

Plastic Waste Upcycling Approaches

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Plastic waste pollution, including non-biodegradable landfills, leaching of toxic chemicals into soil and waterways, and emission of toxic gases into the atmosphere, is significantly affecting our environment. Conventional plastic waste recycling approaches generally produce lower-value materials compared to the original plastic or recover inefficient heat energy. Lately, upcycling or the valorization approach has emerged as a sustainable solution to transform plastic waste into value-added products.

Keywords: plastic waste ; upcycling ; vitrimers ; nanocomposites ; 3D printing ; aromatic products ; biopolymers ; sustainability ; circular economy

1. Vitrimerization

In recent years, vitrimerization has emerged as a promising novel approach to reprocess and recycle intractable waste via dynamic chemistry, which involves dynamic covalent bonds, a special type of covalent bond. Dynamic bonds can be dissociative or associative under external stimuli. Materials synthesized with dynamic covalent bond crosslinks are commonly referred to as covalent adaptive networks (CANs) [1]. Vitrimerization is the process of creating 'vitrimers', a new class of plastic materials with associative dynamic covalent bond crosslinks, where the network integrity is maintained during bond exchanges, whereas the network topology is constantly rearranged. Therefore, vitrimers combine the property advantages of thermoplastic and thermoset materials, such as re-processability, healability, recyclability, shape-memory behavior, and self-adhesion [2]. The vitrimer concept developed for commercial plastic materials can be potentially applied for their recycled waste, including polyesters, such as PET (or any thermoplastics containing ester bonds can be upgraded to vitrimers), and polyolefins (HDPE and PP) [3]. Vitrimer based on commercial PET has been developed by incorporating polyol (containing a tertiary amine structure) into the chain of PET (to furnish reactive hydroxyl groups) and reacting it with diepoxy to obtain the dynamic crosslinked networks [4]. The obtained vitrimer exhibited improved thermal and mechanical properties compared to neat PET, and demonstrated excellent re-processability via extrusion, compression, and injection molding suitable for large-scale industrial production. Caffy et al. [5] synthesized vitrimer from commercial HDPE via a single-step reactive extrusion by combining nitroxide chemistry (for radical grafting of 2,2,6,6-Tetramethyl-4-((2-phenyl-1,3,2-dioxaborolan-4-yl)-methoxy)piperidin-1-oxyl (TEMPO-BE) onto polyethylene) and boronic ester metathesis (as an associative exchange reaction). On the other hand, Saed et al. [6] developed a new extrudable vitrimer from PP, which was functionalized with maleic anhydride (MA) and dynamically crosslinked through thiol–thioester bond exchange using a transesterification catalyst 1,5,7-triazabicyclo[4.4.0]dec-5-ene. The PP vitrimer was demonstrated to be readily re-processable (recycled, remolded, rewelded, and 3D printed) multiple times, and exhibited 25% higher mechanical strength compared to the original PP, with a maximum gel fraction reaching about 55%.

Recently, Kar et al. [7] demonstrated the upcycling of PP bottle waste (PP_b) and PE packaging waste (PE_p) into re-processable high-performance vitrimers (with a gel fraction of 58% and 66%, respectively) using melt extrusion processing. The vitrimers were synthesized by first grafting the plastics with MA, followed by crosslinking with bisphenol A diglycidyl ether (DGEBA) using zinc acetylacetonate hydrate (Zn(acac)₂) as the transesterification catalyst. The vitrimers exhibited thermo-reversible associative bond exchange, thermally triggered shape-memory behavior (with 90% recovery after multiple cycles), and superior mechanical stability compared to the original materials [7]. Moreover, development of vitrimer-based composite materials has also been realized, where an increase in filler concentration generally increases vitrimer temperature, mechanical properties, and self-healing properties [8]. These newly developed vitrimer systems have potential applications in a wide range of industrial sectors, including automotive, aerospace, electronics, and biomedical fields [9].

2. Nanocomposite Fabrication

In nanocomposite fabrication, advanced functional materials with tailored properties are developed by incorporation of functional nanofillers into the plastic waste matrix at desired concentrations. The cost, quality, and application of these nanocomposites depend on the type of plastic waste, property, and quantity of the incorporated nanomaterial, as well as the processing route used for composite fabrication. The fabricated composites can be used as such or can be thermochemically transformed into carbonaceous composites. A summary of value-added nanocomposite structures fabricated using plastic waste is provided in **Table 1**. Waste plastic-based composites fabricated using waste wood, rubber, and crushed glass are generally considered recycled composite materials rather than upcycled materials. Nanocomposites can be fabricated by thermal, mechanical, and solution processing methods.

Table 1. Value-added composite materials made from plastic waste.

Composites	Processing Method	Obtained Properties	Applications	References
rPET/CuO NPs	Electrospinning and chemical precipitation techniques.	Photocatalytic activity efficiency for removing the methylene blue dye up to 99%.	Water treatment and filtration	[10]
rHDPE/CuO NPs	Melt mixing and compression molding.	Increased electron density, mass attenuation coefficient, and effective atomic number for γ -ray energies with point sources 356 keV from ^{133}Ba , 662 keV from ^{137}Cs , and 1332 keV from ^{60}Co .	Radioactive source shielding	[11]
rLDPE/SiO ₂ /TiO ₂ NPs	Melt extrusion, granulation, and compression molding.	Tensile strength of 8.4 MPa, and UV protection factor of 1500+.	UV shielding	[12]
rLDPE/PET/Al/Graphite NPs	Shear milling, melt extrusion, and injection molding.	Thermal conductivity of 1.7 W/mK, and electrical conductivity of 10^{-10} S/cm.	Electronic packaging	[13]
rLDPE/PA/Al nanoflake	Powder mixing and compression molding.	Thermal conductivity in the range of 1.4–4.8 W/mK, and electrical conductivity of 10^{-13} S/cm.	Electronic packaging	[14]
rPS/SnO ₂ NPs	Thermally induced phase separation.	Photodegradation efficiency of rhodamine B dye under UV irradiation up to 98.2%.	Water treatment and filtration	[15]
rPS/TiO ₂ NPs/Al microparticles	Solution mixing and electrospinning.	Water contact angle of 157° (superhydrophobic), and daily water productivity of >1.35 L/m ² .	Fog water-harvesting	[16]

Yasin et al. [10] reported a facile strategy for fabricating PET waste into PET nanofibrous membrane embedded with copper oxide (CuO) nanoparticles (NPs) by electrospinning, where the CuO NPs were synthesized using plant extract and mixed with PET waste solution. The authors demonstrated the photocatalytic efficiency of the fabricated nanocomposite membrane for removal (99% efficiency) of methylene blue (MB) dye, which has potential applications in water treatment and filtration. CuO NPs prepared by the combustion method have also been used in the fabrication (melt mixing and compression molding) of nanocomposite sheets with HDPE waste. The nanocomposite sheets exhibited increased electron density, mass attenuation coefficient, and effective atomic number for γ -ray energies, which have potential applications in enhanced radiation-shielding [11]. In a separate study, Fan et al. [12] demonstrated the application of PE film waste-based porous nanocomposite membranes for UV shielding. The nanocomposites were fabricated by mixing PE waste granules with silicon dioxide (SiO₂) and titanium dioxide (TiO₂) NPs in liquid paraffin, followed by extrusion, granulation, and thermal compression molding. The fabricated composite membrane exhibited tensile strength of 8.4 MPa and a UV protection factor of 1500+ [12]. Wang and co-workers [13] reported a facile route to produce nanocomposites for electronic packaging applications using aluminum (Al)-plastic package waste (APPW). The APPW comprising 70% LDPE, 15% Al, and 15% PET was first finely powdered and mixed with graphite nanoplatelets (GNPs) using solid-state shear milling (S3M) technology, followed by extrusion and injection molding to obtain a high thermally conductive (1.7 W/mK) and high electrically insulating (conductivity of 10^{-10} S/cm) nanocomposite. This work was further extended to fabricate nanocomposites using multilayer plastic package waste comprising 80% LDPE and 20% polyamide, where surface-oxidized Al nanoflakes were powder mixed at different ratios and compression molded into sheets exhibiting thermal conductivity in the range of 1.4–4.8 W/mK and high electrical insulation (conductivity of 10^{-13} S/cm) [14].

Conversely, Assis et al. [15] fabricated PS foams impregnated with tin oxide (SnO₂) NPs using a thermally induced phase separation (TIPS) method, which exhibited a photodegradation efficiency of 98.2% for rhodamine B dye under UV irradiation and can be potentially applied for water treatment and filtration applications. Recently, Uddin et al. [16] fabricated superhydrophobic nanocomposites fibers using recycled expanded PS. The nanocomposite membranes were electrospun from PS solutions comprising various proportions of TiO₂ NPs and Al microparticles, which exhibited a water contact angle of 157°. The fabricated membranes were demonstrated for fog-harvesting capability with daily water productivity of >1.35 L/m². In addition, plastic waste-based transformed nanocomposites, where the plastic is converted into carbonaceous material in the final composite product, have also been reported. For example, Mir et al. [17] reported the synthesis of molybdenum carbide carbon (Mo₂C) nanocomposites using plastic waste (pipette tips) and molybdenum trioxide via an in-situ carburization route. The obtained Mo₂C nanocomposite has potential for hydrogen production and energy storage applications. However, such transformations are considered as chemical upcycling rather than nanocomposite formulations.

3. Additive Manufacturing

Additive manufacturing, or three-dimensional (3D) printing, is a constructive technique for building 3D objects from digital models. The 3D printing of plastics has gained increasing research attention in recent years due to its remarkable potential for fabricating complex structures, customizing the product at will, and reduced lead time and waste, which are advantages in comparison to many traditional manufacturing processes commonly used in industries [18]. Fused filament fabrication or fused deposition modeling (FDM) is the most widely used extrusion-based 3D printing technology for fabrication of value-added products from common polymer-based waste materials [19]. In FDM 3D printing, thermoplastic filaments are heated to their melting point in a nozzle head and deposited as polymer melt in a layer-by-layer fashion on a temperature-controlled bed. Lately, a low-cost, closed-loop, and low-carbon-footprint recycling approach has been realized for the circular economy by utilizing used thermoplastics as feedstock material for fabrication of 3D printing filament using milling and screw extrusion techniques [20][21]. However, the quality of 3D-printed plastic material, such as crystallinity, morphology, thermal, rheological, and mechanical properties, decreases with successive grinding and extrusion events [22]. Therefore, to account for such changes and to improve the property, quality, and value of printed structures, a variety of approaches, such as the addition of additives to control crystallinity, micro-nano fillers or reinforcing agents to improve mechanical/electrical property, rheology modifiers to improve printability, and blending of recycled plastic with virgin material or with another polymer/polysaccharide, have been explored [23]. These 3D printable blends or composite systems not only have the potential to overcome the processability and property limitations of pristine polymer systems, but also provide an opportunity to manufacture customized complex 3D engineering structures and industrial products on demand. A summary of value-added structures that are 3D printed by FDM using blend or composite filaments made from plastic waste is provided in **Table 2**. While the manufacturing technology of composites remains the same when natural polysaccharides and synthetic polymers are added, the processing conditions are tuned to suit their physical properties.

Table 2. Summary of value-added structures that are 3D printed by FDM using composite or blend filaments made from plastic waste.

Filament Material	Extrusion Conditions	3D Printing Parameters	Mechanical Properties of Printed Structures	References
rPET/biochar composite	Single screw extrusion at 250 °C.	Bed temperature of 50 °C, nozzle temperature of 270 °C, layer height of 0.4 mm, print speed of 50 mm/s, nozzle diameter of 0.6 mm.	Tensile strengths in the range of 46–52 MPa, elastic modulus in the range of 0.7–0.9 GPa.	[24]
rPET/cellulose fiber composite	Twin screw extrusion with screw speed of 38–43 rpm, feed port at 200 °C, adjacent zone at 260 °C, main zones at 240 °C, die at 220 °C.	Nozzle temperature of 260 °C, print speed of 30 mm/s.	Impact resistance of 23.30 J/m, and impact strength of 2268 J/m ² .	[25]
rPET/CCFs composite	Co-extrusion.	Bed temperature of 80 °C, nozzle temperature of 230 °C, layer height of 0.2 mm, print speed of 300 mm/s, nozzle diameter of 0.4 mm.	Tensile strength of 604.5 MPa, flexural strength of 318.6 MPa.	[26]

Filament Material	Extrusion Conditions	3D Printing Parameters	Mechanical Properties of Printed Structures	References
rHDPE/PP/PP-MAh blend	Single screw extrusion with screw speed of 20 rpm, feed port at 140 °C, adjacent zone at 150 °C, main zones at 160 °C, die at 155 °C.	Bed temperature of 105 °C, nozzle temperature of 215 °C.	Tensile yield stress of 4.78 MPa, strain of 38.1%.	[27]
rHDPE/CF composite	Twin screw extrusion with screw speed of 30 rpm, feed port at 180 °C, adjacent zone at 185 °C, main zones at 190 °C, die at 200 °C.	Bed temperature of 80 °C, nozzle temperature of 290 °C, nozzle diameter of 0.8 mm.	Tensile yield stress in the range of 18–21 MPa, tensile strengths in the range of 37–64 GPa.	[28]
rPP/cellulose composite	Twin screw extrusion with screw speed of 100 rpm, feed port at 140 °C, adjacent zone at 170 °C, main zones at 180 °C, die at 175 °C.	Bed temperature of 100 °C, nozzle temperature of 220 °C, layer height of 0.2 mm, print speed of 20–50 mm/s, nozzle diameter of 0.8 mm.	Tensile strengths in the range of 13–18 MPa, elastic modulus in the range of 1100–1500 MPa.	[29]
rPP/harakeke fibers, and rPP/hemp fibers composites	Twin screw extrusion with screw speed of 50 rpm, feed port at 150 °C, adjacent and main zones at 170 °C, die at 180 °C.	Nozzle temperature of 230 °C, print speed of 50 mm/min, nozzle diameter of 1.0 mm.	PP/harakeke fibers exhibited tensile strength and Young's modulus in the range of 27–39 MPa and 1612–2767 MPa, respectively, whereas PP/hemp fibers were in the range of 28–38 MPa and 1683–2681 MPa.	[30]
rPP/CBS composite	Twin screw extrusion with screw speed in the range of 6–13 rpm, feed port at 175 °C, and the die at 190 °C.	Bed temperature of 90 °C, nozzle temperature of 250 °C, layer height of 0.25 mm, print speed of 60 mm/s, nozzle diameter of 0.8 mm.	Tensile strengths in the range of 8–15 MPa.	[31]
rPP/RH composite	Twin screw extrusion with screw speed of 9 rpm, feed port at 180 °C, adjacent zone at 185 °C, main zones at 190 °C, die at 195 °C.	Bed temperature of 80 °C, nozzle temperature of 240 °C, print speed of 60 mm/s, nozzle diameter of 0.8 mm.	Tensile strengths in the range of 5–14 MPa.	[32]
rPP/rPET, and rPP/rPS blend	Twin screw extrusion with screw speed of 25 rpm, feed port at 140 °C, adjacent zone at 170 °C, main zones at 240 °C, die at 245 °C.	Bed temperature of 100 °C, nozzle temperature of 260 °C, layer height of 0.2 mm, print speed of 20–50 mm/s, nozzle diameter of 0.5 mm.	PP/PET exhibited maximum tensile strength and elastic modulus of 24 MPa and 980 MPa, respectively, whereas PP/PS exhibited 23 MPa and 1459 MPa.	[33]

Incorporation of biochar has been recognized to improve mechanical, thermal, and electrical properties of polymer composites [34]. Idrees et al. [24] reported the fabrication of melt-compounded recycled PET (rPET)/biochar composite filaments by single-screw extrusion at 250 °C, where the biochar was derived from pyrolysis of packaging waste. The 3D-printed structures from a 5 wt% biochar composite filament showed about 60% increase in tensile modulus over neat PET. Carrete et al. [25] fabricated melt-compounded rPET/cellulose fiber composite filaments by twin-screw extrusion, where the cellulose fibers were derived from denim textile waste. The composite filaments were then 3D printed, where a 10 vol% loading of cellulose fibers showed a 62% increase in impact resistance and 64% increase in impact strength over neat PET. Conversely, Bex et al. [26] reported the 3D printing of rPET/continuous carbon filament fibers (CCFs) composite using a co-extrusion-type fused filament fabrication (FFF) printer, where a 25 wt% loading of CFFs showed more than a 10-fold increase in tensile strength over neat PET. For semicrystalline plastics like HDPE, crystallization-induced shrinkage (warping) is a problem during 3D printing. To overcome this issue, Gudadhe et al. [35] compounded waste-derived HDPE with 10% LLDPE and 0.4% dimethyl dibenzylidene sorbitol (DMDBS) using a twin-screw extruder at 190 °C. The extruded blend filaments were 3D printed at 230 °C, which showed a significant decrease in warpage (<0.6 mm for the 10 mm tall bar). Mejia et al. [27] compounded waste-derived HDPE (90%) with 5% PP and 5.0% PP grafted with maleic anhydride (PP-MAh) using a single-screw extruder at 160 °C. The blend filaments were then 3D printed, which exhibited a 39% increase in tensile yield stress, and a 2.7-fold decrease in strain over neat HDPE. Conversely, Borkar et al. [28] reported the fabrication of melt-compounded rHDPE/carbon fibers (CF) composite filaments by twin-screw extrusion at 200 °C, where the CF was derived from dry offcut fabric (Toray T300 grade). The 3D-printed structures from a 29.5 vol% CF composite filament showed about 11% increase in tensile yield and 188% increase in tensile strength over neat HDPE.

Zander et al. [29] reported the fabrication of rPP/cellulose composite filaments by single-screw extrusion at 180 °C, where the cellulose sources were wastepaper, cardboard, and wood flour. The tensile strength and elastic modulus of 3D-printed composites were obtained in the range of 13–18 MPa and 1100–1500 MPa, respectively, where a 10 wt% cellulose composite filament showed about 38% increase in elastic modulus over neat PP.

Conversely, Stoof et al. [30] fabricated rPP/harakeke fiber and rPP/hemp composite filaments by twin-screw extrusion at 180 °C, where the harakeke and hemp fibers were obtained by alkali digestion. A 30 wt% harakeke composite 3D-printed structure exhibited a tensile strength and Young's modulus of 39 MPa and 2.8 GPa, respectively, which is about a 74% and 214% increase from neat PP. Lately, other composite systems, such as rPP/cacao bean shell (CBS) particles [31] and rPP/rice husk (RH) [32], have also been investigated. CBS addition reduced the characteristic warping effect in 3D printing of rPP by 67% and improved the tensile strength and fracture strain of rPP specimens printed at 90° (compared to 0°), where higher particle fracture, filler–matrix debonding, and matrix breakage were observed for samples printed at 0° [31]. Conversely, rPP/RH composite that was 3D printed at 0° exhibited a relatively higher tensile strength compared to the 90° 3D-printed sample [32]. In a separate study, Zander et al. [33] processed blends of waste PP, PET, and PS into filaments for 3D printing, and studied the effect of styrene ethylene butylene styrene (SEBS) and maleic anhydride-functionalized SEBS as the compatibilizer on the resulting mechanical and thermal properties. The 3D-printed rPP/PET and rPP/PS blends exhibited the highest tensile strength of 24 MPa and 22 MPa, respectively, which is about 26% and 16% increase from neat PP. Recently, post-processing heat treatment of 3D-printed parts has also been shown to enhance the mechanical properties [36]. Moreover, the 3D-printed products can also be reprocessed for nanocomposite formulation after the desired use.

4. Catalytic Transformation of Waste Plastic for the Production of Fine Chemicals and Carbon Materials

Plastic waste can be used as an important feedstock material for the preparation of value-added platform chemicals. Conventional approaches used for chemical recycling of plastic waste include pyrolysis (typically using inert atmosphere at 400–800 °C), gasification (typically using air, oxygen, or steam at >700 °C), and solvolysis (typically using solvent medium at 80–280 °C) [37]. However, these techniques are energy intensive and face several challenges, including higher temperature, lower control over product selectivity, longer duration, etc. To overcome such difficulties, researchers have explored the application of different catalysts for transformation of plastic waste into various value-added products under milder conditions (i.e., upcycling) [38]. A summary of catalysts applied for plastic waste upcycling is given in **Table 3**.

Table 3. Summary of catalysts applied for the valorization of plastic waste.

Plastic	Catalyst	Process	Products	Reference
HDPE	HZSM-5 zeolite	Pyrolysis	Ethylene	[39]
PE, PP, PS	Fe/Al ₂ O ₃	Pyrolysis	Amorphous carbon, carbon nanotubes, hydrogen	[40]
PP	Ni-Cu/La ₂ O ₃	Pyrolysis	Multiwalled carbon nanotubes, carbon nanofibers	[41]
LDPE	Fe-Mo/MgO	Pyrolysis	Carbon nanotubes, carbon nanofibers, graphene	[42]
PET, PE, PP	Ni/ZSM-5	Gasification	Syngas	[43][44]
HDPE	Ni/CeO ₂ -ZrO ₂	Gasification	Hydrogen-rich syngas	[45]
HDPE	Ni-Fe/CNT-PC	Gasification	Hydrogen-rich syngas	[46]
PP	Ni/Al ₂ O ₃	Gasification	Hydrogen-rich syngas	[47]
LDPE	Pt/S-ZrO ₂	Hydrogenolysis	Liquid fuels	[48]
LDPE	Pt/USY zeolite	Hydrogenolysis	Alkanes	[49]
PE	Pt/SrTiO ₃	Hydrogenolysis	Lubricants	[50]
HDPE	Pt/SiO ₂ /mSiO ₂	Hydrogenolysis	Alkanes	[51]
LDPE	Pt/WZrO ₂	Hydrogenolysis	Alkanes	[52]
LDPE	Pt/WO ₃ /ZrO ₂ /HY zeolite	Hydrogenolysis	Liquid fuels	[53]

Plastic	Catalyst	Process	Products	Reference
PE, PP	Ru/C	Hydrogenolysis	Alkanes, liquid fuels, lubricants	[54][55][56]
PE, PP	Ru/CeO ₂	Hydrogenolysis	Liquid fuels	[57]
PP	Ru/TiO ₂	Hydrogenolysis	Lubricants	[58]
LDPE	Ru/WZrO ₂	Hydrogenolysis	Alkanes	[59]
PET, PS	Ru/Nb ₂ O ₅	Hydrogenolysis	Arenes	[60]
LDPE, HDPE, PP	Ru/ZrO ₂	Hydrogenolysis	Alkanes, liquid fuels	[61]
LDPE, PP, PS	Ru/FAU zeolite	Hydrogenolysis	Grid-compatible gas streams	[62]
PET	Co/TiO ₂	Hydrogenolysis	Arenes	[63]
PET	CuNa/SiO ₂	Hydrogenolysis	Alcohol, aromatics	[64]
PET	Pt/C, Ru-Cu/SiO ₂	Tandem solvolysis–hydrogenation	Cycloalkanes, aromatics	[65]
PE	Pt/γ-Al ₂ O ₃	Tandem hydrogenolysis–aromatization	Long-chain alkylaromatics	[66]
PP, PS	TiO ₂	Photoreforming	Hydroxyl, carbonyl, and carbon-hydrogen groups	[67]
LDPE	ZnO ₂	Photoreforming	Hydroperoxides, peroxides, carbonyl, and unsaturated groups	[68]
PET	CdS/CdO _x	Photoreforming	Hydrogen	[69]
PET	CNx/Ni ₂ P	Photoreforming	Hydrogen	[70]
PE	Pt/TiO ₂	Tandem solvolysis–photoreforming	Alkene, alkane	[71]
PET	Pd/NF	Electroreforming	Hydrogen	[72]
PET	CoNi _{0.25} P	Electroreforming	Hydrogen	[73]

5. Industrial Biotechnology

Recent developments in biotechnology, such as enzyme-mediated biocatalytic depolymerization of plastic waste into value-added chemicals and utilization of depolymerized plastic waste as feedstock material for microbe-mediated biopolymer synthesis have emerged as sustainable and efficient methods for plastic waste upcycling [74][75]. The “green” nature of these transformations without the use of any hazardous substances provides an eco-friendly route for plastic waste upcycling.

References

- Podgórski, M.; Fairbanks, B.D.; Kirkpatrick, B.E.; McBride, M.; Martinez, A.; Dobson, A.; Bongiardina, N.J.; Bowman, C.N. Toward Stimuli-Responsive Dynamic Thermosets through Continuous Development and Improvements in Covalent Adaptable Networks (CANs). *Adv. Mater.* 2020, 32, e1906876.
- Zheng, J.; Png, Z.M.; Ng, S.H.; Tham, G.X.; Ye, E.; Goh, S.S.; Loh, X.J.; Li, Z. Vitrimers: Current research trends and their emerging applications. *Mater. Today* 2021, 51, 586–625.
- Hayashi, M. Implantation of Recyclability and Healability into Cross-Linked Commercial Polymers by Applying the VitriMer Concept. *Polymers* 2020, 12, 1322.
- Qiu, J.; Ma, S.; Wang, S.; Tang, Z.; Li, Q.; Tian, A.; Xu, X.; Wang, B.; Lu, N.; Zhu, J. Upcycling of Polyethylene Terephthalate to Continuously Reprocessable Vitrimers through Reactive Extrusion. *Macromolecules* 2021, 54, 703–712.
- Caffy, F.; Nicolai, R. Transformation of polyethylene into a vitriMer by nitroxide radical coupling of a bis-dioxaborolane. *Polym. Chem.* 2019, 10, 3107–3115.

6. Saed, M.O.; Lin, X.; Terentjev, E.M. Dynamic Semicrystalline Networks of Polypropylene with Thiol-Anhydride Exchangeable Crosslinks. *ACS Appl. Mater. Interfaces* 2021, 13, 42044–42051.
7. Kar, G.P.P.; Saed, M.O.; Terentjev, E.M. Scalable upcycling of thermoplastic polyolefins into vitrimers through transesterification. *J. Mater. Chem. A* 2020, 8, 24137–24147.
8. Hubbard, A.M.; Ren, Y.; Papaioannou, P.; Sarvestani, A.; Picu, C.R.; Konkolewicz, D.; Roy, A.K.; Varshney, V.; Nepal, D. Vitriimer Composites: Understanding the Role of Filler in Vitriimer Applicability. *ACS Appl. Polym. Mater.* 2022, 4, 6374–6385.
9. Alabiso, W.; Schlögl, S. The Impact of Vitrimers on the Industry of the Future: Chemistry, Properties and Sustainable Forward-Looking Applications. *Polymers* 2020, 12, 1660.
10. Yasin, S.; Zeebaree, S.S.; Zeebaree, A.S.; Zebari, O.H.; Saeed, I. The Efficient Removal of Methylene Blue Dye Using CuO/PET Nanocomposite in Aqueous Solutions. *Catalysts* 2021, 11, 241.
11. Mahmoud, M.E.; El-Sharkawy, R.M.; Allam, E.A.; Elsaman, R.; El-TaHER, A. Fabrication and characterization of phosphotungstic acid—Copper oxide nanoparticles—Plastic waste nanocomposites for enhanced radiation-shielding. *J. Alloys Compd.* 2019, 803, 768–777.
12. Fan, C.; Luo, Y. Polyethylene film waste-derived porous nanocomposites with superior mechanical robustness and excellent UV resistance as supported substrates for the development of multifunctional materials. *J. Mater. Sci.* 2020, 55, 10942–10952.
13. Yang, S.; Bai, S.; Duan, W.; Wang, Q. Production of Value-Added Composites from Aluminum–Plastic Package Waste via Solid-State Shear Milling Process. *ACS Sustain. Chem. Eng.* 2018, 6, 4282–4293.
14. Yang, S.; Li, W.; Bai, S.; Wang, Q. Fabrication of Morphologically Controlled Composites with High Thermal Conductivity and Dielectric Performance from Aluminum Nanoflake and Recycled Plastic Package. *ACS Appl. Mater. Interfaces* 2019, 11, 3388–3399.
15. de Assis, G.C.; Skovroinski, E.; Leite, V.D.; Rodrigues, M.O.; Galembeck, A.; Alves, M.C.; Eastoe, J.; de Oliveira, R.J. Conversion of “Waste Plastic” into Photocatalytic Nanofoams for Environmental Remediation. *ACS Appl. Mater. Interfaces* 2018, 10, 8077–8085.
16. Uddin, N.; Desai, F.J.; Subeshan, B.; Rahman, M.M.; Asmatulu, E. Sustainable atmospheric fog water generator through superhydrophobic electrospun nanocomposite fibers of recycled expanded polystyrene foams. *Surfaces Interfaces* 2021, 25, 101169.
17. Mir, R.A.; Pandey, O. Waste plastic derived carbon supported Mo₂C composite catalysts for hydrogen production and energy storage applications. *J. Clean. Prod.* 2019, 218, 644–655.
18. Jandyal, A.; Chaturvedi, I.; Wazir, I.; Raina, A.; Haq, M.I.U. 3D printing—A review of processes, materials and applications in industry 4.0. *Sustain. Oper. Comput.* 2022, 3, 33–42.
19. Fico, D.; Rizzo, D.; Casciaro, R.; Corcione, C.E. A Review of Polymer-Based Materials for Fused Filament Fabrication (FFF): Focus on Sustainability and Recycled Materials. *Polymers* 2022, 14, 465.
20. Zhao, P.; Rao, C.; Gu, F.; Sharmin, N.; Fu, J. Close-looped recycling of polylactic acid used in 3D printing: An experimental investigation and life cycle assessment. *J. Clean. Prod.* 2018, 197, 1046–1055.
21. Pavlo, S.; Fabio, C.; Hakim, B.; Mauricio, C. 3D-Printing Based Distributed Plastic Recycling: A Conceptual Model for Closed-Loop Supply Chain Design. In *Proceedings of the 2018 IEEE International Conference on Engineering, Technology and Innovation (ICE/ITMC)*, Stuttgart, Germany, 17–20 June 2018; pp. 1–8.
22. Vidakis, N.; Petousis, M.; Tzounis, L.; Maniadi, A.; Velidakis, E.; Mountakis, N.; Kechagias, J.D. Sustainable Additive Manufacturing: Mechanical Response of Polyamide 12 over Multiple Recycling Processes. *Materials* 2021, 14, 466.
23. Zander, N.E. Recycled Polymer Feedstocks for Material Extrusion Additive Manufacturing. In *Polymer-Based Additive Manufacturing: Recent Developments*; ACS Symposium Series; American Chemical Society: Washington, DC, USA, 2019; Volume 1315, pp. 37–51.
24. Idrees, M.; Jeelani, S.; Rangari, V. Three-Dimensional-Printed Sustainable Biochar-Recycled PET Composites. *ACS Sustain. Chem. Eng.* 2018, 6, 13940–13948.
25. Carrete, I.A.; Quiñonez, P.A.; Bermudez, D.; Roberson, D.A. Incorporating Textile-Derived Cellulose Fibers for the Strengthening of Recycled Polyethylene Terephthalate for 3D Printing Feedstock Materials. *J. Polym. Environ.* 2021, 29, 662–671.
26. Bex, G.J.P.; Ingenhut, B.L.J.; Cate, T.T.; Sezen, M.; Ozkoc, G. Sustainable approach to produce 3D-printed continuous carbon fiber composites: “A comparison of virgin and recycled PETG”. *Polym. Compos.* 2021, 42, 4253–4264.

27. Mejia, E.B.; Al-Maqdi, S.; Alkaabi, M.; Alhammadi, A.; Alkaabi, M.; Cherupurakal, N.; Mourad, A.I. Upcycling of HDPE Waste using Additive Manufacturing: Feasibility and Challenges. In Proceedings of the 2020 Advances in Science and Engineering Technology International Conferences (ASET), Dubai, United Arab Emirates, 4 February–9 April 2020; pp. 1–6.
28. Borkar, A.; Hendlmeier, A.; Simon, Z.; Randall, J.D.; Stojcevski, F.; Henderson, L.C. A comparison of mechanical properties of recycled high-density polyethylene/waste carbon fiber via injection molding and 3D printing. *Polym. Compos.* 2022, 43, 2408–2418.
29. Zander, N.E.; Park, J.H.; Boelter, Z.R.; Gillan, M.A. Recycled Cellulose Polypropylene Composite Feedstocks for Material Extrusion Additive Manufacturing. *ACS Omega* 2019, 4, 13879–13888.
30. Stoof, D.; Pickering, K. Sustainable composite fused deposition modelling filament using recycled pre-consumer polypropylene. *Compos. Part B Eng.* 2018, 135, 110–118.
31. Morales, M.A.; Maranon, A.; Hernandez, C.; Porras, A. Development and Characterization of a 3D Printed Cocoa Bean Shell Filled Recycled Polypropylene for Sustainable Composites. *Polymers* 2021, 13, 3162.
32. Morales, M.; Martinez, C.A.; Maranon, A.; Hernandez, C.; Michaud, V.; Porras, A. Development and Characterization of Rice Husk and Recycled Polypropylene Composite Filaments for 3D Printing. *Polymers* 2021, 13, 1067.
33. Zander, N.E.; Gillan, M.; Burckhard, Z.; Gardea, F. Recycled polypropylene blends as novel 3D printing materials. *Addit. Manuf.* 2019, 25, 122–130.
34. Das, C.; Tamrakar, S.; Kiziltas, A.; Xie, X. Incorporation of Biochar to Improve Mechanical, Thermal and Electrical Properties of Polymer Composites. *Polymers* 2021, 13, 2663.
35. Gudadhe, A.A.; Bachhar, N.; Kumar, A.; Andrade, P.; Kumaraswamy, G. Three-Dimensional Printing with Waste High-Density Polyethylene. *ACS Appl. Polym. Mater.* 2019, 1, 3157–3164.
36. Amza, C.; Zapciu, A.; Constantin, G.; Baci, F.; Vasile, M. Enhancing Mechanical Properties of Polymer 3D Printed Parts. *Polymers* 2021, 13, 562.
37. Jiang, J.; Shi, K.; Zhang, X.; Yu, K.; Zhang, H.; He, J.; Ju, Y.; Liu, J. From plastic waste to wealth using chemical recycling: A review. *J. Environ. Chem. Eng.* 2022, 10, 106867.
38. Wang, C.; Han, H.; Wu, Y.; Astruc, D. Nanocatalyzed upcycling of the plastic wastes for a circular economy. *Co-ord. Chem. Rev.* 2022, 458, 214422.
39. Diaz-Silvarrey, L.S.; Zhang, K.; Phan, A.N. Monomer recovery through advanced pyrolysis of waste high density polyethylene (HDPE). *Green Chem.* 2018, 20, 1813–1823.
40. Cai, N.; Li, X.; Xia, S.; Sun, L.; Hu, J.; Bartocci, P.; Fantozzi, F.; Williams, P.T.; Yang, H.; Chen, H. Pyrolysis-catalysis of different waste plastics over Fe/Al₂O₃ catalyst: High-value hydrogen, liquid fuels, carbon nanotubes and possible reaction mechanisms. *Energy Convers. Manag.* 2021, 229, 113794.
41. Aboul-Enein, A.A.; Awadallah, A.E. Production of nanostructure carbon materials via non-oxidative thermal degradation of real polypropylene waste plastic using La₂O₃ supported Ni and Ni–Cu catalysts. *Polym. Degrad. Stab.* 2019, 167, 157–169.
42. Aboul-Enein, A.A.; Awadallah, A.E. Production of nanostructured carbon materials using Fe–Mo/MgO catalysts via mild catalytic pyrolysis of polyethylene waste. *Chem. Eng. J.* 2018, 354, 802–816.
43. Al-Asadi, M.; Miskolczi, N.; Eller, Z. Pyrolysis-gasification of wastes plastics for syngas production using metal modified zeolite catalysts under different ratio of nitrogen/oxygen. *J. Clean. Prod.* 2020, 271, 122186.
44. Yao, D.; Yang, H.; Chen, H.; Williams, P.T. Investigation of nickel-impregnated zeolite catalysts for hydrogen/syngas production from the catalytic reforming of waste polyethylene. *Appl. Catal. B Environ.* 2018, 227, 477–487.
45. Wu, S.-L.; Kuo, J.-H.; Wey, M.-Y. Design of catalysts comprising a nickel core and ceria shell for hydrogen production from plastic waste gasification: An integrated test for anti-coking and catalytic performance. *Catal. Sci. Technol.* 2020, 10, 3975–3984.
46. Zhang, S.; Zhu, S.; Zhang, H.; Liu, X.; Xiong, Y. High quality H₂-rich syngas production from pyrolysis-gasification of biomass and plastic wastes by Ni–/Porous carbon catalyst. *Int. J. Hydrogen Energy* 2019, 44, 26193–26203.
47. Arregi, A.; Abbas-Abadi, M.S.; Lopez, G.; Santamaria, L.; Artetxe, M.; Bilbao, J.; Olazar, M. CeO₂ and La₂O₃ Promoters in the Steam Reforming of Polyolefinic Waste Plastic Pyrolysis Volatiles on Ni-Based Catalysts. *ACS Sustain. Chem. Eng.* 2020, 8, 17307–17321.
48. Utami, M.; Wijaya, K.; Trisunaryanti, W. Pt-promoted sulfated zirconia as catalyst for hydrocracking of LDPE plastic waste into liquid fuels. *Mater. Chem. Phys.* 2018, 213, 548–555.

49. Bin Jumah, A.; Anbumuthu, V.; Tedstone, A.A.; Garforth, A.A. Catalyzing the Hydrocracking of Low Density Polyethylene. *Ind. Eng. Chem. Res.* 2019, 58, 20601–20609.
50. Celik, G.; Kennedy, R.M.; Hackler, R.A.; Ferrandon, M.; Tennakoon, A.; Patnaik, S.; LaPointe, A.M.; Ammal, S.C.; Heyden, A.; Perras, F.A.; et al. Upcycling Single-Use Polyethylene into High-Quality Liquid Products. *ACS Central Sci.* 2019, 5, 1795–1803.
51. Tennakoon, A.; Wu, X.; Paterson, A.L.; Patnaik, S.; Pei, Y.; Lapointe, A.M.; Ammal, S.C.; Hackler, R.A.; Heyden, A.; Slowing, I.I.; et al. Catalytic upcycling of high-density polyethylene via a processive mechanism. *Nat. Catal.* 2020, 3, 893–901.
52. Vance, B.C.; Kots, P.A.; Wang, C.; Hinton, Z.R.; Quinn, C.M.; Epps, T.H.; Korley, L.T.; Vlachos, D.G. Single pot catalyst strategy to branched products via adhesive isomerization and hydrocracking of polyethylene over platinum tungstated zirconia. *Appl. Catal. B Environ.* 2021, 299, 120483.
53. Liu, S.; Kots, P.A.; Vance, B.C.; Danielson, A.; Vlachos, D.G. Plastic waste to fuels by hydrocracking at mild conditions. *Sci. Adv.* 2021, 7, eabf8283.
54. Rorrer, J.E.; Beckham, G.T.; Román-Leshkov, Y. Conversion of Polyolefin Waste to Liquid Alkanes with Ru-Based Catalysts under Mild Conditions. *JACS Au* 2021, 1, 8–12.
55. Jia, C.; Xie, S.; Zhang, W.; Intan, N.N.; Sampath, J.; Pfaendtner, J.; Lin, H. Deconstruction of high-density polyethylene into liquid hydrocarbon fuels and lubricants by hydrogenolysis over Ru catalyst. *Chem Catal.* 2021, 1, 437–455.
56. Rorrer, J.E.; Troyano-Valls, C.; Beckham, G.T.; Román-Leshkov, Y. Hydrogenolysis of Polypropylene and Mixed Polyolefin Plastic Waste over Ru/C to Produce Liquid Alkanes. *ACS Sustain. Chem. Eng.* 2021, 9, 11661–11666.
57. Nakaji, Y.; Tamura, M.; Miyaoka, S.; Kumagai, S.; Tanji, M.; Nakagawa, Y.; Yoshioka, T.; Tomishige, K. Low-temperature catalytic upgrading of waste polyolefinic plastics into liquid fuels and waxes. *Appl. Catal. B Environ.* 2021, 285, 119805.
58. Kots, P.A.; Liu, S.; Vance, B.C.; Wang, C.; Sheehan, J.D.; Vlachos, D.G. Polypropylene Plastic Waste Conversion to Lubricants over Ru/TiO₂ Catalysts. *ACS Catal.* 2021, 11, 8104–8115.
59. Wang, C.; Xie, T.; Kots, P.A.; Vance, B.C.; Yu, K.; Kumar, P.; Fu, J.; Liu, S.; Tsilomelekis, G.; Stach, E.A.; et al. Polyethylene Hydrogenolysis at Mild Conditions over Ruthenium on Tungstated Zirconia. *JACS Au* 2021, 1, 1422–1434.
60. Jing, Y.; Wang, Y.; Furukawa, S.; Xia, J.; Sun, C.; Hülsey, M.J.; Wang, H.; Guo, Y.; Liu, X.; Yan, N. Towards the Circular Economy: Converting Aromatic Plastic Waste Back to Arenes over a Ru/Nb₂O₅ Catalyst. *Angew. Chem. Int. Ed.* 2021, 60, 5527–5535.
61. Tamura, M.; Miyaoka, S.; Nakaji, Y.; Tanji, M.; Kumagai, S.; Nakagawa, Y.; Yoshioka, T.; Tomishige, K. Structure-activity relationship in hydrogenolysis of polyolefins over Ru/support catalysts. *Appl. Catal. B Environ.* 2022, 318, 121870.
62. Lee, W.-T.; Bobbink, F.D.; van Muyden, A.P.; Lin, K.-H.; Corminboeuf, C.; Zamani, R.R.; Dyson, P.J. Catalytic hydrocracking of synthetic polymers into grid-compatible gas streams. *Cell Rep. Phys. Sci.* 2021, 2, 100332.
63. Hongkailers, S.; Jing, Y.; Wang, Y.; Hinchiranan, N.; Yan, N. Recovery of Arenes from Polyethylene Terephthalate (PET) over a Co/TiO₂ Catalyst. *ChemSusChem* 2021, 14, 4330–4339.
64. Gao, Z.; Ma, B.; Chen, S.; Tian, J.; Zhao, C. Converting waste PET plastics into automobile fuels and antifreeze components. *Nat. Commun.* 2022, 13, 3343.
65. Tang, H.; Li, N.; Li, G.; Wang, A.; Cong, Y.; Xu, G.; Wang, X.; Zhang, T. Synthesis of gasoline and jet fuel range cycloalkanes and aromatics from poly(ethylene terephthalate) waste. *Green Chem.* 2019, 21, 2709–2719.
66. Zhang, F.; Zeng, M.; Yappert, R.D.; Sun, J.; Lee, Y.-H.; LaPointe, A.M.; Peters, B.; Abu-Omar, M.M.; Scott, S.L. Polyethylene upcycling to long-chain alkylaromatics by tandem hydrogenolysis/aromatization. *Science* 2020, 370, 437–441.
67. Nabi, I.; Bacha, A.-U.; Li, K.; Cheng, H.; Wang, T.; Liu, Y.; Ajmal, S.; Yang, Y.; Feng, Y.; Zhang, L. Complete Photocatalytic Mineralization of Microplastic on TiO₂ Nanoparticle Film. *iScience* 2020, 23, 101326.
68. Tofa, T.S.; Kunjali, K.L.; Paul, S.; Dutta, J. Visible light photocatalytic degradation of microplastic residues with zinc oxide nanorods. *Environ. Chem. Lett.* 2019, 17, 1341–1346.
69. Uekert, T.; Kuehnel, M.F.; Wakerley, D.W.; Reisner, E. Plastic waste as a feedstock for solar-driven H₂ generation. *Energy Environ. Sci.* 2018, 11, 2853–2857.
70. Uekert, T.; Kasap, H.; Reisner, E. Photoreforming of Nonrecyclable Plastic Waste over a Carbon Nitride/Nickel Phosphide Catalyst. *J. Am. Chem. Soc.* 2019, 141, 15201–15210.
71. Pichler, C.M.; Bhattacharjee, S.; Rahaman, M.; Uekert, T.; Reisner, E. Conversion of Polyethylene Waste into Gaseous Hydrocarbons via Integrated Tandem Chemical–Photo/Electrocatalytic Processes. *ACS Catal.* 2021, 11, 9159–9167.

72. Shi, R.; Liu, K.-S.; Liu, F.; Yang, X.; Hou, C.-C.; Chen, Y. Electrocatalytic reforming of waste plastics into high value-added chemicals and hydrogen fuel. *Chem. Commun.* 2021, 57, 12595–12598.
 73. Zhou, H.; Ren, Y.; Li, Z.; Xu, M.; Wang, Y.; Ge, R.; Kong, X.; Zheng, L.; Duan, H. Electrocatalytic upcycling of polyethylene terephthalate to commodity chemicals and H₂ fuel. *Nat. Commun.* 2021, 12, 467.
 74. Zhu, B.; Wang, D.; Wei, N. Enzyme discovery and engineering for sustainable plastic recycling. *Trends Biotechnol.* 2022, 40, 22–37.
 75. Blank, L.M.; Narancic, T.; Mampel, J.; Tiso, T.; O'Connor, K. Biotechnological upcycling of plastic waste and other non-conventional feedstocks in a circular economy. *Curr. Opin. Biotechnol.* 2020, 62, 212–219.
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