

Chemical Recycling of Polymethacrylates

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Abstract: Polymethacrylates, including poly(methyl methacrylate) (PMMA), are produced on a large scale for applications ranging from optics to construction, yet their end-of-life fate remains largely linear. Chemical recycling to regenerate the monomer (depolymerization) offers a promising route to circularity, but conventional methods such as pyrolysis rely on high-temperature random scission pathways that suffer from poor selectivity and undesirable side reactions. Recent advances have demonstrated that polymethacrylates synthesized by controlled radical polymerizations can undergo efficient depolymerization under milder conditions through reactivation of thermally labile chain-end functionalities. Emerging mid-chain-initiated depolymerization strategies further extend low temperature chemical recycling to polymers produced by conventional free-radical polymerization. This review highlights these developments, comparing mechanisms, limitations, and opportunities towards scalable, energy-efficient chemical recycling of polymethacrylates to support a more sustainable plastic economy.

Keywords: Chemical recycling · Depolymerization · PMMA · Pyrolysis



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1. Introduction

Polymethacrylates represent a versatile class of vinyl polymers that have found widespread use across industries ranging from automotive and construction to medical devices and consumer products. The methacrylate backbone, derived from easily functionalized methacrylic ester monomers, allows for the design of materials with tunable properties through control over side-chain composition, molecular weight, and polymer architecture. Poly(methyl methacrylate) (PMMA) is the most commercially significant, marketed under trade names such as Plexiglas[®], Perspex[®], and Lucite[®]. PMMA is valued for its optical clarity, mechanical strength, and weatherability, making it a lightweight, shatter-resistant alternative to glass in applications including windows, displays, and lenses. With global production estimated at

3.9 million metric tons in 2020 and a projected compound annual growth rate of 3.4%, the demand for PMMA is growing,^[1] highlighting the increasing importance of developing sustainable strategies to reduce waste and recover value from end-of-life materials.^[2,3] However, it has been estimated that only around 10% of PMMA waste is currently recycled, with the vast majority either incinerated or landfilled.^[1] This review focuses on recent advances in the chemical recycling of polymethacrylates to regenerate the monomers, with an emphasis on techniques which allow lower temperature depolymerization.

1.1 Mechanical Recycling of PMMA

As with almost all plastics, the reasons for low recycling rates are complex and include both economic factors, such as the costs of collection, sorting, and processing, and practical limitations related to the quality of recycled material compared to virgin-grade PMMA.^[4,5] The most common recycling method for PMMA is mechanical recycling, where waste material is shredded and extruded^[6] or dissolved, precipitated, and extruded.^[7] However, these processes typically subject the polymer chains to thermal and mechanical stress, leading to chain scission, reduced molecular weight, and inferior properties in the recycled product.^[5] Mechanically recycled plastics often require blending with virgin material and additives to mitigate these negative effects.^[8] As a result, PMMA obtained solely through mechanical recycling may be unsuitable for high-value applications, limiting its commercial viability and contributing to the continued disposal of most end-of-life PMMA by incineration or landfill.

2. Pyrolysis of PMMA and Necessity of High Temperature Chemical Recycling

Chemical recycling to monomer, commonly referred to as depolymerization, offers a potential alternative to mechanical recycling in which waste PMMA is chemically converted back to its constituent monomer, methyl methacrylate (MMA). This monomer can then be repolymerized to produce material with properties comparable to virgin-grade PMMA, enabling closed-loop recycling.^[2]

As with all addition polymerizations, the formation of polymethacrylates is a reversible process governed by a thermody-

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ruthenium catalyst facilitated homolytic cleavage of the carbon-chlorine bond at the polymer chain-end, generating a radical capable of depropagating. Under optimized conditions, depolymerization proceeded at temperatures as low as 100 °C, producing up to 24% MMA monomer. The yield could be further improved by applying an iterative evaporation strategy, in which the monomer was removed from the system to shift the equilibrium toward further depolymerization, consistent with Le Chatelier's principle. This study demonstrated that catalyst-mediated chain-end activation could enable selective depolymerization of PMMA under significantly milder conditions than pyrolysis.

While the ruthenium-catalyzed system provided an important proof-of-concept, the relatively low monomer yield and reliance on a precious metal catalyst limited its broader practical appeal. Building on this foundation, Matyjaszewski and coworkers demonstrated that copper-based ATRP catalysts could be exploited to achieve more efficient depolymerization of polymethacrylates. In particular, they showed that copper(II) complexes, such as CuCl_2 in combination with tris(2-pyridylmethyl)amine (TPMA) as a ligand, could catalyze the depolymerization of a range of chlorine-terminated polymethacrylates at relatively mild temperatures (Fig. 2A).^[29,30] Under optimized conditions, depolymerization proceeded at 170 °C, affording monomer yields exceeding 70%. More recently, Anastasaki and coworkers reported that the inclusion of small amounts of volatile co-solvents, such as acetone, allowed these depolymerizations to be carried out in vessels open to air, without the need for sealed or inert reaction conditions.^[31] In parallel to copper catalysis, iron-based systems have also been explored as alternatives for promoting the depolymerization of ATRP-synthesized polymethacrylates. Iron salts are inexpensive, abundant, and generally considered less toxic than many other transition metals, making them attractive candidates for sustainable catalysis. Iron(II) halides, such as FeCl_2 or FeBr_2 , as well as Fe(0) can promote the depolymerization of chlorine-terminated PMMA under conditions similar to those used for copper catalysis.^[32]

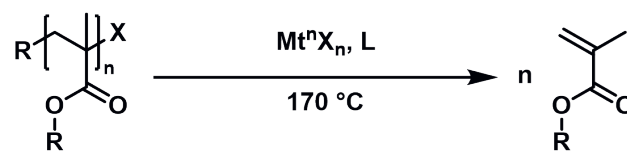
While these metal-catalyzed systems show considerable promise, the loss of halogen end-groups at elevated temperatures through intramolecular lactonization limits depolymerization efficiency by deactivating chain-ends.^[29,30] Lowering the reaction temperature is therefore important to suppress side reactions and improve monomer recovery. However, 170 °C is typically required to promote the reduction of Cu(II) to Cu(I) , which sustains the catalytic cycle. To address this, Anastasaki, Matyjaszewski and coworkers developed a photothermal depolymerization method using iron catalysts, where light-induced photoreduction enabled efficient depolymerization at as low as 100 °C (Fig. 2B).^[33] This approach also allowed temporal control by switching the process on and off with light. More recently, the addition of initiators for continuous activator regeneration (ICAR) has also been employed to lower the required temperatures for both copper and iron-catalyzed depolymerizations by facilitating Cu(I) or Fe(II) generation through a radical-mediated pathway (Fig. 2C).^[34]

3.2 Reversing RAFT Polymerization

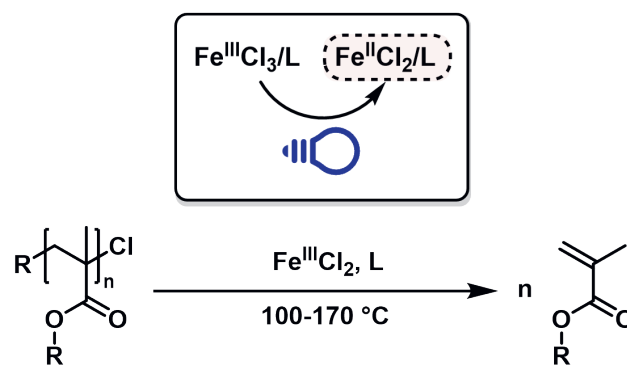
Reversible addition-fragmentation chain-transfer (RAFT) polymerization makes use of a chain-transfer agent (CTA), usually a thiocarbonylthio compound, to mediate propagation *via* degenerative chain transfer. In this process, growing chains rapidly exchange between active propagating radical and dormant CTA-capped species allowing control over molecular weight and dispersity while preserving end-group fidelity. As with ATRP, the presence of a reactive end-group enables post-polymerization modifications and chain extension reactions.

In 2018, Gramlich and coworkers reported the first example of depolymerization of RAFT-synthesized polymethacrylates.^[35] They demonstrated that bottlebrush polymers of poly(PDMSMA)

A) Atom Transfer Radical Depolymerization



B) Photothermal Depolymerization



C) ICAR Depolymerization

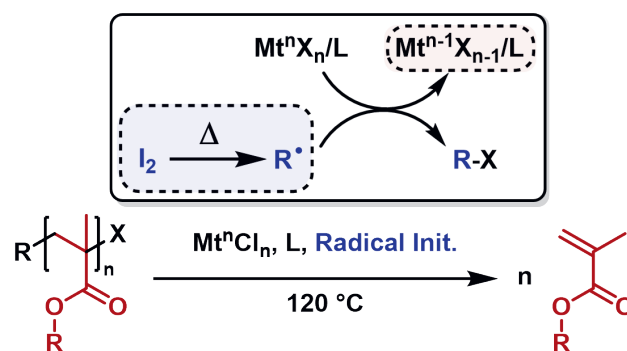


Fig. 2. Selected approaches to atom transfer radical depolymerization. Figure adapted from Fig. 1 of *J. Am. Chem. Soc.* **2023**, *145*, 21146-21151, licensed under CC-BY-4.0 <https://creativecommons.org/licenses/by/4.0/>

and poly(oligo(ethylene glycol) methyl ether methacrylate) (POEGMA), bearing trithiocarbonate end-groups, could undergo depolymerization in 1,4-dioxane at 70 °C, reaching 35% monomer recovery after 56 hours. Although the initiation mechanism was not fully established, the authors suggested that chain-end re-activation or external radical sources could be responsible. A key conclusion of the study was that depropagation can significantly affect the polymerization of bulky macromonomers, highlighting the need for careful selection of reaction conditions, such as high monomer concentration and low temperature, to avoid depropagation.

In 2022, Anastasaki and coworkers reported the first depolymerization of RAFT-synthesized polymethacrylates bearing non-bulky substituents, including PMMA (Fig. 3).^[36] Depolymerization in 1,4-dioxane at 120 °C afforded up to 92% monomer recovery under optimized conditions. Repolymerization of the recovered monomer and CTA produced new polymers with low dispersity, demonstrating the potential for closed-loop chemical recycling of both the polymer and the RAFT agent. A subsequent study analyzing the small-molecule products implicated solvent-derived radicals, generated from dioxane, as the primary initiators of depolymerization.^[37] These radicals were proposed to add to the RAFT end-group, triggering fragmentation and generating a propagating radical capable of initiating depropagation. Building

on this insight, the use of conventional free-radical initiators has been explored to facilitate chain-end activation more efficiently, leading to improved depolymerization rates^[38] and higher polymer loadings.^[39]

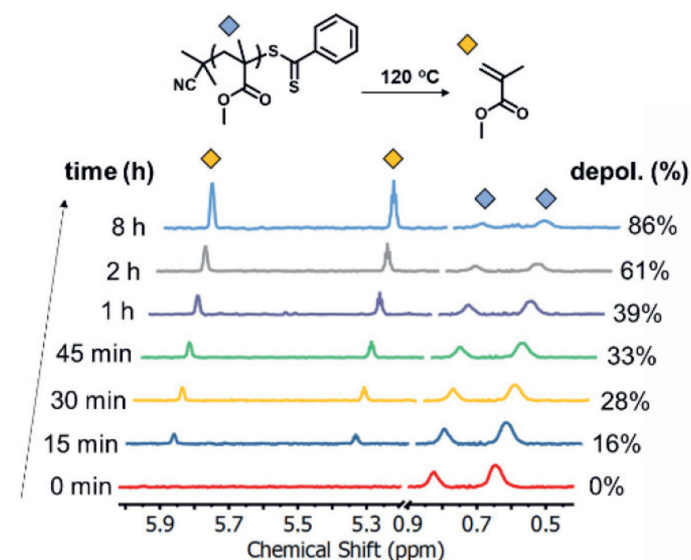


Fig. 3. NMR spectra from the depolymerization of RAFT-synthesized PMMA at 120 °C in 1,4-dioxane. Figure adapted from Fig. 2b of *J. Am. Chem. Soc.* **2022**, *144*, 4678-4684, under CC-BY-NC-ND 4.0 (<https://creativecommons.org/licenses/by-nc-nd/4.0/>).

More efficient activation of polymethacrylate chain-ends to achieve depolymerization has also been demonstrated using photochemical approaches. For example, in 2022, Sumerlin and coworkers reported that LED irradiation could directly homolytically cleave the terminal thiocarbonylthio group, generating polymeric radicals capable of initiating depolymerization.^[40] This method enabled faster depolymerization of PMMA than purely thermal activation *via* solvent-derived radicals and allowed the reaction temperature to be reduced to 100 °C. In a related strategy, photoinduced electron transfer (PET) catalysis has also been shown to enhance RAFT depolymerization, with a report from Anastasaki and coworkers demonstrating that using eosin Y as a photocatalyst not only accelerated depolymerization but also broadened the range

of compatible solvents.^[41] Boyer and coworkers also showed that employing zinc tetraphenylporphyrin (ZnTPP) as a photocatalyst in depolymerization can confer a degree of oxygen tolerance.^[42]

3.3 Controlled Depolymerization

A potential advantage of depolymerizing polymers synthesized by controlled radical polymerizations is the opportunity to mediate depolymerization in a controlled manner, similar to the forward polymerization process. In such a process, molecular weight decreases gradually with conversion, instead of slow activation and fast unzipping of chains. This could allow tuning of polymer molecular weight or sequencing of complex block copolymer architectures, which would be difficult to achieve using methods such as pyrolysis which rely on random chain scission. In 2024, Anastasaki and coworkers demonstrated that increasing the effective concentration of deactivating species in RAFT depolymerization, either by increasing the polymer concentration (and thus the end-group concentration) or by adding exogenous, high-activity chain transfer agents, enabled a controlled depolymerization process in which molecular weight decreased progressively as the reaction proceeded.^[43] This behavior was further illustrated by the sequential release of different monomers during the depolymerization of block copolymers (Fig. 4), providing experimental evidence for controlled, sequence-selective depolymerization. A more recent study demonstrated that electron-withdrawing Z-groups on the chain transfer agent enhance deactivation efficiency, enabling controlled depolymerization at temperatures as low as 90 °C.^[44] Controlled depolymerization has also been demonstrated in PET-RAFT systems, where suppression of thermal initiation through careful tuning of reaction conditions, combined with sufficient deactivation, resulted in progressive shifts in molecular weight and enabled ideal temporal control over the depolymerization process.^[45]

3.4 Bulk depolymerization of PMMA

Industrial pyrolysis of polymethacrylates is typically carried out as a bulk process, which reduces solvent handling, processing costs, and downstream purification steps.^[7] In contrast, all of the depolymerization strategies described so far for polymers synthesized by controlled polymerization have been demonstrated in solution. While the use of solvents lowers the ceiling temperature by diluting the polymer,^[9] they also introduce additional cost, complexity, and environmental considerations. This has motivated researchers to explore whether the selectivity and mild activation pathways of end-group initiated depolymerization can be

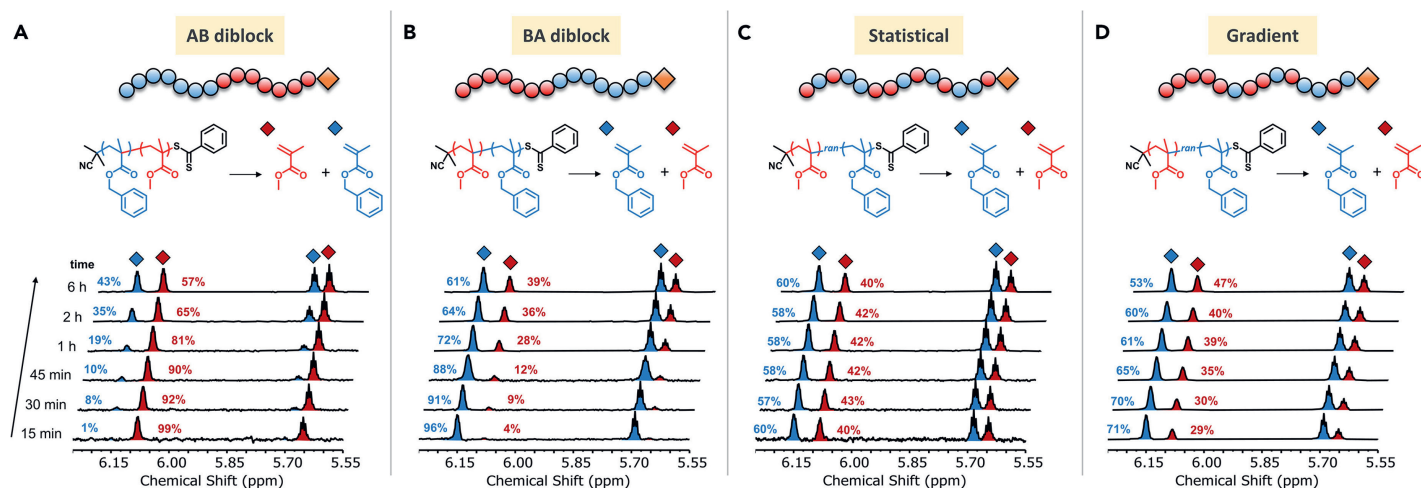


Fig. 4. NMR spectra showing controlled depolymerization of diblock (A and B), statistical (C), and gradient (D) copolymers of MMA and benzyl methacrylate by RAFT depolymerization. Reprinted from *Chem* **2024**, *10*, H. S. Wang, K. Parkatzidis, T. Junkers, N. P. Truong, A. Anastasaki, 'Controlled radical depolymerization: Structural differentiation and molecular weight control', 388-401, Copyright 2024, with permission from Elsevier.

combined with the operational simplicity of solvent-free processing. Sumerlin and coworkers used α,ω -functionalized PMMA, containing both a thiocarbonylthio and a thermally labile phthalimide unit, to achieve depolymerization in bulk on a multi-gram scale at 210–220 °C (Fig. 5).^[46] Matyjaszewski and coworkers also showed that adding copper catalysts to ATRP-derived PMMA enabled rapid depolymerization in bulk, achieving 84% monomer recovery within 15 minutes at 230 °C using short-path distillation.^[47] Additionally, Anastasaki and coworkers demonstrated that RAFT and ATRP PMMA modified to macromonomers with unsaturated chain-ends depolymerized in bulk at 220 °C, yielding up to 90% monomer recovery.^[48]

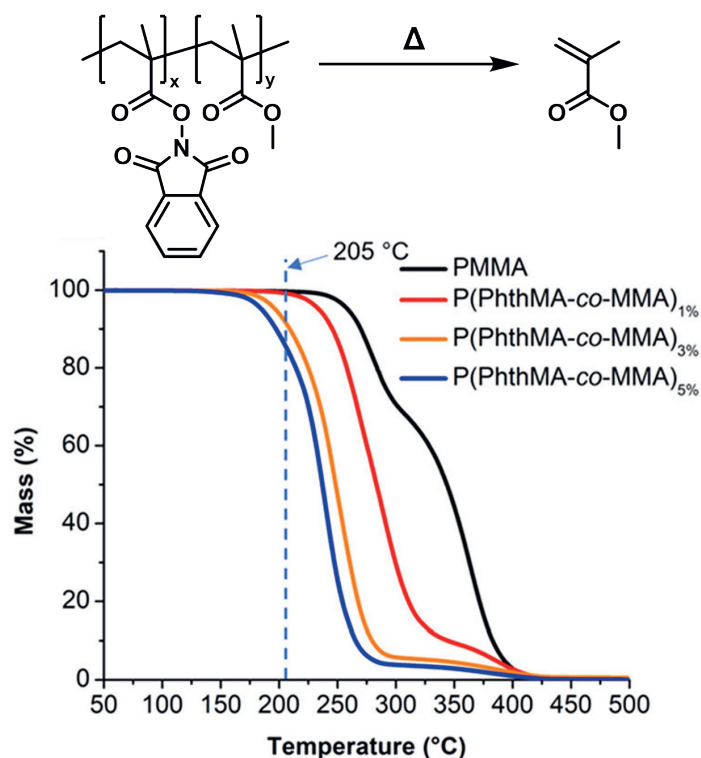


Fig. 5. Copolymers of MMA with PhthMA, giving bulk depolymerization at lower temperature as seen by thermogravimetric analysis (TGA). Reprinted (adapted) with permission from *J. Am. Chem. Soc.* **2024**, *146*, 6217–6224. Copyright 2024 American Chemical Society.

4. Emerging Strategies for Low-Temperature Depolymerization of Free Radical PMMA

While low-temperature depolymerization of polymethacrylates synthesized by reversible deactivation radical polymerization (RDRP) has shown significant promise, the industrial use of these materials remains limited.^[49] In contrast, free radical polymerization remains the dominant method for producing PMMA due to its simplicity, cost-effectiveness, and robustness. However, these polymers lack defined end-groups, making them unsuitable for chain-end mediated depolymerization strategies. As a result, achieving efficient depolymerization of free radical PMMA at lower temperatures requires alternative approaches that do not rely on end-group fidelity.

4.1 Reactive Comonomers

One strategy to enable depolymerization of free radically synthesized PMMA involves the incorporation of reactive comonomers that introduce thermally labile sites, or ‘weak points’, along the polymer backbone. Sumerlin and coworkers found that free radical copolymerization of MMA with 1–5% of *N*-hydroxyphthalimide methacrylate (PhthMA) gave polymers

which depolymerized to >90% MMA in bulk at ~290 °C, whereas PMMA homopolymers reached only ~50% depolymerization conversion.^[50] This approach was also shown to be effective for ultra-high molecular weight copolymers and crosslinked PMMA networks. In a separate study, Diao, He, Liu, and coworkers employed a similar comonomer strategy to lower the depolymerization temperature of PMMA synthesized by conventional free radical polymerization.^[51] Copolymerization of MMA with small amounts of α -methylstyrene was carried out at 85 °C, yielding a material with physical properties comparable to PMMA but capable of depolymerizing in bulk at 180–210 °C to afford >90% MMA.

4.2 Direct Activation of PMMA

Commercial PMMA often contains small amounts of comonomers, such as butyl acrylate, which are added to improve thermal and mechanical properties. These comonomers can act as ‘stops’ along the polymer chain, disrupting the continuous methacrylic backbone needed for efficient unzipping and thereby limiting the effectiveness of many of the low temperature approaches discussed thus far. Effective low temperature depolymerization of commercial grade PMMA could therefore require activation at multiple loci along the chain, in order to overcome the interruptions introduced by comonomers.

In an approach similar to the end-group initiated techniques discussed in section 3, Sumerlin and coworkers utilized mechanochemically promoted chain scission of postconsumer PMMA in the presence of PhthMA monomer to effectively end-functionalize the resultant PMMA macroradicals with a labile phthalimide unit. This end-functional PMMA was found to be suitable for bulk depolymerization with an onset temperature as low as 170 °C.^[52]

A more recent study by Anastasaki and coworkers introduced a strategy based on hydrogen atom transfer (HAT) to generate radicals along the backbone of PMMA.^[53] The approach uses chlorinated solvents, such as dichlorobenzene, which produce chlorine radicals upon visible light irradiation. These chlorine radicals abstract hydrogen atoms from the polymer backbone, forming unstable carbon-centered radicals that undergo *beta*-scission, fragmenting the chain and enabling depolymerization under thermodynamically favorable conditions. Heating solutions of PMMA in chlorinated solvents between 90 and 150 °C under violet LED irradiation resulted in near-quantitative depolymerization, with monomer yields exceeding 98%. Size exclusion chromatography (SEC) of samples taken during the reaction showed a decrease in both the polymer peak area and the number-average molecular weight, consistent with simultaneous chain scission and depolymerization. Multiple scission events per chain allowed this method to be tolerant to copolymers of MMA with small amounts of butyl acrylate. Remarkably, even commercial-grade Plexiglas[®], including pigmented samples, could be effectively depolymerized under these conditions, highlighting the robustness and broad applicability of the approach (Fig. 6).

5. Conclusions

Over the past decade, significant progress has been made toward enabling the chemical recycling of PMMA and related polymethacrylates at temperatures well below those required for traditional pyrolysis. Early studies focused on polymers synthesized *via* RDRP, where end-group reactivation enabled selective depolymerization under relatively mild conditions. This foundation has since expanded to include methods applicable to conventional free radical PMMA, culminating in a HAT-based approach that achieves efficient depolymerization directly from commercial materials.

While these advances represent important milestones, further development is still needed. Advancing the mechanistic understanding of chain activation, particularly mid-chain initiation,

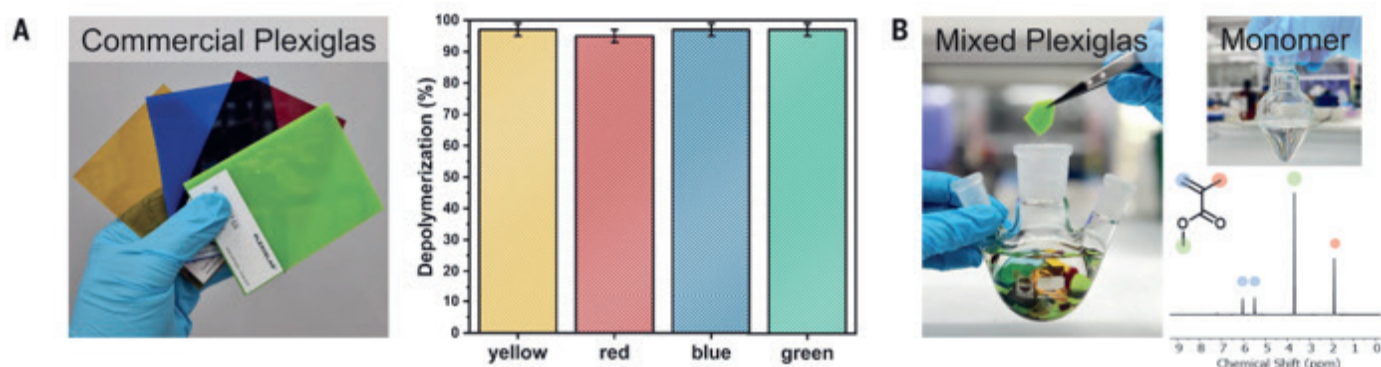


Fig. 6. A) Commercial Plexiglas® samples achieving >95% depolymerization conversion regardless of colour. B) Depolymerization of mixed Plexiglas® showing the NMR spectrum of the pure monomer. From *Science* **2025**, 387, 874-880. Reprinted with permission from AAAS.

could be important for improving the efficiency and robustness of depolymerization processes. In parallel, further exploration of controlled depolymerization may offer new opportunities to selectively reduce polymer molecular weight while partially recovering the monomer, enabling the repurposing of existing materials into new forms. Continued progress in these areas will be key to advancing low temperature depolymerization from a promising laboratory concept to a viable industrial solution for circular PMMA.

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