Selective Degradation of End-of-Life Poly(lactide) via Alkali-Metal-Halide Catalysis

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The recycling of poly(lactide) (PLA) is studied. Using methanol and alkali-metal-halides as catalyst, PLA is degraded to produce methyl lactate, which is a suitable chemical for the regeneration of PLA. An excellent degree of degradation (conversions: >99%, yields: >99%) is achieved in less than 20 min utilizing microwave heating. Remarkably, PLA goods are successfully converted into methyl lactate.

Nowadays, petrochemical-based plastics have a significant impact on society, which is underlined by the countless applications, the yearly increasing volume of production, and the deep interweavement with human life. However, several issues arise with plastics after the service life ends. In more detail, the current management for end-of-life (EoL) plastic streams is mainly based on energy recovery, (landfill storage), recycling (mechanical and downcycling), and unregulated release to the ecosphere. Finally, these treatments convert chemical functions of the polymers into harmful chemicals, for instance, carbon dioxide, consequently affecting on the anthroposphere (e.g., global warming). Importantly, for fresh polymers the declining fossil resources are required. Moreover, environmental concerns (e.g., high stability against biodegradation, absorption of chemicals) are caused by end-of-life plastic streams, which are not reaching the collecting system, but discarded directly in the ecosphere. An attractive alternative to petrochemical-based plastics is plastics originated from renewables (biopolymers and bio-based polymers), but so far the rate of production is significantly lower. One example from this group is poly(lactide) (1), which is accessible from lactic acid (via polycondensation) or the lactic acid derivative lactide (via ring-opening-polymerization, ROP). Lactic acid is available from biochemical processes originated from CO₂, H₂O, and energy.

Poly(lactide) (PLA) is manufactured in an annual scale of ≈0.2 million tons and has interesting plastic abilities, which enables utilization in a wide range (e.g., food packaging, pharmaceuticals). In contradiction to the majority of fossil-based plastics, poly(lactide) is to some extent biodegradable depending on the conditions and environment and is degraded to, for instance, CO₂; therefore, environmental issues are reduced if PLA is not treated within the current waste management. Nonetheless, the breakdown of PLA in composting plants is regularly not fully accomplishable in the scheduled time frame. Conversely, EoL polylactide can be exposed to incineration for energy recovery. In contrast to petrochemical-based plastics a nearly carbon-neutral manner can be visualized, since carbon dioxide “embedded” in PLA and the carbon dioxide released from EoL PLA are in a narrow section of the timeline.

Moreover, the EoL plastics can be handled by recycling (mechanical recycling or downcycling) to generate products with identical or lower quality, nonetheless after some sequences the plastics will be send to biological degradation or energy recovery, due to loss of quality. Even if several advantages of polymers based on renewables are evident, the production needs cultivable acreage. Noteworthy, by doing so the acreage is blocked and cannot be applied for the cultivation of edibles and cultivation of energy or virgin biotopes have to be transformed to agriculture areas. A possibility to avoid the competitive situation can represent the chemical recycling composed of depolymerization and polymerization reactions. Initially, the depolymerization/degradation enables the conversion of the EoL plastic/polymer to monomers or useful compounds. Subsequently, the monomers are utilized as feed in polymerizations to create new polymers. Advantageously, compared to mechanical recycling/downcycling the quality and abilities of the plastics can be adjusted at the polymerization step or the plastic formation stage and is independent of the quality of the EoL plastic. Noteworthy, cultivable acreage required for PLA-plastics can be minimized. A number of investigations have been reported including the hydrolysis, alcoholysis, and (catalytic) pyrolysis or thermal degradation of PLA. For instance, the alcoholysis of PLA, especially the methanolysis, represents an interesting approach toward chemical recycling (Scheme 1). The EoL PLA (1) is degraded with the degradation reagent methanol and with the support of a catalyst to form as main chemical methyl lactate (2). Subsequently,
methyl lactate (2) can be transformed to compound 3 (lactide), which is a fundamental compound in the industrial production chain of PLA (Scheme 1).[63–70] As second product methanol is produced, which can be recycled for further degradation; therefore, reduced amounts of waste are generated and the whole process is made highly atom efficient. With respect to the degradation process, a number of organocatalysts have been proven to be active, but often long reaction times and “costly” organocatalysts are required; hence, improvements are needed. For example, ionic liquids (ILs) have been established as catalysts.[56,71–75] It was supposed that the alcohol is activated by the anion and the carbonyl function by the cation. In this context, we wondered if straightforward alkali metal halides can mimic the action of ILs.[76–78] Furthermore, alkali metal salts can have benefits regarding “Green Chemistry” and SDGs considerations.[79] For instance, we recently studied the potential of the organocatalyst 4-N,N-dimethylaminopyridine (DMAP).[57] However, low TOFs (amount of substance of 2/(amount of substance catalysts) $h^{-1} \approx 178 h^{-1}$) were observed and moreover to access DMAP several synthetic steps, which create waste, are required starting from fossil resources.[80] In this context, we account our studies on microwave-assisted degradation of EoL PLA catalyzed by alkali metal halides (Scheme 2).

Initially, as model for optimization of the degradation reaction conditions a transparent plastic cup containing poly(lactide) (1a, $M_n = 57 \ 500 \ g \ mol^{-1}$, $M_w = 240 \ 800 \ g \ mol^{-1}$, $D = 4.2$, PLLA) as major component was investigated (Table 1). Small pieces of the PLA 1a (1.35 mmol with respect to the monomer unit, implying that 1a contains 100% of PLA), an excess of methanol (23.1 equivalents with respect to the monomer unit of 1a) and catalytic amounts of KF (5.0 mol% with respect to the monomer unit of 1a) were added to a microwave glass vial. After sealing the vial the stirred mixture was reacted for 10 min at 180 °C applying microwave heating (MW) (Table 1, entry 1). During

![Scheme 1. Potential life cycle of PLA.](image)

![Scheme 2. Chemical recycling concept for PLA (option A: conversion of 2 to the industrial relevant building block 3, which can be used as precursor for the synthesis of PLA via ring opening polymerization; option B: direct polymerization of 2 via transesterification).](image)
heating/degradation process the PLA was completely dissolved resulting in the formation of a homogeneous solution. After that, an aliquot of the mixture was added CDCl3 for determination of the yield by 1H NMR spectroscopy. The existence of the desired degradation product 2 was indicated by the appearance of a singlet at 3.76 ppm. The signal corresponds to the methyl ester functionality of 2. Moreover, the occurrence of 2 was confirmed by a doublet at 2.89 ppm (3 \( J_{HH} = 5.20 \text{ Hz} \)), which is in alignment with the \(-\text{CHOH}\)-functionality. Moreover, a doublet at 1.39 ppm (3 \( J_{HH} = 6.91 \text{ Hz} \), 3 \( J_{HH} = 5.20 \text{ Hz} \), \( \text{CH}_3\text{CHOH}\)-functionality) were monitored. Relation of the integral of the \( \text{CH}_3\text{CHOH}\)-function to the integral of the \( \text{CH}_3\text{CHOR}\)-function of the polymer/oligomer leftover (≈5.17 ppm) revealed nearly complete transformation of 1a to 2, since a yield of 98% (2) was calculated, and showed excellent catalyst abilities of the potassium fluoride (Table 1, entry 1). Importantly, no addition of extra solvents was necessary to perform the degradation, which is beneficial with respect to the Green Chemistry Principles.\[^{[8]}\] Interestingly, half of the catalyst loading exhibited the generation of 2 in an excellent yield (Table 1, entry 2).

Table 1. Degradation of 1a using alkali-metal-halide-catalysis—optimization of reaction conditions.

| Entry\[^{[8]}\) | Catalyst | Catalyst loading [mol\%] | MeOH [equiv.] | \( T \) [°C] | \( t \) [min] | Yield 2 [%]\[^{[8]}\) |
| --- | --- | --- | --- | --- | --- | --- |
| 1 | KF | 5.0 | 23.1 | 180 | 10 | 98 |
| 2 | KF | 2.5 | 23.1 | 180 | 10 | 99 |
| 3 | KF | 5.0 | 23.1 | 180 | 5 | 99 |
| 4 | KF | 5.0 | 23.1 | 180 | 60 | >99 |
| 5 | KF | 1.0 | 23.1 | 180 | 10 | 98 |
| 6 | KF | 0.5 | 23.1 | 180 | 10 | 68 |
| 7 | – | – | 23.1 | 180 | 10 | <1 |
| 8\[^{[9]}\) | KF | 5.0 | 11.6 | 180 | 10 | >99 |
| 9\[^{[a]}\) | KF | 5.0 | 5.8 | 180 | 10 | 23 |
| 10\[^{[b]}\) | KF | 5.0 | 1.0 | 180 | 10 | <1 |
| 11 | KF | 5.0 | 23.1 | 160 | 10 | 99 |
| 12 | KF | 5.0 | 23.1 | 140 | 10 | 98 |
| 13 | KF | 5.0 | 23.1 | 120 | 10 | n.d.\[^{[c]}\) |
| 14 | LiCl | 5.0 | 23.1 | 180 | 10 | 50 |
| 15 | NaF | 5.0 | 23.1 | 180 | 10 | n.d.\[^{[c]}\) |
| 16 | NaCl | 5.0 | 23.1 | 180 | 10 | n.d.\[^{[c]}\) |
| 17 | NaI | 5.0 | 23.1 | 180 | 10 | 49 |
| 18 | KCl | 5.0 | 23.1 | 180 | 10 | n.d.\[^{[c,d]}\) |
| 19 | KBr | 5.0 | 23.1 | 180 | 10 | n.d.\[^{[c,d]}\) |
| 20 | KI | 5.0 | 23.1 | 180 | 10 | 64 |
| 21\[^{[e]}\) | KF | 2.5 | 23.1 | 180 | 20 | 93 |

\[^{[a]}\) Conditions: 1a (transparent PLA cup), 1.35 mmol with respect to the monomer unit, it is presumed that 1a is composed of 100% of PLA), catalyst (0–5 mol\%, 0–0.0675 mmol with respect to the monomer unit of 1a), MeOH (5.8–23.1 equiv. with respect to the monomer unit of 1a), THF (0–2.0 g), temperature: 120–180 °C (microwave heating), time: 5–60 min; \[^{[b]}\) A homogeneous solution was noticed. The yield of 2 bases on 1H NMR spectroscopy; \[^{[c]}\) Addition of THF (1.0 g); \[^{[d]}\) Addition of THF (2.0 g); \[^{[e]}\) A precipitate was formed; \[^{[f]}\) 8.1 mmol scale of 1a.

Moreover, the reaction was extended to 60 min at 180 °C to investigate the stability of product 2 under reaction conditions, but no decomposition reactions were noticed (Table 1, entry 4). From this starting point the loading of KF was studied (Table 1, entries 5–7). Decreasing the KF load toward 1.0 mol% the degradation product 2 was still formed in excellent yield (98%). Noteworthy, from the presented catalyst activity a turnover frequency (TOF) of ≈588 h\(^{-1}\) was calculated, which is among the highest TOFs reported so far. In case of a further decrease to 0.5 mol% KF a yield of 68% of 2 after cooling to room temperature was noticed (Table 1, entry 6) (TOF ≈816 h\(^{-1}\)). For instance, with our recently published DMAP system lower TOFs have been obtained (≈178 h\(^{-1}\)).\[^{[77]}\) Notable, in the absence of KF no formation of 2 was detected by NMR, therefore KF is essential for the degradation process (Table 1, entry 7). Following, the effect of the quantity of methanol on the degradation was studied (Table 1, entries 8–10). Noteworthy, in case of lower methanol loadings (1.0–11.6 equiv.) tetrahydrofuran was used as (co)solvent. Still excellent activity (>99% yield) was observed in the presence of 11.6 equiv. MeOH, while a decrease to 5.8 equiv. resulted in a reduced yield of 23%. In addition, the
impact of the temperature was studied (Table 1, entries 11–13). A yield of 98% for 2 was realized at a decreased temperature of 140 °C, while after 10 min at 120 °C a precipitation of 1 after cooling to room temperature was noted. Finally, a test of a range of different alkali-metal-halides was performed (Table 1, entries 14–20). Moderate-to-good yields (49–64%) were realized with 5 mol% of lithium chloride, sodium iodide, and potassium iodide. Afterward, the degradation of PLA was performed on an 8.1 mmol scale under optimized conditions (2.5 mol% KF, 23.1 equiv. methanol, 180 °C, 20 min, MW). In this regard, product 2 was obtained in an excellent yield of 93% as monitored by 1H NMR (Table 1, entry 21).

Moreover, methyl lactate was isolated after performing the reaction for 20 min at 180 °C. In more detail, the mixture was transferred to distillation apparatus and 2 and methanol were distilled off from the catalyst under reduced pressure, to circumvent side/back processes (polymerization). Subsequently, the mixture of 2 (b.p. ≈ 144 °C) and methanol (b.p. ≈ 65 °C) was separated by distillation. 2 was isolated in 134 wt% (based on the weight of 1a) or 92% isolated yield (presumption that 1a contains 100% PLA) (Table 2, entry 1).

Noteworthy, the collected methanol (19.7 equiv., 89%) can be reused/recycled for next degradation. In addition, we investigated the reuse of the KF (Figure 1). In this regard, the reaction conditions applied were in accordance to the experiment in Table 1, entry 21. After 20 min (180 °C) the reaction was cooled to room temperature and a sample was subjected to 1H NMR. From the reaction mixture methanol and methyl lactate were removed in vacuum. The catalyst containing residue in the flask was transferred with the aid of methanol to a vial.

### Table 2. Potassium fluoride-catalyzed degradation of PLA goods.

| Entrya) | Product | $M_n$ [g mol$^{-1}$]b) | $M_w$ [g mol$^{-1}$]b) | $D^b$ | Yield of 2 [wt%]c) | Yield of 2 [%]d) |
|---------|---------|------------------------|------------------------|-------|---------------------|-----------------|
| 1       | Transparent cup (1a) | 57 500 | 240 800 | 4.2 | 134 | 92 |
| 2       | Transparent disposable food box (1b) | 43 600 | 236 000 | 5.4 | 104 | 72 |
| 3       | Transparent Sushi box cover (1c) | 54 700 | 212 700 | 3.9 | 87 | 60 |
| 4       | Transparent plastic sheet (1d) | 150 400 | 262 000 | 1.7 | 83 | 57 |
| 5       | Transparent bottle (1e) | 99 100 | 254 900 | 2.6 | 141 | 97 |
| 6       | Drinking straw with green strips (1f) | 112 100 | 239 900 | 2.1 | 70 | 48 |
| 7       | Disposable fork with talcum powder (1g) | 59 000 | 124 100 | 2.1 | 62 | 43 |
| 8       | Lid for espresso mugs (contains ≈ 20–30% talcum powder) (1h) | 56 100 | 177 500 | 3.2 | 78 | 54 |
| 9       | Black lid for coffee mugs (used) (1i) | 75 400 | 241 100 | 3.2 | 72 | 50 |
| 10i)    | Black lid for coffee mugs (used) (1i) | – | – | – | 92 | 64 |
| 11      | Sushi box (black base) (1j) | 80 300 | 148 400 | 1.8 | 84 | 58 |
| 12      | Pink ice cream spoon (1k) | 57 600 | 133 000 | 2.3 | 105 | 72 |
| 13i)    | Yellow ice cream spoon (used) (1k) | – | – | – | 122 | 92 |
| 14      | Coffee mug (paper with PLA coating) (1l) | 86 400 | 221 000 | 2.6 | 4 | 3 |

a) Conditions: 1a–1l (583.2 mg, 8.1 mmol with respect to the monomer unit), KF (11.8 mg, 2.5 mol%, 0.202 mmol with respect to the monomer unit of 1), MeOH (6.0 g, 186.3 mmol, 23.1 equiv. with respect to monomer unit of 1), temperature: 180 °C (microwave heating), time: 20 min; b) $M_n$, $M_w$, and $D$ were obtained from GPC measurements; c) The mass of 2 was linked to the mass of the initial PLA good. Yield wt% = ($m$(2)/$m$(1)) × 100; d) The amount of substance of 2 was linked to the amount of substance of the initial PLA good (presumption: PLA good contains 100% of PLA. Yield % = ($n$(2)/$n$(1)) × 100; e) Used with coffee, milk, and sugar. The PLA product was not cleaned before degradation; f) Contaminations: coffee, sugar, and milk. The PLA good was dried before degradation, but not purified.
containing a new portion of end-of-life PLA. Afterward, the degradation was carried out at a temperature of 180 °C for 20 min. Until the third run no change in the reaction outcome was monitored, while afterward the yield of 2 drops to 54% in the fifth run. Nevertheless, a reuse/recycling of potassium fluoride is possible, which is in contrast to our earlier reported DMAP system.\[58\] The distilled-off mixture of methanol and methyl lactate from the catalyst recycling experiments were combined and after removal of methanol methyl lactate was isolated in a total yield of 78%. In addition, the time–yield plot relationship was recorded at 140 °C (Figure 2). The sample taken after 120 s was investigated by gel permeation chromatography (GPC) analysis revealing the disappearance of long chain polymers.

With respect to the underlying degradation mechanism we assume that the alkali metal halide can act as dual catalyst in accordance to ionic liquid catalysts applied in the methanolysis of polycarbonate (Scheme 3).\[56,71–75\] The potassium cation can function as Lewis acid and coordinates the oxygen of the carbonyl function and by doing so the carbonyl carbon is activated and methanol can attack (oxygen atom of the methanol) the group as nucleophile. The nucleophilicity of the methanol can be increased by the interaction of the fluoride anion with the proton of methanol via hydrogen bonding. Based on that, the PLA chain is dissected and results in the formation of smaller chains and by repeating the process as final product methyl lactate is formed.

Afterward, a selection of PLA products was directed to degradation (Table 2, entries 1–14). Besides the transparent PLA cup further transparent products such as a food box (1b), a cover of a Sushi box (1c), a PLA sheet (1d), and the body of a bottle (1e) were studied (Table 2, entries 2–5). Methyl lactate was isolated in yields of up to 141 wt% (the yield base on the weight of the plastic, which not only contains the polymer PLA. The plastic also contains an unknown amount of additives, etc.; therefore, the precise amount of PLA is not known) or up to 97% (presumption that the plastic contains 100% PLA). Note-worthy, if the PLA product (drinking straw with green stripes (1f)) contains dyes, the yield (70 wt%, 48%) is to some extent

**Figure 1.** Degradation of PLA: catalyst recycling experiments [Conditions: 1a (583.2 mg, 8.1 mmol with respect to the monomer unit), KF (first run: 11.8 mg, 2.5 mol%, 0.202 mmol with respect to the monomer unit of 1a), MeOH (6.00 g, 187.2 mmol, 23.1 equiv. with respect to the monomer unit of 1a), temperature: 180 °C (microwave heating), time: 20 min. Second run catalyst leftover from first run; third run catalyst leftover from second run; fourth run catalyst leftover from third run; fifth run catalyst leftover from fourth run; the yield was determined by 1H NMR.

**Figure 2.** Degradation of PLA: time–yield plot [Conditions: 1a (583.2 mg, 8.1 mmol with respect to the monomer unit), KF (5 mol%, 0.404 mmol with respect to the monomer unit of 1a), MeOH (6.00 g, 187.2 mmol, 23.1 equiv. with respect to the monomer unit of 1a), temperature: 140 °C (microwave heating); the yield was determined by 1H NMR.

**Scheme 3.** Tentative degradation mechanism of end-of-life PLA in the presence of potassium fluoride.

**Scheme 4.** Scale-up degradation of end-of-life PLA performed with conventional heating in an autoclave [Conditions: 1m (102.7 mmol), MeOH (2.47 mol; 24.0 equiv. with respect to the monomer unit of 1m, potassium fluoride (5.13 mmol, 5.0 mol% with respect to the monomer unit of 1a), temperature: 140 °C (conventional heating), time: 60 min].
lower (Table 2, entry 6). Moreover, PLA goods containing additive talcum powder (≈20–30%) were tested (Table 2, entries 7–8). Good yields of 62–78 wt% or 43–54% were realized for a disposable fork (1g) and a lid for espresso mug (1h). Note, taking the amount of talcum powder into account the yields of methyl lactate are comparable to the transparent PLA products.
Next, black PLA products were studied (Table 2, entries 9–11). Yields of 2 in the range of 72–84 wt% or 50–58% were obtained. Remarkably, a used coffee mug lid (1), which was contaminated with coffee/milk/sugar leftovers, even a higher yield of 92 wt% or 64% was noticed (Table 2, entry 10). An analogous observation was made for spoons 105 wt% or 72% versus 122 wt% or 92% (Table 2, entries 12–13). Finally, the degradation of a disposable coffee mug composed of paper and a PLA coating was studied (Table 2, entry 14). However, a low yield of 2 of 4 wt% or 3% was noticed, since the paper component is the main component in the coffee mug.

Besides, for plastic cups (1a) a scale-up reaction was carried out (Scheme 4). In more detail, the reaction was performed in accordance to the conditions stated in Table 1 entry 12 by charging an autoclave (160 mL) with 7.4 g of 1a. The reaction was carried out at 140 °C (conventional heating) and the reaction time was extended to 30 min. Following the standard purification/isolation protocol 8.63 g of 2 was attained, which relates to a yield of 81%. Moreover, the optical rotation of the isolated 2 was measured to get information of the optical purity of the product. An enantiomeric excess of ≈−93% (of (−)-methyl l-lactate) was obtained, which is in line with the starting polymer and revealed almost no racemization.

In addition, we studied the potassium fluoride catalyzed degradation of PLA (1a) accompanied by different end-of-life plastics containing carbonyl functionalities, which can be seen as a fraction of the waste streams. Recently, we have demonstrated the abilities of alkali metal halides as catalyst in the depolymerization of end-of-life poly(bisphenol A carbonate) (3). Poly carbonate 3 was converted in the presence of methanol and potassium fluoride as catalyst to the building blocks bisphenol A and dimethyl carbonate, which can be reused as starting materials for fresh polycarbonate.[82,83] Since the reaction conditions are comparable, we investigated the combination of the degradation of a mixture of (solvent-assisted mixing) end-of-life poly(lactide) 1a and the depolymerization of end-of-life poly(bisphenol A carbonate) 3 (Scheme 5). In more detail, to a mixture of 1a (5.4 mmol) and 3 (2.7 mmol) was added methanol (124.9 mmol; 23.1 equivalents with respect to the monomer unit of 1a and 46.3 equiv. with respect to the repeating unit of 3) and catalytic amounts of potassium fluoride (0.135 mmol, 2.5 mol% with respect to the monomer unit of 1a, 5.0 mol% with respect to the monomer unit of 3). The mixture was stirred at 160 °C (microwave heating) for 20 min. After that, the mixture was analyzed by 1H NMR revealing the formation of degradation products methyl lactate derived from PLA (NMR yield: 97%) and bisphenol A (NMR yield: 86%) and dimethyl carbonate derived from polycarbonate 3.[85,86] Besides, the degradation products can be easily separated by distillation. First, methyl lactate, dimethyl carbonate, and methanol were quickly distilled off from the catalyst and 4 under vacuum. The residue was recrystallized from chloroform to isolate 4 as colorless crystals in 70% yield. Subsequently, the mixture of 2, 5, and methanol was separated by distillation. Keeping the temperature below 100 °C 5 and methanol were collected, while 2 remains in the distillation flask and was isolated in 91% yield. In consequence, a side-by-side degradation of PLA and poly(bisphenol A carbonate) with the same catalyst is feasible. Moreover, poly(ethylene terephthalate) (PET) (6) or polyamide 6 (Nylon 6) (7) was added to the mixture of poly(lactide) (1a), methanol, and catalytic amounts of potassium fluoride (Scheme 5). In both experiments, the degradation of 1a was achieved and 2 was isolated in 79% and 54%, respectively. Conversely, the PET and Nylon 6 were not depolymerized and were recovered from the reaction mixture.

In summary, we have installed an easy-to-adopt degradation method as an initial module for the chemical recycling of EoL poly(lactide) (PLA). In more detail, the degradation was performed with methanol as degradation reagent to obtain methyl lactate as product. Noteworthy, as catalyst simple alkali metal halides are required to realize excellent yields of 2 (>99%) within short reaction times (5 min) under microwave heating. Applying potassium fluoride as catalyst turnover frequencies up to ≈588 h⁻¹ were obtained. The principle of operation of the concept was proven in the degradation of a variety of PLA goods.

Noteworthy, the produced methyl lactate can be the basis for the synthesis of lactide, the starting material for new PLA as reported in the literature. Finally, the degradation of PLA has the potential to add some value to the UN’s Sustainable Development Goals (SDGs), e.g., SDG 13 (responsible use of natural resources), SDGs 2,3 (food security, land use), SDGs 9,11 (clean/green technologies, waste management/urban mining), and SDG 12 (supply chain, more efficient, and less consumptive technologies, recycling, waste streams). Upcoming investigations will focus on the advance of the degradation and the establishment of a continuous method of operation.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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[81] The TOF was calculated by using the H NMR yield: $\text{TOF} = \frac{\text{mol product}}{(\text{mol catalyst} \times \text{h})}$. The TOF was calculated using the yield of 2 after 10 min, because the determination of the TOF at a yield of 50% was not possible, due to technical limitations.

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