0 \times 10^4$  and  $1.0 \times 10^5$  showed a sharp decrease after the reaction started, indicating a rapid depolymerization of



**Fig. 1** 1,3-Diketones used for ligand.



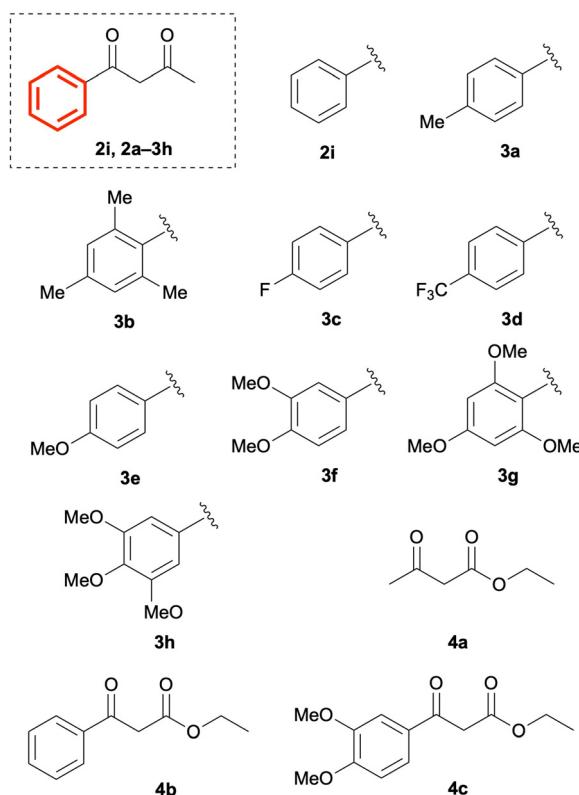


Fig. 2 Structure of benzoyl acetate ligands and 1,3-ketoesters.

Table 3 Effects of substituents on aromatic ring in benzoyl acetone-type ligands and 1,3-ketoesters<sup>a</sup>

Entry	Ligand	Catalyst	Yield <sup>b</sup> [%]	
			BPA	DMC
1	<b>2i</b>	<b>La-2i</b>	45	45
2	<b>3a</b>	<b>La-3a</b>	34	35
3	<b>3b</b>	<b>La-3b</b>	19	19
4	<b>3c</b>	<b>La-3c</b>	23	24
5	<b>3d</b>	<b>La-3d</b>	35	36
6	<b>3e</b>	<b>La-3e</b>	37	38
7	<b>3f</b>	<b>La-3f</b>	70(96) <sup>c</sup> [nd <sup>d</sup> ]	68(93) <sup>c</sup> [nd <sup>d</sup> ]
8	<b>3g</b>	<b>La-3g</b>	42	41
9	<b>3h</b>	<b>La-3h</b>	51	48
10	<b>4a</b>	<b>La-4a</b>	52	51
11	<b>4b</b>	<b>La-4b</b>	61	61
12	<b>4c</b>	<b>La-4c</b>	39	32

<sup>a</sup> All reactions were performed using 1.02 g of powdered PC prepared through toluene pre-treatment (4 mmol, based on BPA-CO units) and 0.04 mmol of La complexes in 40 mL of methanol at 60 °C for 2 h.

<sup>b</sup> Determined by GC analysis. <sup>c</sup> Yield at 8 h. <sup>d</sup> The reaction using untreated PC pellet. The methanolysis was conducted at 60 °C for 2 h.

large polymer molecules. This change in average molecular weight suggested that depolymerization occurred not only at the terminals of polymer chains but also at random positions along the chain. Interestingly, a new peak with a molecular weight around 2500 began to appear at approximately

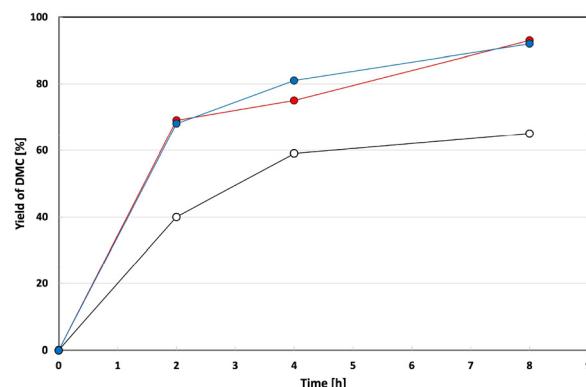


Fig. 3 Comparison of reaction profile for the reactions using catalyst 1, La-2h, and La-3f. Symbols: white, 1; blue, La-2h; red, La-3f.

30 minutes from the beginning. Although the prepared PC powder remained largely insoluble in methanol, a low-polymerization PC prepared by us was found to be able to divide into methanol-insoluble (Component A, Fig. 5) and soluble components (Component B, Fig. 5). GPC analysis revealed that the components with molecular weights of 2500 or higher significantly decreased in solubility in methanol (see also Scheme S1†). The peak of this insoluble component is matched the newly appeared peak in the GPC profile. These observations suggest a mechanism in which the polymer undergoes random methanol attack along the chain, leading to the formation of low-molecular-weight oligomers with a molecular weight of around 2500 that are initially insoluble in methanol. These oligomers subsequently undergo further depolymerization into a homogeneous system, rapidly converting to monomers. In the same catalytic system, using PC pellets without toluene pre-treatment, methanolysis at 80 °C resulted in some degree of molecular weight reduction and monomerization (SI-6†). However, the rate of this process was extremely slow, and after 24 h, the majority of the polymer still maintained an average molecular weight of approximately

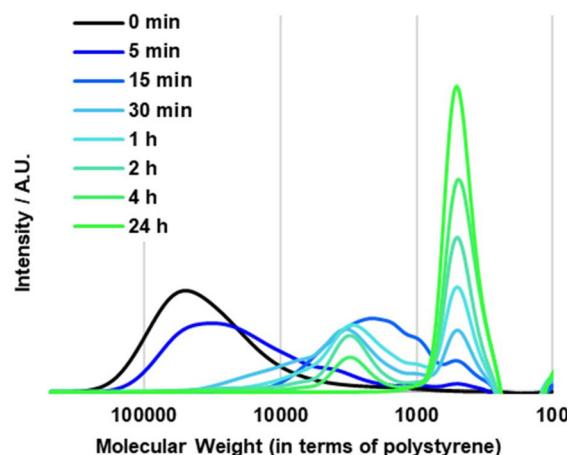


Fig. 4 Time-resolved GPC analysis of methanolysis reaction.



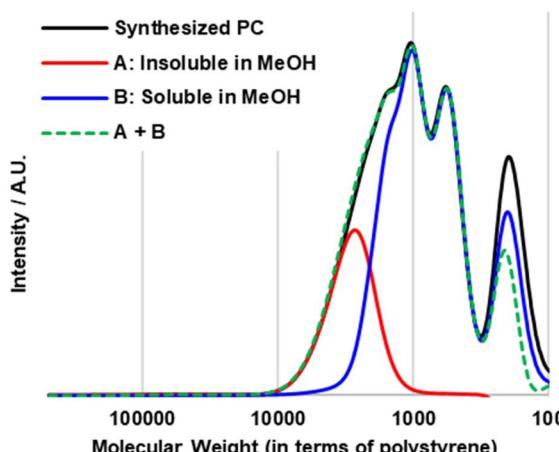
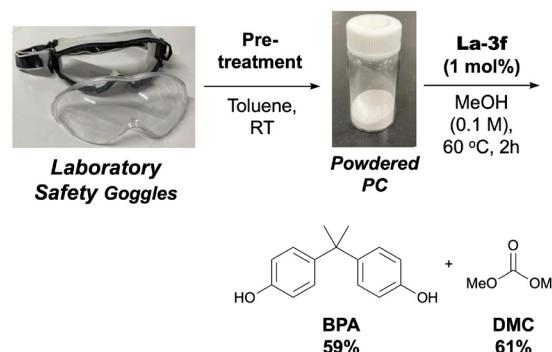


Fig. 5 Solubility of synthesized PC in methanol by GPC analysis.

10 000. This observation underscores the significant impact of toluene pre-treatment on the outcomes of this study. These studies provide examples of observing the dynamic behaviour of polymer depolymerization using GPC. The findings offer valuable insights that could influence the design of future reactions and contribute significantly to the understanding of depolymerization and chemical recycling processes in another polymer.

A key feature of this study is the successful low-temperature methanolysis achieved by leveraging the dispersibility of PC in toluene. This allows for initial powdering of the PC, which is then used as the starting material. While disposing of the toluene after each pre-treatment step would be inefficient, the recovered toluene can be reused in subsequent pre-treatment processes, addressing this concern. In our experiments, powdered PC was obtained after toluene evaporation. Notably, the PC after pre-treatment remains dispersed, not dissolved, in toluene, enabling separation *via* filtration. During filtration, impurities from the original PC and small amounts of soluble PC components will be present in the recovered toluene and may accumulate with repeated reuse (see ESI, SI-10†). Despite this, the energy requirements for toluene dispersion and recovery are estimated to be low. Furthermore, the addition of a pre-treatment step does not significantly hinder the chemical recycling of PC-based plastic products, as demonstrated with actual products.

To illustrate this, we depolymerized the lens portion of polycarbonate safety goggles. After removing the band, the lens material was coarsely shredded. These fragments were soaked in toluene overnight, and following the procedures described earlier, a powder was obtained. IR spectroscopy analysis showed the powder's spectrum to be nearly identical to that of reagent-grade polycarbonate (SI-9†), indicating the lens material's equivalence to the reagent-grade PC. Subjecting this powder to methanolysis with catalyst **La-3f** for 2 h yielded 59% BPA and 61% DMC (Scheme 2), assuming complete PC conversion. This demonstrates equivalent reaction progress between recycled PC



Scheme 2 Methanolysis of PC derived from a post-consumer PC product.

powder from products and reagent-grade PC powder. However, comparing our results with those of other studies using actual products is challenging due to difficulties in rigorously characterizing the diverse products involved.<sup>38</sup> Our demonstration aims to highlight that the pre-treatment-induced reaction acceleration applies beyond virgin polycarbonate.

## Conclusions

Using La complexes with 1,3-dicarbonyl-type ligands, we have demonstrated that the depolymerization of polycarbonate *via* transesterification in methanol proceeds under mild temperature conditions below the solvent boiling point. The structure of the ligand requires a moderate space for activation of the carbonyl moiety in the polymer yet possesses an electron-rich functional group. The results strongly suggest that the Lewis basicity of the formed La complex is also important for high catalytic activity. Optimized complexes achieved high conversion efficiencies of 69% in 4 h and 92% in 8 h, as determined by GC analysis, even at the low temperature of 60 °C. GPC analysis revealed a sharp drop in the molecular weight of polycarbonate immediately after the start of the reaction, indicating that cleavage of ester bonds occurred at random positions in the polymer chain. In addition, the appearance of a peak corresponding to a methanol-insoluble component with a molecular weight of about 2500 during the reaction progress indicates that the polymer-degradation reaction proceeds in a heterogeneous system. The reactivity of depolymerization in such a heterogeneous system is considered to be closely related to the molecular weight distribution. In future studies, we plan to further investigate the changes in molecular weight distribution during the pre-treatment process of polymers and their impact on reactivity, not limited to polycarbonate.

## Experimental

### Preparation of PC powder

PC (20.1 g) in pellet form was added to toluene (2 L) and stirred at room temperature for more than 12 h. The obtained



white suspension was filtered through a stainless steel mesh with an aperture of 500 µm. The solvent was then removed under reduced pressure to obtain a white solid. The white solid was ground in a mortar and dried under vacuum at 80 °C overnight to obtain a white powder (6.6 g). Particle size distribution measurement in methanol confirmed that the prepared PC powder possessed two peaks of 105 µm and 384 µm. This bimodal distribution likely arises from the aggregation of smaller primary particles rather than distinct size classes. Ultrasonic treatment induced a transition from larger aggregates to smaller particles. SEM images further suggest that the primary particles consist of even smaller sub-particles. Particle size distribution and SEM images are available in the ESI (SI-3 and SI-4,† respectively).

### Synthesis of La complexes<sup>28,29</sup>

1,3-Diketones and β-ketoesters were purchased or synthesized according to reported methods. In a typical experiment, a ligand (20 mmol, 10 equiv.) was placed in a 100 mL flask, and a solution of 0.5 M NaOMe in methanol (40 mL) was added with stirring until the ligand dissolved completely. L LaCl<sub>3</sub> (2 mmol) dissolved in methanol (1 mL) was then added drop-wise to the flask with stirring. After stirring for more than 6 h, the precipitated solid was collected by filtration and dried under vacuum at room temperature overnight.

### Methanolysis of PC powder

PC powder (101.7 mg, 0.4 mmol, based on the BPA-CO unit) and a La complex (4 µmol, 1 mol% relative to PC) were placed in a sealed tube reactor. Methanol (4 mL) was added to the reactor, and the atmosphere was replaced with argon. The reactor was then sealed tightly and stirred at 60 °C for 2 h. After this period, the reactor was allowed to cool to room temperature. Decane was added as an internal standard, and the mixture was diluted with methanol. After removing the reaction residue with a membrane filter, the methanol solution was analyzed by GC-FID.

### Data availability

The data supporting this article have been included as part of the ESI.†

### Conflicts of interest

There are no conflicts to declare.

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