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Depolymerization of Waste Plastics to Monomers and Chemicals Using a Hydrosilylation Strategy Facilitated by Brookhart’s Iridium(III) Catalyst

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ABSTRACT: Plastic waste management is a major concern. While the societal demand for sustainability is growing, landfilling and incineration of waste plastics remain the norm and methods able to efficiently recycle these materials are desirable. Herein, we report the depolymerization, under mild conditions, of oxygenated plastics in the presence of hydrosilanes with the cationic pincer complex [Ir(PCP)H(THF)][B(C₆F₅)₄] (PCP = 1,3-(tBu₂P)₂C₆H₃) as catalyst. The iridium catalyst, with a low loading (0.3-1 mol%), proves selective toward the formation of silyl ethers or the corresponding alkanes depending only on the reaction temperature. It is noteworthy that the depolymerization of real household waste plastics such as PET (from plastic bottles) and polylactic acid (PLA) from 3D printer filaments is not altered by the presence of dye or other plastic’s additives.
Introduction

Plastics are ubiquitous in our modern society. Thanks to their lightness, their ease to be molded and their low costs, these polymers have forced their use in various applications such as electronics, packaging, medicine, etc. It is estimated that 4-6 % of the annual production of fossil resources are used to supply the 335 million tons of plastics consumed worldwide.\(^1\) In consequence, millions of metric tons of waste are generated every year, which are either incinerated or deposited in landfills. This open cycle poses ecological problems, causing pollution issues and emissions of large quantities of the greenhouse gas CO\(_2\).\(^2\) The increasing awareness of the environmental issues associated with plastic wastes generates a public demand for sustainability and renewability in the plastic industries. In this context, a conglomerate of industries and associations \(^3\) has called for the implementation of a circular economy of plastics lying on three main axes: drastically reducing the leakage of wastes in the environment, decoupling plastic production from fossil resources and up-taking recycling processes.\(^4\)

The reuse of manufactured polymers remains challenging because of the high density of chemical functionalities in the polymer matrix, the presence of additives in plastics and/or the blend of different materials. Besides incineration and landfilling, physical and chemical recycling technologies are appealing approaches to improve the conservation of carbonaceous materials.\(^5\) The former processes consist of melting and remodeling the plastic, although the second generation plastics have more limited usages because of the presence of impurities. In contrast, chemical processes can afford the recovery of pure chemicals, useful for the production of recycled virgin quality plastics as well as new chemicals.\(^6\) In this respect, the development of new depolymerization systems recently emerged in the scientific community.\(^4-6\) Classical
aminolysis, hydrolysis or transesterification reactions have been widely repurposed in the catalytic depolymerization of polyesters\textsuperscript{7-9} and polycarbonates.\textsuperscript{10,11} Alternatively, the cleavage of C–O bonds can be achieved through reduction methods.\textsuperscript{12} Robertson and co-workers tackled the recycling of waste plastics via the catalytic hydrogenation of oxygenated polymers with the soluble Ru(II) pincer complexes 1 and 2 (Scheme 1).\textsuperscript{13} Selective deconstruction of a wide range of polycarbonates and polyesters into the corresponding valuable alcohols or carboxylic acids was achieved with excellent yields. Hydrogenation of the polymers proceeded in relatively harsh conditions (54 atm H\textsubscript{2}, 160°C). Taking advantage of the kinetically and thermodynamically favored hydrosilylation of carbonyl functions, our group recently reported the efficient catalytic reduction of a range of natural and manufactured oxygenated polymers, at room temperature, with B(C\textsubscript{6}F\textsubscript{5})\textsubscript{3} (3) as a catalyst.\textsuperscript{14} Nonetheless, this Lewis acid catalyst is incompatible with a number of solvents and its price is close to precious metal (B(C\textsubscript{6}F\textsubscript{5})\textsubscript{3} \textasciitilde 110 €/mmol, [Ir(COD)Cl]\textsubscript{2} \textasciitilde 40 €/mmol of Ir, COD = 1,5-cyclooctadiene [Sigma-Aldrich]). It is also poorly selective, and demands high loadings (2-10 mol%). These drawbacks call for more stable, efficient catalysts.
Scheme 1: Catalysts for the reductive depolymerization of plastics and polymers

The cationic Ir(III) complex 4 is a well-known hydrosilylation catalyst exhibiting similar chemical behavior as borane 3$^{15}$ (Scheme 1). The electrophilic activation of hydrosilanes by Lewis acid 4 favors reduction of oxygenated functions such as carbonyl$^{16}$ or carboxyl$^{17}$ groups as well as ethers,$^{18,19}$ leading to the corresponding silylated alcohols or alkanes. $^{20}$ While complex 4 proved to be very efficient in hydrosilylation reactions with organic substrates, its utilization in the reduction of poly-functional materials is scarce$^{21,22}$ and Brookhart et al. reported the unique example of depolymerization of polyethylene glycol (PEG) into silylated ethylene glycol with 4 within 4 h at 65°C.$^{18}$ Here we report the efficient use of the Brookhart’s complex in the catalytic depolymerization of polyesters and polycarbonates with hydrosilanes. Silyl ethers or hydrocarbons are formed selectively by controlling the temperature of the reaction, thus
demonstrating the wide applicability of the iridium(III) catalyst in the depolymerization of oxygenated polymers.

**Results and discussion**

One of the major drawbacks in the chemical treatment of polymers is generally their insolubility in classical organic solvents which can prevent reactions or considerably slow down their kinetics. The first attempt at hydrosilylation of plastics was thus undertaken with low melting point polymers, such as the aliphatic polycaprolactone (PCL) (mp = 60°C), to form homogeneous or biphasic solutions by heating. Surprisingly, at room temperature, insoluble commercial pellets of PCL (diameter ≈ 0.4 cm) rapidly reacted in presence of the mild reductant triethylsilane (Et₃SiH, 3 eq) and of the iridium catalyst 4 (0.3 mol%) in chlorobenzene. After 2 hours, no residual solid remained, ¹H NMR and DEPT 135 ¹³C NMR as well as GC-MS analyses of the clear solution revealed a complete and selective depolymerization of PCL into the silylated 1,6-hexanediol (5), obtained as the unique product in quantitative yield (Scheme 2, eq. 1). Compound 5 was isolated pure in a preparative scale (94% from 3 mmol of PCL).

Successful depolymerization of PCL under these mild conditions led us to consider a variety of aliphatic and aromatic polyesters which differ by the nature of the spacer between the carboxyl groups and with higher melting points than PCL. Though PCL, poly(dioxanone) (PDO), and poly(lactic acid) (PLA) are biodegradable polymers, their chemical recycling remain relevant to conserve the raw carbonaceous materials as well as to avoid, upon degradation, the release in the environment of CO₂ (or CH₄ in anaerobic conditions)²³,²⁴ and the additives present in the plastic matrix. PDO was found inert in reduction conditions used for PCL but further heating to 65 °C proved sufficient to successfully depolymerize PDO into a mixture of
the two silylethers **DEG-Si** and **EG-Si** along with gaseous ethane and the siloxane **Et₃SiOSiEt₃** (Scheme 2, eq. 2). While **DEG-Si** was the major product (82%), a complete selectivity could not be achieved suggesting that the hydrosilylation of **PDO** and **DEG-Si** are concomitant. Formation of **EG-Si** was reached in excellent yield (89%) under forcing conditions, when using an excess of the hydrosilane (10 equiv.) with higher temperature and prolonged reaction time (90 °C, 48 h) (Scheme 2, eq. 3).

**Scheme 2**: Reduction of **PCL**, **PDO** and **PLA** into silylated alcohols with **Et₃SiH** and 4.
A similar behavior has been observed with PLA (Scheme 2, eq. 4). Although the depolymerization of PLA required longer reaction times than PDO (60 h vs 48 h), the reaction occurs in presence of the iridium complex 4 (0.5 mol%) and Et₃SiH (3 equiv.) to yield the silylated propylene glycol PG-Si (64%) and the over-reduced product nPrOSi (31%). As for the deconstruction of PDO, the reduction of PLA and PG-Si seems to be kinetically competitive and PG-Si and nPrOSi were observed simultaneously during the progressive reduction of PLA (see SI, section II.5). Nonetheless, 1 mol% of the Ir catalyst and an excess of the hydrosilane (10 equiv.) were sufficient to yield to nPrOSi in 92% yield along with ethane and Et₃OEtSiEt₃ at 90 °C (Scheme 2, eq. 5). Interestingly, strictly similar results have been obtained from the treatment of a soluble ground powder of commercial PLA as well as large insoluble pieces of coloured PLA from 3D printer plastic (white, blue and red; ≈0.4x0.3x0.3 cm), showing the high tolerance of the Ir catalyst 4 towards the contaminants present in these PLA plastics (dyes and additives).

Despite the structural similarities between polyglycolic acid (PGA) and poly-3-hydroxybutyric acid (P3HB) with PLA, these materials were left untouched in the presence of 4 and Et₃SiH regardless the quantity of reductant, the temperature and the reaction times (up to 2 weeks) (Scheme 2, eq. 6). These distinct behaviours in hydrosilylation are obviously not related to different steric congestions nor to the solubility of the polymers (soluble PLA vs insoluble PCL).

We then investigated the depolymerization of PES (polyethylene succinate), an aliphatic biodegradable copolymer of ethylene glycol and butanedioic acid. In presence of excess Et₃SiH (6 equiv.) and a 0.3 mol% 4, PES was readily depolymerized at 65 °C to the silylated butanediol 6 and ethylene glycol EG-Si (Scheme 3, eq. 7). Interestingly, the disilylether 6 was produced almost quantitatively (99%). EG-Si was obtained in lower yield (23%) due to its further
reduction to ethane and siloxane \( \text{Et}_3\text{SiOSiEt}_3 \). PET from commercial Dacron© fibers or small chunks of Evian© bottles were finely ground and dried. The powders were then converted at 70°C, using slight excess \( \text{Et}_3\text{SiH} \) (6 equiv.) and 1 mol% of the iridium complex 4, into two silyl ethers: silylated 1,4-benzenedimethanol (BDM-Si) and EG-Si in 63% and 48% yield (Scheme 3, eq. 8).

Scheme 3: Reductive depolymerization of PES, and PET with \( \text{Et}_3\text{SiH} \) and catalyst 4

Ethane and \( \text{Et}_3\text{SiOSiEt}_3 \) were also present as products of the reduction of EG-Si and prolonged heating (2 weeks at 90 °C) led to para-xylene (1,4-Me-benzene) and ethane. The formation of such silyl ethers is attractive as these products can be used as sources of alkoxide groups in Ullman’s coupling reactions to prepare ethers.\textsuperscript{25-28} Alternatively, the useful alcohols could be obtained through hydrolysis of the silyl ethers. In the case of PET, hydrolysis of BDM-Si
affords the diol 1,4-benzenedimethanol (BDM) which is a valuable building block for the production of pesticides, perfumes or dyes (see SI, section IV.1). \(^{29,30}\)

Interestingly, the reduction of PET required harsher conditions than PLA with the present hydrosilylation system, which is consistent with the chemical behavior noted by Robertson and co-workers under hydrogenation conditions.\(^{13}\) However, we observed a different behavior with the organoborane B(C\(_6\)F\(_5\))\(_3\) (3) and 16 h were needed to depolymerize PLA with a slight excess of Et\(_3\)SiH and 5 mol% 3 at room temperature, while the same reaction was achieved within 3 h with PET.\(^{14}\) As already observed with natural polymers,\(^{22}\) catalyst 4 proved less reactive than B(C\(_6\)F\(_5\))\(_3\) requiring longer reaction times and higher temperatures for both PLA and PET. Nevertheless, B(C\(_6\)F\(_5\))\(_3\) in presence of excess silane favored rapid (24 h) cleavage of all the C-O bonds at room temperature with formation of alkanes and siloxane as the sole products. The iridium catalyst 4 exhibits a higher selectivity allowing the conservation of functionalities, such as silylethers. Moreover, when 5 mol% of the borane 3 are required to treat PLA, the iridium catalyst completed the deconstruction of this polymer with only 0.5 mol% demonstrating a higher stability.

The successful reductive depolymerization of polyesters with 4 and Et\(_3\)SiH led us to consider the reduction of more oxygenated polymers. The reduction of polycarbonates is of interest as organic carbonates are classically more difficult to reduce, compared to esters, because of the delocalization of the oxygen lone pairs over the CO\(_3\) group.\(^{31}\) Catalytic reductive hydrosilylation of polycarbonates (and plastics in general) was only reported in 2015 by our group with the organocatalyst B(C\(_6\)F\(_5\))\(_3\), \(^{14}\) while Robertson and co-workers reported in 2014 the catalytic hydrogenation of polycarbonates (and polyesters) with Ru(II) complexes (\textit{vide supra})\(^{13}\).
We first considered polypropylene carbonate (PPC) viewed as the assemblage of the two monomers, propylene oxide and CO$_2$. Surprisingly, reductive depolymerization of PPC using the hydrosilylation conditions of PLA (0.5 mol% of 4 and 4 equiv. Et$_3$SiH) at 65 °C proceeded more rapidly and more selectively than what was observed with PLA (3 h versus 60 h) affording MeOSiEt$_3$ and disilylether PG-Si in quantitative yield (Scheme 4, eq. 9). MeOSiEt$_3$ results from the reduction of the carbonate groups and no other by-products, such as Et$_3$SiOSiEt$_3$, CH$_4$ or "PrOSi, were detected. The selectivity of the reaction indicated that the hydrosilylation of the polycarbonate was faster than deoxygenation of PG-Si (see scheme 4). This difference in reactivity could be exploited for the selective depolymerization of a mixture containing pieces of PPC (insoluble) and a fine powder of PLA (soluble), suspended in chlorobenzene. In contrast to what expected from their respective solubility, PPC reacted faster than PLA. The $^1$H and $^{13}$C NMR spectra unveiled the formation of PG-Si from PPC which rapidly disappeared in the medium while soluble PLA was left untouched (see SI, section V.3). This comparative experiment highlights that the reactivity of polymers is not necessarily related to their solubility. While the reduction of carbonyl groups usually follows the order of reactivity ketones > esters > carbonates, Brookhart’s catalyst 4 exhibits a different reactivity. In fact, when a mixture of propylene carbonate and rac-lactide are exposed to 1 mol% 4 and an excess Et$_3$SiH, the monofunctional carbonate is reduced before the lactide monomer (see SI, section V.2). The selective hydrosilylation of PPC over PLA hence translates a difference in reactivity between monofunctional carbonates and esters. Yet, this observation should not be taken as a general rule (vide infra).
Scheme 4: Selective reduction of polycarbonates to their corresponding silylated alcohols with the 4/hydrosilane system

Depolymerization of PC-BPA under conditions similar to PPC is slower (6 h vs 3 h) and afforded the disilylated phenol BPA-Si in excellent yield (88%) (Scheme 4, eq. 10). MeOSiEt₃ was not detected by NMR. Yet, evolution of gaseous CH₄ was observed by ¹H NMR (in a closed J-Young NMR tube) along with Friedel-Craft-like products in the form of methylbisphenol derivatives (see SI, section II.9). Of interest, hydrolysis of the hydrosilylation products from
**PPC and PC-BPA** afforded the corresponding alcohols, propylene glycol and BisPhenol A (BPA), respectively. They correspond to the monomers of the original polymers. For example, BPA was obtained in 88 % yield with 2.5 eq. of hydrated tetrabutylammonium fluoride (TBAF) in THF within 12 h (Scheme 4, eq. 11). An attractive alternative to TBAF involves the use of sodium hydroxide in methanol and water for the hydrolysis of silyl ethers. Yet, hydrolysis with sodium hydroxide proved inefficient when applied to triethylsilyl ethers. In fact, such transformation requires more Lewis acidic silicon derivatives such as alkoxyisilane ([RO]₃SiH). This finding led us to consider the silicon industrial waste 1,1,3,3-tetramethyldisiloxane (TMDS) as a potential reductant. Treatment of 1 mmol of PC-BPA with TMDS (4 eq) and 4 (0.5 mol%) led to the formation of a gel after 12 h at 65 °C. Notably, no Friedel-Craft product could be observed in the hydrosilylation products using TMDS. Hydrolysis of the crude mixture with NaOH in water/methanol enabled the formation of the monomer BPA in 83% yield, after 2 h (Scheme 4, eq. 12).

TMDS being a cost-efficient source of hydrides, its potential in the reductive depolymerization of plastics was further assessed. In contrast to the conditions described in eq. 1 (Scheme 2), the reduction of PCL does not occur at room temperature with the dihydrosiloxane. Nevertheless, in presence of 0.3 mol% of the iridium complex 4 and 3 equiv. of TMDS (6 equiv. of hydrides), the polymer was completely decomposed within 8 h at 65°C into numerous silyl ethers which could be further hydrolysed to 1,6 hexanediol with the NaOH/MeOH system (yield of 68 % overall, scheme 5, eq. 13). The need for a convergent depolymerization led us to consider the hydrosilylation of PCL at 110 °C to reduce the intermediate ethers to alkanes. In presence of 0.3 mol% 4 with 6 equiv. of TMDS (Scheme 5, eq.14), in situ ¹H NMR and DEPT 135 ¹³C NMR spectra revealed complete reduction of PCL into hexane and siloxanes in quantitative yield.
Surprisingly, after removing hexane, excess **TMDS** and the solvent under reduced pressure, a non-volatile viscous liquid was isolated. $^{13}$C NMR, $^{29}$Si NMR as well as FT-IR analyses confirmed the formation of polydimethylsiloxane **PDMS**. SEC chromatography was performed to further characterize the resulting polysiloxane and unveiled a high molar weight and a high polydispersity ($\text{Mn} = 1054 \text{ g.mol}^{-1}$, $\text{Mw} = 23209 \text{ g.mol}^{-1}$ and $\text{PD} = 22.0$). Chromatograms also revealed a complex distribution of the molecular weights.

**Scheme 5**: Reductive depolymerization of **PCL** and **PLA** with the system 4/TMDS to the corresponding hydrocarbons.
Regardless the further used of silyl ethers,\textsuperscript{25-28, 35} stoichiometric quantities of silicon by-products are formed, at the expense of a low atom economy for the whole process. In this respect, the use of TMDS for the complete deoxygenation of plastic wastes is more attractive than Et\textsubscript{3}SiH, as it enables the formation of a valuable by-product, PDMS,\textsuperscript{36}, which is widely used as a lubricant, as a food additive, as silicone in breast implants,\textsuperscript{37} etc.\textsuperscript{38} TMDS would be especially meaningful in the depolymerization of bio-sourced polymers, in order to produce a well-defined fuel by reduction of the bio-based carbon matrix and a silicon polymer. PLA was considered as the ideal candidate to test this idea. In presence of 1 mol\% of the iridium catalyst 4 and 6 equiv. of TMDS, PLA was fully deconstructed at 110 °C into propane and polysiloxane PDMS (Scheme 5, eq. 15). Because of its volatility, the yield of propane could not be evaluated but the concentration of C\textsubscript{3}H\textsubscript{8} was sufficient to detect it by \textsuperscript{1}H and \textsuperscript{13}C DEPT NMR (see SI, section III.2). Decreasing the amount of TMDS to 2.0 equiv. did not affect the rate of the reaction leading to propane and polysiloxane, using the same conditions of loadings catalyst, temperature, and time. After removal of the volatiles, a silicon rubber of PDMS remained in the flask (Scheme 5, eq. 16). Similarly to the PDMS silicon oil obtained from PCL depolymerization, the silicon rubber exhibits a complex mass distribution with at least two observable peaks in SEC chromatograms and a high polydispersity. (M\textsubscript{n} = 776 g.mol\textsuperscript{-1}, M\textsubscript{w} = 13976 g.mol\textsuperscript{-1}, and PD = 18.0).

In addition to ethers and esters, Brookhart and co-workers demonstrated that catalyst 4 is able to reduce halogeno-alkyl compounds.\textsuperscript{39,40} The reduction of halogenated polymers was thus attempted with the dechlorination of poly(vinyl chloride) PVC (scheme 6, eq. 17). While no reaction occurs at 65 °C and 90 °C, \textsuperscript{13}C NMR reveals that, in the presence of 0.8 mol\% 4 and 2
equiv. of Et₃SiH, insoluble pieces of PVC were reduced at 110 °C into soluble poly(ethylene) PE. The corresponding chlorosilane Et₃SiCl was formed in 39% yield.

When a mixture of PVC and PLA was exposed to Et₃SiH and catalyst 4, PLA was left untouched at 110 °C, although its depolymerization was observed at 65 °C in the absence of PVC (see Scheme 2, Eq. 4). In parallel, the less reactive PVC was dechlorinated to PE and Et₃SiCl (see SI, section V.4). This reactivity trend is surprising considering that PVC is typically less reactive than PLA. More mechanistic investigations are certainly needed to decipher the origins of these unexpected selectivities. Importantly, a different behavior was observed when catalyst 4 was replaced with B(C₆F₅)₃. In this case, only the polyester (PLA) was depolymerized (see SI, section V.5). Overall, we can conclude that the reactivity of isolated polymers towards a depolymerization method does not transpose directly to mixtures of plastic wastes, where reactivity trends can be inverted.

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\text{Scheme 6: Reductive dechlorination of PVC to PE with the system 4/Et₃SiH.}
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Conclusions

In conclusion, Brookhart’s iridium(III) complex 4 has been utilized for the first time for the depolymerization of a range of oxygenated polymers, comprising household wastes. These include polyesters (PET, PCL, PLA, etc.) and polycarbonates (PC-BPA, PPC). In the presence of hydrosilanes, catalyst 4 cleaves C–O bonds in esters and carbonates to yield silyl ethers and
alkanes. A chlorinated polymer such as PVC was dechlorinated to PE, using the same protocol. Importantly, it was found that the reactivity of isolated esters, carbonates or well-defined polymers does not directly transpose to mixtures of polymers.

The method presents the advantage of regenerating the original monomers or valuable chemicals in a pure form. For instance, bis-phenol A could be isolated in 83% yield from a sequence involving the depolymerization of PC-BPA with 4 and TMDS and the subsequent hydrolysis of the reaction products with NaOH in methanol. In addition, the reduction of bio-based polymers such as PLA with TMDS affords bio-fuels, in the form of hydrocarbons, and PDMS (a valuable silicon polymer), in a process that valorizes both the carbon and silicon feedstocks.

Although catalyst 4 proved very efficient in the plastic depolymerization with low catalyst loadings, its price remains a major obstacle for a scale-up process development. In addition, the energy intensive production of hydrosilanes combined with the generation of silicon by-products calls for alternative methods of reduction. In this perspective, surrogates of hydrosilanes, such as sylilformates (R₃SiOCHO)⁴²-⁴⁴, may offer promising alternatives in the future by using renewable hydride sources (e.g. formic acid).

ASSOCIATED CONTENT

Supporting Information.

The following files are available free of charge.

Supplementary equations, detailed descriptions of experimental methods, and results are provided in the Supporting Information.

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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Synopsis

Several oxygenated plastics are efficiently depolymerized into valuable chemicals under mild hydrosilylation conditions with the use of molecular iridium catalyst.