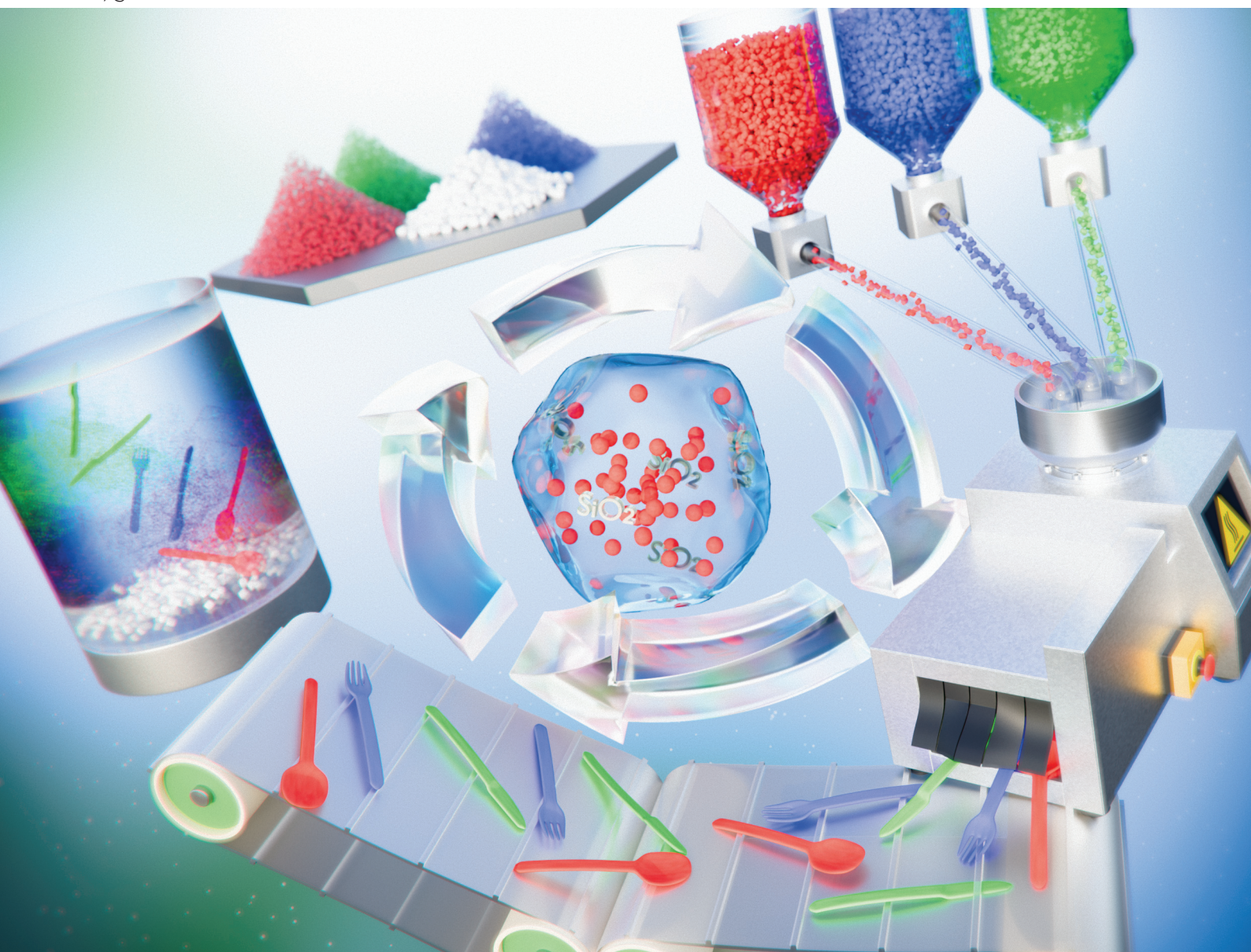


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Sustainable and recyclable polymer coloring system *via* size-tunable, pigment-encapsulated silica microspheres: enabling circular economy of plastics

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The transition toward a circular economy for functional thermoplastics is urgently required to improve resource efficiency and reduce reliance on petroleum-derived feedstocks. Currently, the recycling of colored plastics is severely limited because molecularly dispersed dyes and pigments are difficult to remove selectively, inevitably leading to low-value downcycling. To address this, we developed a “recyclable-by-design” platform using colorant-embedded silica (SiO₂) microspheres synthesized *via* a scalable spray-drying method. These micrometer-sized carriers (typically from 4 to 6 μm in diameter) feature a dense silica shell that provides enhanced thermal stability, protecting encapsulated colorants during industrial melt-processing at 250 °C. By utilizing the particle size as an information carrier, we demonstrated the effective separation of multiple colorants and high-quality resins (PMMA and PS) from mixed waste streams *via* sequential filtration. Experimental results indicate that both the recovered polymers and SiO₂-embedded colorants maintain their original optical and physical characteristics. Furthermore, thermal treatment at 500 °C enables the complete decomposition of the organic colorants, leaving only benign, earth-abundant silica residue. This ensures that the final solid byproduct is environmentally innocuous, providing a practical and sustainable pathway for circular polymer composites. This dual-functional approach, which offers both high-value upcycling and a clean mineral end-state, provides a practical and sustainable pathway for closed-loop functional polymer composites.

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1. This work advances green chemistry by introducing a “recyclable-by-design” strategy for functional thermoplastics to address the challenge of selective colorant removal. Utilizing silica microspheres as physical information carriers enables the separation of colorants and polymers, providing a proactive solution to mitigate conventional downcycling.
2. Experimental results demonstrate the selective separation of multiple colorants and recovered PMMA/PS resins *via* sequential filtration. Colorant-embedded silica remains stable during 250 °C melt-processing, and thermal treatment at 500 °C decomposes organic colorants into benign silica residue, ensuring a safe mineral end-of-life.
3. Future efforts should focus on optimizing the energy requirements of the separation and thermal processes. Expanding the platform to a broader range of engineering plastics and integrating bio-based materials would further enhance the sustainability and environmental profile of circular polymer composites.

1. Introduction

The escalating accumulation of plastic waste represents one of the most formidable environmental challenges of the 21st century, necessitating an urgent paradigm shift from a linear

“take-make-dispose” economy to a robust circular economy.^{1–7} Although commodity thermoplastics, including poly(methyl methacrylate) (PMMA) and polystyrene (PS), are ubiquitous in industrial applications due to their superior optical and mechanical properties, their sustainable management remains fundamentally flawed. Currently, mechanical recycling is the primary route for polymer recovery. However, this process is plagued by the contamination of functional additives.

Colorants are among the most ubiquitous of these additives, serving critical roles in aesthetic appeal, product identifi-

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cation, light shielding, and decoration. These colorants are broadly classified into insoluble pigments and soluble dyes. Inorganic pigments, such as titanium dioxide, iron oxides, carbon black, and complex metal oxides, are extensively utilized owing to their exceptional opacity, thermal stability, and weather resistance. Organic pigments, including azo, phthalocyanine, quinacridone, perylene, and diketopyrrolopyrrole-based derivatives, are employed to achieve vivid hues and high tinting strength, whereas dyes such as anthraquinones and various fluorophores are selected when optical transparency or specific photophysical functions are required. Recent trends in plastic coloration have increasingly focused on high-performance organic pigments and multifunctional colorants engineered with enhanced heat resistance, weatherability, and optical activity. Despite their functional benefits, conventional colorants raise significant sustainability concerns. These include potential toxicity, long-term environmental persistence, and migration or leaching under specific conditions. Most importantly, because these conventional colorants are typically dispersed directly into the polymer matrix at the molecular or nano-scale, they cannot be selectively removed during conventional recycling processes (Fig. 1a). This inherent limitation leads to “downcycling,” which is defined as the co-processing of multi-colored plastic streams into low-value and aesthetically degraded materials that are typically grey or black, thereby severing the loop of high-value material circulation.^{8–12}

The fundamental barrier to high-purity polymer recycling lies in the lack of “design for disassembly” at the additive level. While chemical recycling involving the depolymerization of plastics into their constituent monomers has emerged as a potential solution, the high energy intensity and complex purification steps often diminish the overall sustainability of the process. A more atom-economical approach would involve the physical separation of intact polymer chains from colorants. This challenge is particularly acute for transparent amorphous polymers, where maintaining optical clarity while ensuring pigment stability and recoverability is a delicate balance. While several encapsulation strategies have been explored, they frequently fail to withstand the rigorous thermomechanical conditions of industrial processing such as injection molding, where temperatures often exceed 250 °C.^{13–17}

Building upon our previous research, we addressed these stability issues by developing a scalable spray-drying strategy to synthesize micrometer-sized silica (SiO₂) microspheres (typically from 4 to 6 μm in diameter) with a uniquely dense and hermetic shell. The core of this technology lies in the optimization of the sol–gel precursor chemistry, which ensures the uniform dispersion of even difficult-to-incorporate nanomaterials, such as carbon-dots (C-dots),^{18,19} without compromising their intrinsic optical functions. During the rapid evaporation process in the spray dryer, the silica matrix undergoes extensive polycondensation, forming a non-porous and physically robust exterior layer that effectively shields the

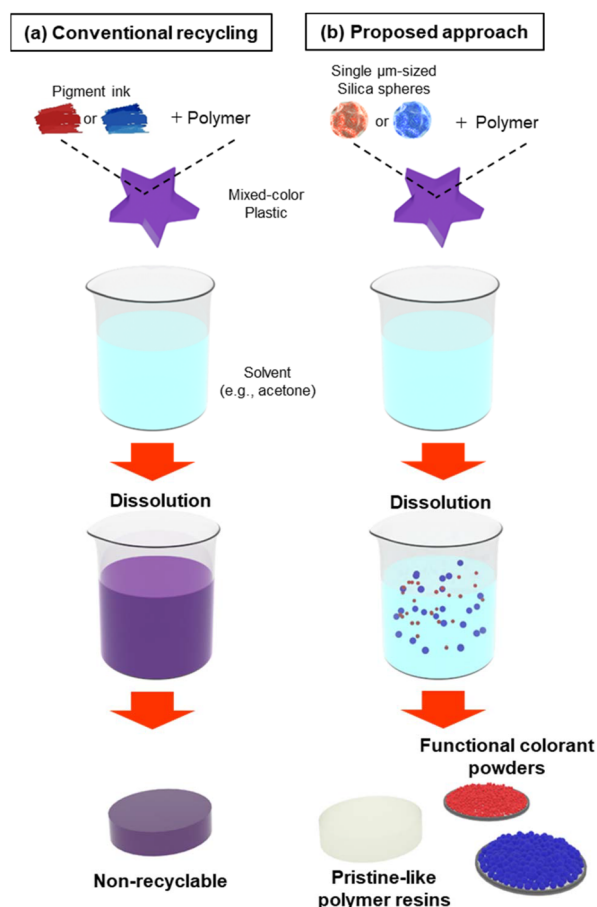


Fig. 1 Schematic comparison of colorant-recycling strategies for functional plastics. (a) Conventional recycling approach: organic pigments or dyes are molecularly dispersed within the polymer matrix. Upon solvent-based dissolution, these colorants cannot be selectively removed, resulting in a mixed-color solution (e.g., purple) and subsequent “downcycling” into low-value, aesthetically degraded materials. (b) Proposed “recyclable-by-design” approach: by embedding colorants within micrometer-sized SiO₂ spheres (typically from 4 to 6 μm in diameter), the colorants exist as discrete, chemically inert carriers. Following polymer (e.g., PMMA, PS) dissolution, these microspheres remain intact and can be precisely separated from the polymer solution. This methodology enables the simultaneous recovery of high-purity, virgin-like polymer resins and functional colorant powders, facilitating a closed-loop, high-value circular economy.

embedded colorants from oxygen and moisture, and helps survival in the high-shear environments during processing.¹⁸ This architecture provides the remarkable thermal and chemical stability necessary for maintaining functionality throughout the entire plastic lifecycle, yet the broader potential of these microspheres as a universal platform for high-purity recycling remained to be fully explored.^{20–23}

In this study, we present a transformative “recyclable-by-design” strategy that leverages the physical robustness and tunable architecture of these spray-dried SiO₂ microspheres to overcome the colorant-contamination bottleneck in plastic recycling (Fig. 1b). We demonstrate that this colorant-embedding platform is not limited to C-dots but is universally appli-

cable to a diverse library of commercial organic molecules, including fluorescent dyes (rhodamine B and fluorescein) and organic industrial pigments (azo and phthalocyanine derivatives). By substituting traditional molecularly dispersed colorants with these discrete silica carriers, we create a composite system, integrated into major resins like PMMA and PS, that can be deconstructed at will.^{24,25}

The core innovation of our approach is the utilization of the microspheres' particle size as an encoded information carrier for automated color sorting. By synthesizing specific colors within discrete size regimes, such as large-diameter blue-emitting spheres and small-diameter red-emitting spheres, we enable the high-purity separation of multiple colorants from a single mixed-polymer solution *via* a simple and sequential filtration process (as illustrated in the schematic diagram of Fig. 1b). We provide experimental evidence that the recovered polymer matrices retain their virgin-like physical and optical characteristics, allowing for high-value "upcycling" without material fatigue. Furthermore, the inherent architecture of these SiO₂-embedded colorants allows for the clean removal of organic components through thermal treatment (*e.g.*, at 500 °C), leaving behind only benign, earth-abundant silica. This feature provides an additional layer of environmental safety, aligning with the principle of "design for degradation" by ensuring that the materials do not persist as hazardous waste even if they exit the recycling loop. This strategy offers a scalable and economically viable pathway toward the realization of a truly closed-loop plastic economy, directly addressing the global imperatives of the Sustainable Development Goals (SDGs).^{26–30}

2. Materials and methods

2.1 Materials

Tetraethyl orthosilicate (TEOS, >95.0% (GC)) was procured from Fujifilm Wako Pure Chemical Corporation. Ethanol (99.5%), acetone (99.5%), toluene (99.5%) and nitric acid (HNO₃) were obtained from Fujifilm Wako Pure Chemical Corporation. Carbon dots (C-dots), specifically Fuji GQD 818, were procured from Fuji Pigment Co., Ltd and utilized without further purification.^{18,19} The concentrated nitric acid was diluted to 1 M with deionized water prior to use. Red and blue colorants, namely an azo pigment and a phthalocyanine pigment, respectively, were supplied by Fuji Pigment Co., Ltd and used as received without further purification. Orange-red fluorescent dye, rhodamine B (maximum emission wavelength: *ca.* 590 nm), and green fluorescent dye, fluorescein (maximum emission wavelength: *ca.* 520 nm), were purchased from FUJIFILM Wako Pure Chemical Corporation and used without further purification. Poly(methyl methacrylate) (PMMA, ACRYPET® VH001, Mitsubishi Chemical) and polystyrene (PS, degree of polymerization, DP = 2000, FUJIFILM Wako Pure Chemical) pellets were used as received.

2.2 Preparation of colorant-embedded SiO₂ microspheres

Colorant-embedded microspheres were synthesized *via* a scalable spray-drying method based on our previously reported

protocol.¹⁸ The precursor sol was prepared by mixing tetraethyl orthosilicate (TEOS, 350 mL, 1.49 mol, accounting for ~95% purity), ethanol (45 mL, 0.77 mol), deionized (DI) water (40 mL, 2.22 mol), and 1 M HNO₃ aqueous solution (30 mL) in a lidded 500 mL polypropylene (PP) container. To incorporate the functional colorants, the specific solvent components were substituted with colorant dispersions or solutions to achieve a final concentration of 0.6 wt% relative to the SiO₂ matrix. Specifically, for C-dots and red/blue colorants, the colorants were pre-dispersed in DI water, which was then used as the water source for the sol. Conversely, for water-insoluble dyes (rhodamine B and fluorescein), the colorants were dissolved in ethanol and utilized as the ethanol component of the sol. After thorough stirring, the resulting sols were aged for 24 h at room temperature. In all cases, the aged sols remained stable, transparent, and low-viscosity, with no observable gelation or precipitation. The aged sol was fed into a spray dryer (Preci, TR160) using a rotating disk atomizer operating at 18 000 rpm. The atomized droplets were rapidly dried in a chamber maintained at 80 °C. The resulting powdered microspheres were collected from the bottom outlet, meticulously washed with DI water and ethanol using an ultrasonic cleaner to remove surface-adsorbed species, and finally dried in an oven at 60 °C for 2 h.

2.3 Preparation of colorant-embedded SiO₂/polymer composites

To prepare colored composite materials, PMMA or PS pellets were used as polymer matrices. First, colorant-embedded SiO₂ microspheres (5 wt% relative to polymer matrices) were melt-compounded with PMMA or PS to produce a masterbatch using a twin-screw extruder (KZW15TW, Technovel Corporation). Extrusion was carried out at a heater temperature of 250 °C to achieve maximum homogeneity and eliminate any localized aggregation of the microspheres within the polymer matrix. The same temperature conditions were applied for both polymer systems. Subsequently, the obtained masterbatches were injection-molded using a benchtop injection molding machine (Mold Lock, Century Innovation Co., Ltd). The PS specimens were injection-molded into a spoon shape at a melt temperature of 250 °C and a mold temperature of 140 °C. All injection-molding processes were conducted under identical conditions. To demonstrate the practical applicability of the proposed system in food packaging, multi-layered PS food trays were fabricated. Initially, a PS solution was prepared by completely dissolving 30 g of PS in 100 mL of toluene within a sealable glass container. Subsequently, 2 g of C-dot-embedded SiO₂ microspheres (C-dot concentration: 0.6 wt% relative to SiO₂) were dispersed into the solution. The resulting mixtures were cast onto a polytetrafluoroethylene (PTFE) plate using a bar coater to form a functional composite layer. During the drying process, the film was peeled from the PTFE substrate while it was still in a soft and tacky state. This film was then applied onto the surface of a commercial PS food tray (133 × 80 × 19 mm; CP Chemical). By utilizing the residual toluene in the film to partially soften the interface, a

robust, seamless bond was formed between the composite layer and the tray substrate. The integrated multilayered tray was then dried completely to ensure full solvent removal before further characterization and dissolution tests. To demonstrate the size-selective recovery of colorants, dual-color SiO₂/PMMA composites were prepared by incorporating two distinct types of colorant-embedded microspheres. Prior to mixing, the microspheres were pre-sorted by size using standard sieves: red-emitting microspheres (rhodamine B-embedded) were collected as the fraction that did not pass through a 40-mesh sieve, while green-emitting microspheres (fluorescein-embedded) were collected as the fraction that passed through a 100-mesh sieve. These size-classified microspheres were added to a PMMA solution (dissolved in acetone) such that the total silica content was 5 wt% relative to the polymer matrix. Approximately equal weight ratios of the red and green microspheres were thoroughly dispersed in the polymer solution to ensure a uniform distribution of both colors. The resulting viscous composite mixture was then cast into silicone animal-shaped molds and dried to obtain the final dual-color specimens for subsequent separation and recycling tests.

2.4 Dissolution recovery test of polymer composites

The dissolution and recovery behavior of the polymer composites was evaluated using shaped molded parts. For the PMMA system, a heart-shaped specimen containing C-dot-embedded SiO₂ microspheres (0.6 wt%) was suspended in a lidded glass container filled with 40 mL of acetone using a metal clip. The solution was stirred using a magnetic stirrer, and the dissolution process was recorded *via* time-lapse photography at regular intervals. Upon complete dissolution, the resulting turbid mixture was allowed to stand to facilitate the sedimentation of the SiO₂ microspheres. The supernatant suspension and the precipitate were separately collected and subjected to centrifugation to recover the powders. The obtained powders were washed by re-dispersing them in 20 mL of acetone, followed by centrifugation; this washing cycle was repeated three times to ensure the removal of residual polymer. Finally, the acetone used during the recovery process was removed *via* vacuum distillation to reclaim both the PMMA resin and the solvent.

A similar dissolution recovery protocol was applied to the PS injection-molded spoons. To account for the solubility of the matrix, toluene was employed as the solvent instead of acetone. The PS spoons were completely dissolved in toluene to liberate the embedded colorant-containing SiO₂ microspheres. The subsequent separation, centrifugal washing (three times with toluene), and solvent evaporation steps were performed identically to the PMMA recovery process to evaluate the feasibility of high-purity material circulation in PS-based systems.

2.5 Characterization

The morphology of the embedded SiO₂ microspheres was observed using a scanning electron microscope (SEM; FlexSEM

1000II, Hitachi). Photoluminescence (PL) spectra of the powder samples were recorded using a fluorescence spectrophotometer (FP-8300, JASCO). To minimize strong light scattering at the powder surface and ensure adequate excitation penetration, the powders were loaded into a dedicated measurement cell and immersed in ethylene glycol, which possesses a refractive index closely matched to that of silica. Three-dimensional fluorescence spectra were acquired by scanning excitation wavelengths from 350 to 550 nm at 1 nm intervals, with the range adjusted to 400 to 600 nm specifically for samples containing fluorescein and rhodamine B. Additionally, the fluorescence intensity under excitation at 450 nm was measured using a fluorescence spectrophotometer (RF-5300PC, Shimadzu). Particle size distributions were measured by dynamic image analysis (DIA) instead of laser diffraction (LD) in order to avoid the influence of surface roughness using a digital microscope (VHX series, Keyence). The specific surface area of the colorant-embedded SiO₂ microspheres was determined using a specific surface area analyzer (BELSORP MAX II, Microtrac). For each surface area analysis, approximately 1 g of sample was weighed into a glass container and degassed at 120 °C under reduced pressure for 24 h. Subsequently, a 100 mg portion was transferred to a sample tube and further pre-treated at 120 °C for 12 h using a degassing apparatus (BELPREP VAC II, Microtrac) before recording the final sample mass. Three samples were analyzed simultaneously to ensure identical measurement conditions, and the adsorption cycles were repeated twice to confirm reproducibility. Photographs of luminescent samples were taken using colored glass filters to suppress the influence of excitation light. For excitation at 365 nm, a SCF-50S-44Y filter (Sigma Koki) was used, while for excitation at 405 nm, a SCF-50S-48Y filter (Sigma Koki) was employed. Changes in the molecular weight of PMMA before and after injection molding were evaluated by gel permeation chromatography (GPC; HLC-8220 GPC, Tosoh) calibrated with six polystyrene standards dissolved in chloroform (Fujifilm Wako Pure Chemical Corporation). Chemical structure changes in recycled PMMA and polystyrene were analyzed using Fourier transform infrared spectroscopy (FT-IR; FT/IR-4600, JASCO). Changes in the embedded SiO₂ microspheres mixed with recycled water-dispersible pigment ink were evaluated by absorbance measurements using a spectrophotometer (V-770, JASCO). UV-vis absorption spectra were recorded in a double-beam mode using a quartz glass cell over a wavelength range of 400 to 700 nm at a constant scan speed of 400 nm min⁻¹. Mechanical tensile properties were evaluated using a universal testing machine (AUTOGRAPH AGS-X 10 kN, SHIMADZU) at a cross-head speed of 10 mm min⁻¹. The injection-molded specimens were prepared with cross-sectional dimensions of 4 mm × 10 mm (corresponding to a cross-sectional area of 40 mm²). To systematically evaluate the impact of the SiO₂ microsphere incorporation on the polymer matrix, specimens prepared from neat polystyrene (PS) pellets and those containing the silica microspheres (0.6 wt%) were tested under identical conditions. DSC and TGA profiles were acquired using a DSC

2500SR and a TGA Q500SR instrument (TA Instruments), respectively. The analyzed sample set comprised neat injection-molded PMMA, the C-dot-silica composite masterbatch, and the final injection-molded PMMA composite. For each run, approximately 5 mg of material was precisely shaved from the specimen, loaded into an aluminum pan, and the exact mass was recorded *via* the instrument software. DSC scans were performed from room temperature to 150 °C at a heating rate of 10 °C min⁻¹. TGA scans were conducted from room temperature to 350 °C at a heating rate of 10 °C min⁻¹. Both thermal analyses were carried out under a continuous nitrogen flow to accurately replicate the oxygen-suppressed environment inside the commercial injection-molding machine.

3. Results and discussion

3.1 Versatility of the spray-drying strategy for encapsulating diverse colorants

To establish the versatility of our spray-drying strategy, we first demonstrated that the embedding process is universally applicable to a variety of commercially available colorants, including organic red/blue pigments, carbon dots (C-dots)^{18,19} and fluorescent dyes. As shown in Fig. 2a, silica microspheres incorporating these diverse colorants were successfully synthesized as fine, vibrantly colored powders. The average diameter of the microspheres is 5 μm. The consistency in the “dimpled” surface morphology across all samples, regardless of the embedded guest molecules, was confirmed by scanning

electron microscopy (SEM) (Fig. 2b and S1). These colorant-embedded silica microspheres were readily incorporated into PMMA matrices to form uniformly colored composite objects (Fig. 2c and S2).¹⁸ To clarify the internal architecture of these composites for readers unfamiliar with our previous report, a schematic representation of the composite structure is provided in Fig. 2d. This diagram illustrates that the individual colorants are hermetically sealed within the dense silica microspheres, which are, in turn, uniformly dispersed as independent structural units throughout the PMMA matrix. This hierarchical structure is key to maintaining the optical integrity of the colorants while preventing their direct contact with the polymer or external environment.

The structural integrity and hermetic nature of the silica shell were rigorously evaluated. Similar to the C-dot-embedded microspheres, the nitrogen (N₂) adsorption-desorption isotherms for these colorant-embedded silica microspheres yielded specific surface areas below the limit of quantitation (LOQ, <1 m² g⁻¹), indicating a remarkably dense structure with negligible accessible porosity (Fig. S3). This lack of porosity is crucial for the stable retention of guest molecules. Indeed, aqueous leaching tests confirmed the exceptional barrier properties of the silica shell; no discoloration of the supernatant was observed even after the microspheres were immersed in deionized water for over 24 hours (Fig. S4). These results confirm that the spray-drying method effectively sequesters diverse colorants within a chemically inert and thermally robust silica matrix (Fig. S5). This “hermetic embedding” ensures that the colorants remain as discrete, non-leachable units, which serves as a fundamental prerequisite for the solvent-based recovery and sorting processes discussed in the subsequent sections.^{31–33}

3.2 Solvent-based recovery and separation of constituent materials

The robust and non-leachable nature of the colorant-embedded silica microspheres enables a highly efficient, solvent-based separation process. To demonstrate the feasibility of recovering both the polymer matrix and the coloring agents, a C-dot-embedded SiO₂/PMMA composite was immersed in acetone. Acetone was selected as the primary solvent because of its exceptional ability to dissolve PMMA and its low boiling point, which facilitates energy-efficient recovery *via* distillation.

As captured in the time-lapse photographs in Fig. 3a, the composite, suspended by a wire, underwent gradual dissolution over approximately 4 hours. During this process, the PMMA dissolved into the liquid phase, while the C-dot-embedded SiO₂ microspheres were released from the matrix and settled at the bottom of the vessel. Upon mechanical agitation, the mixture formed a turbid suspension that exhibited intense fluorescence under UV illumination, confirming the presence of dispersed SiO₂ microspheres throughout the solvent (Fig. 3b). Subsequent centrifugation allowed for the pristine separation of the two components. The supernatant was completely transparent and, crucially, exhibited no detect-

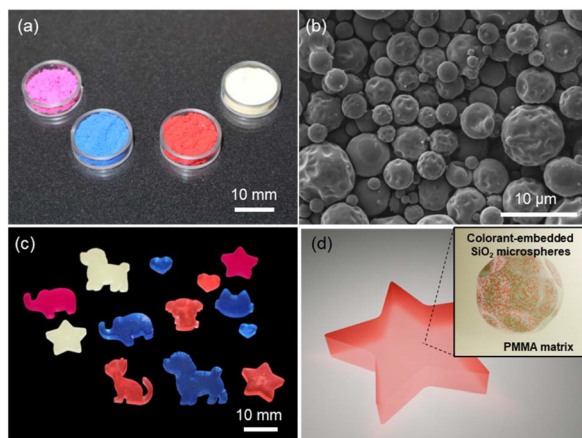


Fig. 2 (a) Optical photographs of spray-dried SiO₂ microspheres incorporating a diverse library of commercial colorants (pigments, fluorescent dyes, and C-dots), obtained as vibrantly colored powders. (b) SEM micrograph of the synthesized C-dot-embedded SiO₂ microspheres showing the characteristic “dimpled” surface morphology and structural uniformity. (c) Optical photograph of various animal- and star-shaped PMMA composites containing different colorant-embedded SiO₂ carriers, taken under 405 nm light. (d) Schematic representation illustrating the hierarchical “recyclable-by-design” structure: guest colorants are hermetically sealed within dense silica microspheres, which are independently dispersed as discrete structural units throughout the PMMA matrix.

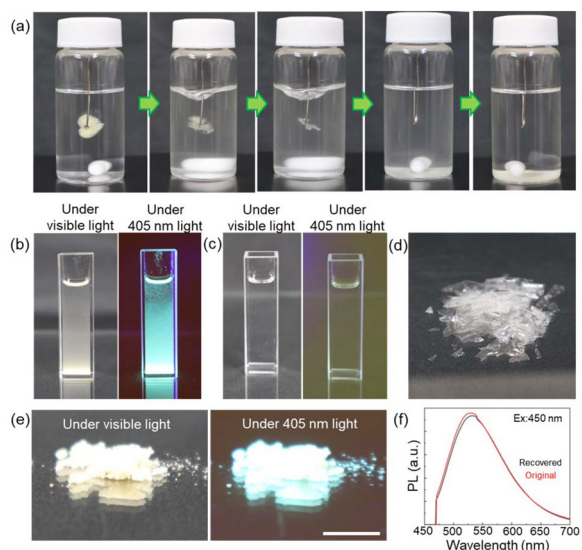


Fig. 3 (a) Time-lapse photographs showing the gradual dissolution of a C-dot-embedded SiO₂/PMMA composite in acetone. (b) Photographs of the resulting turbid suspension containing released microspheres and dissolved PMMA, shown under visible light (left) and 405 nm light (right). (c) Photographs of the transparent supernatant obtained after centrifugation, demonstrating the absence of colorant leaching under visible and UV light. (d) Optical photograph of the high-purity PMMA flakes recovered after solvent evaporation. (e) Photographs of the recovered C-dot-embedded SiO₂ microspheres exhibiting intense fluorescence under 405 nm light. (f) Photoluminescence (PL) spectra ($\lambda_{\text{ex}} = 450$ nm) comparing the original microspheres (red line) and the recovered microspheres (black line), confirming the preservation of optical integrity throughout the recycling process.

able fluorescence under UV light (Fig. 3c). This result was further confirmed by UV-Vis spectroscopy (Fig. S6), which confirmed that the supernatant consisted solely of the dissolved PMMA, with no trace of C-dot leaching. By evaporating the acetone from this supernatant, the PMMA was successfully recovered in the form of transparent flakes (Fig. 3d). Simultaneously, the C-dot-embedded SiO₂ microspheres collected as a precipitate were recovered, retaining their vibrant fluorescence under UV light (Fig. 3e). Notably, the photoluminescence (PL) spectra of these recovered microspheres were nearly identical to those of the original microspheres prior to composite formation (Fig. 3f), demonstrating that the optical integrity of the colorants is perfectly preserved throughout the entire recycling cycle. This dual recovery of high-purity polymer and intact coloring agents, along with the reclamation of the solvent *via* distillation, underscores the potential of this system as a sustainable solution for the polymer industry.

3.3 Multi-cycle recyclability and quality preservation of recovered materials

The practical utility of the recovered PMMA and colorant-embedded SiO₂ microspheres was evaluated through a multi-cycle re-molding demonstration. Unlike conventional recycling, which often results in color contamination or material degradation, our system allows for the complete restoration of

constituent materials to their near-virgin states. As shown in Fig. 4a, we first fabricated a cat-shaped composite by dispersing the C-dot-embedded SiO₂ microspheres in an acetone-based PMMA solution, followed by casting into a mold and subsequent solvent evaporation to obtain the solid object. This initial composite was then subjected to the solvent-based separation process to recover the silica particles and the polymer. These recovered materials were subsequently re-molded into a dog-shaped composite (2nd generation) and then a koala-shaped composite (3rd generation) by repeating the same recovery and fabrication cycle. The successful formation of these intricate shapes across three generations visually demonstrates that the processing characteristics of the PMMA and the structural integrity of the microspheres are fully preserved.

The optical and chemical stability of the materials throughout these repeated cycles was rigorously characterized. The photoluminescence (PL) spectra of the cat, dog, and koala composites exhibited identical profiles (Fig. 4b), confirming that the C-dots remained hermetically sealed and protected within the silica matrix, even after multiple exposures to solvents and thermal processing. Furthermore, the chemical structure of the PMMA matrix remained remarkably stable. The Fourier-transform infrared (FT-IR) spectra (Fig. 4c) showed no alterations in the characteristic vibration bands of PMMA, such as the carbonyl (C=O) stretching at 1730 cm⁻¹, indicating that the polymer did not undergo significant oxidative degradation or structural change. This was further supported by GPC analysis (Fig. S7), which revealed that the molecular weight distribution of the PMMA remained unchanged across all generations. The versatility of this approach was further confirmed by using red pigment-embedded SiO₂ micro-

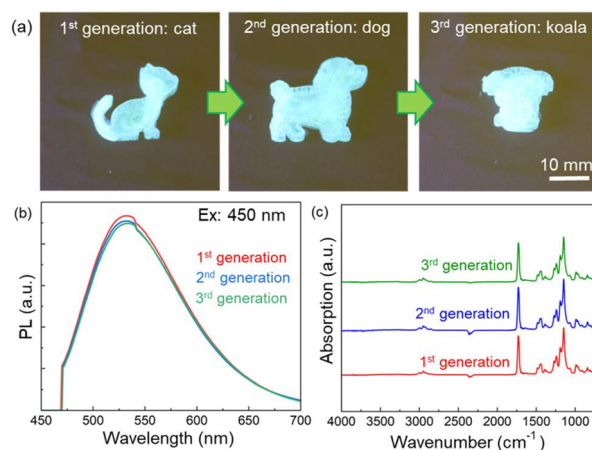


Fig. 4 (a) Optical photographs demonstrating the closed-loop recycling process: recovered materials were sequentially re-molded into 1st generation (cat), 2nd generation (dog), and 3rd generation (koala) composites. All images were taken under 405 nm light. (b) Photoluminescence (PL) spectra ($\lambda_{\text{ex}} = 450$ nm) of the C-dots-embedded SiO₂ microspheres across three recycling generations, confirming the consistent optical performance of the encapsulated colorants. (c) FT-IR spectra of the PMMA matrix across three recycling generations, indicating no significant chemical degradation of the polymer.

spheres/PS composite. These composites also exhibited consistent absorption spectra throughout the recycling cycles (Fig. S8). These collective findings demonstrate that our silica-embedding strategy effectively “decouples” the colorant from the polymer matrix during the recycling process. This ensures that both components can be recovered and reused indefinitely without the loss of optical or mechanical performance, providing a robust platform for high-value closed-loop recycling of colored plastics.

3.4 Practical demonstrations: seamless product recycling and industrial processing

To showcase the real-world applicability of our system, we extended the demonstration to functional forms mimicking commercial products: multilayered food trays and injection-molded cutlery (Fig. 5).^{34,35} To verify the versatility of our approach with different polymer matrices, we utilized PS, a material ubiquitous in food packaging, for the food tray demonstration. We fabricated a colored composite sheet consisting of C-dot-embedded SiO₂ microspheres dispersed in a PS matrix and applied it onto a conventional PS food tray (Fig. 5a). Although a solution-based casting method was utilized here as a bench-scale proof of concept due to laboratory equipment limitations, this step can be readily translated to

large-scale industrial packaging lines. On a commercial scale, the colored functional layer can be seamlessly integrated with the structural polymer base using standard high-throughput manufacturing technologies, such as in-mold labeling (IML), co-extrusion, or inline lamination during the continuous thermoforming process. Notably, because both the tray substrate and the functional colored layer consist of the same polymer type (PS), this multilayered architecture is inherently compatible with collective dissolution processing. Unlike conventional colored packaging where inseparable pigment leaching often leads to contaminated polymer recovery (Fig. S9), our “decoupling” strategy allows the entire product to be treated as a single feedstock for potential solvent-based recovery. This design ensures that even complex, multi-component consumer goods can be rendered fully recyclable by aligning the matrix chemistry with our separable silica-encapsulated colorants. Furthermore, we demonstrated industrial feasibility *via* injection molding. Using a masterbatch prepared from recovered materials, we successfully molded colored spoons at a processing temperature of 250 °C (Fig. 5b–d). While many organic pigments typically decompose above 200 °C, our silica shell effectively acted as a thermal barrier, preserving the colorants’ integrity during high-temperature processing (Fig. S10). These results highlight that our silica-embedding platform not only maintains the aesthetic and functional requirements of consumer products but also ensures their fundamental compatibility with high-value, closed-loop recycling systems (Fig. 5e).^{36,37} Overall, this “recyclable-by-design” approach inherently simplifies end-of-life management. While the primary goal is high-purity material recovery, the system also allows for the thermal decomposition of organic colorants at elevated temperatures (e.g., 500 °C), reverting the material to its mineral origin as benign, earth-abundant silica. This design philosophy aligns with the green chemistry principle of “design for degradation” by preventing the long-term persistence of chemical additives in the environment, providing a robust and sustainable solution for the plastic circular economy.

Finally, we achieved high-fidelity sorting of mixed-color waste by “coding” the physical dimensions of the silica microspheres. To demonstrate this, we prepared a bicolor composite containing two types of silica carriers with distinct size distributions: large fluorescein-embedded microspheres (obtained using a 40-mesh sieve, $>ca. 425\ \mu\text{m}$) and small red-pigment-embedded microspheres (obtained using a 100-mesh sieve, $<ca. 150\ \mu\text{m}$).^{38–41} As shown in Fig. 6a, these microspheres were dispersed within a single PMMA matrix to form a composite exhibiting an interfacial mixture of both colorants. Upon dissolution of the composite in acetone, the mixed solid residue was subjected to mechanical sieving using a 100-mesh screen. Due to the pre-designed size difference, the larger fluorescein-containing particles were retained on the sieve, while the smaller red particles passed through, allowing for the successful recovery of each colorant with high visual and optical purity (Fig. 6b and c). This multi-functional recovery platform transforms complex, multi-colored plastic waste back into discrete, high-purity, and reusable resources, providing a robust

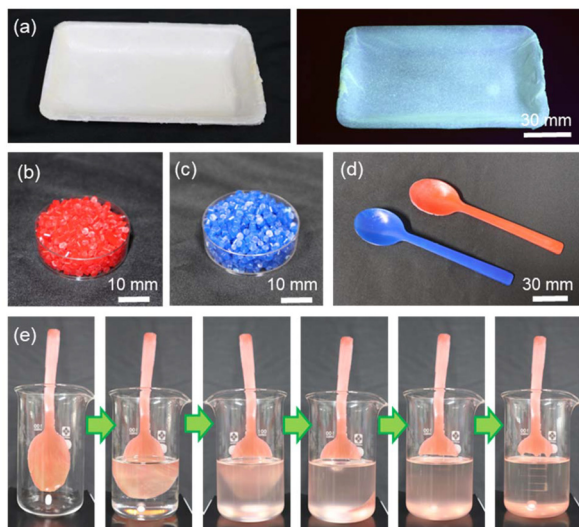


Fig. 5 Real-world applications and demonstration of the closed-loop recycling system. (a) PS food trays mimicking commercial multilayered packaging. Left: photograph under visible light. Right: photograph under 405 nm light, showing the seamless integration of the C-dot-embedded SiO₂/PS composite layer. (b and c) Masterbatches re-formulated from recovered silica carriers: (b) red-pigment-embedded and (c) blue-pigment-embedded carriers compounded with polymer resin at a high concentration for subsequent molding. (d) Injection-molded colored spoons fabricated at an industrial processing temperature of 250 °C using the recovered materials, demonstrating the thermal protection afforded by the silica shells. (e) Time-lapse photographs showing the complete solvent-based dissolution of an injection-molded spoon in toluene, confirming that the molded products retain full recyclability and can be returned to their constituent high-purity resources.

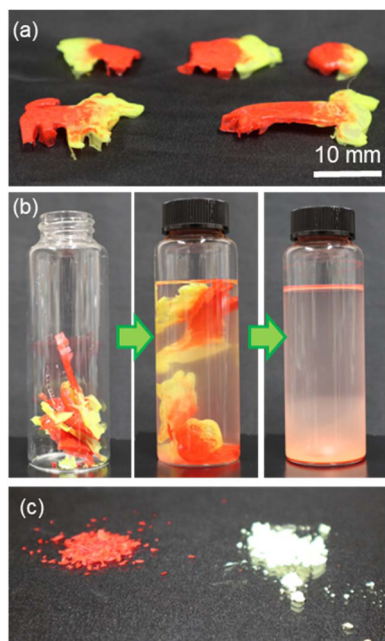


Fig. 6 High-fidelity sorting of mixed-color plastic enabled by size-coded silica carriers. (a) Optical photograph of a bicolor PMMA composite featuring an interfacial mixture of two distinct silica carriers: large fluorescein-embedded microspheres and small red-pigment-embedded microspheres. (b) Sequential photographs showing the solvent-based liberation of silica carriers; the PMMA matrix is completely dissolved in acetone, leaving a mixture of solid colorant carriers at the bottom of the vial. (c) Visual demonstration of the successfully sorted colorants after mechanical sieving using a 100-mesh screen.

solution for the “color contamination” problem in conventional recycling.

3.5 Quantitative sustainability evaluation and green metrics

To critically assess the “green advance” and intrinsic sustainability of our recyclable-by-design platform, a quantitative evaluation was performed using established green chemistry metrics: Process Mass Intensity (PMI) and environmental factor (*E*-factor). These metrics were rigorously applied to our closed-loop recycling stage based on the empirical material balance and com-

pared comprehensively with state-of-the-art conventional recycling technologies (Table 1). As summarized in Table 1, conventional mechanical recycling inherently features a very low PMI and *E*-factor because it avoids secondary chemical solvents; however, this approach fundamentally fails to address the “color contamination” bottleneck, leading to unavoidable downcycling and cascading material degradation. On the other end of the spectrum, traditional chemical recycling (*e.g.*, thermal or catalytic depolymerization) can regenerate virgin-like monomers with high reaction mass efficiency (RME) for the polymer matrix itself. However, it requires severe operational temperatures (typically 300–500 °C) and involves complex, energy-intensive purification steps to remove molecularly dispersed colorants, resulting in a high total material burden (PMI of 1.5–5.0 for the polymer recovery phase alone). In sharp contrast, our dissolution–filtration–distillation platform operates within an exceptionally mild energy window, requiring only room temperature for polymer dissolution and 56 °C for acetone distillation. From a mass balance perspective, our bench-scale demonstration successfully achieved an empirical solvent recovery rate (SRR) of 90.9% *via* a simple vacuum trap. Based on this non-optimized empirical dataset, the baseline PMI and *E*-factor for our polymer/colorant separation process were calculated to be 3.62 and 2.62, respectively (see SI for full calculation details). These baseline values are already highly competitive with conventional chemical depolymerization. Furthermore, upon scaling up to an optimized industrial infrastructure where automated closed-loop fluidics eliminate handling remnants and high-efficiency columns achieve a standard 99% solvent recovery, the projected PMI and *E*-factor dramatically drop to 1.28 and 0.28, respectively, directly challenging the low-mass-intensity profile of mechanical processing. Furthermore, according to established solvent selection guides (*e.g.*, CHEM21), acetone is classified as a “recommended” green solvent due to its low toxicity, high biodegradability, and benign environmental hazard profile. Consequently, the combination of a low operational temperature, low chemical hazard profile, and highly competitive mass intensity mathematically demonstrates that our size-coded sorting and dissolution platform offers a rigorous “green advance” that fulfills the strict requirements for a circular plastics economy.

Table 1 Comprehensive comparison of plastic recycling technologies and their quantitative green metrics

Recycling strategy	Sorting/color separation capability	Target matrix quality	Primary operating temperature	Solvent/chemical hazard (CHEM21)	Estimated PMI (polymer recovery)	<i>E</i> -Factor (polymer recovery)
Mechanical recycling	None (leads to grey/black mixture)	Degraded (downcycling)	200–260 °C (melt extrusion)	None	~1.0–1.1	~0.0–0.1
Chemical recycling (depolymerization)	Difficult/requires heavy post-purification	Virgin-like	300–500 °C (pyrolysis/solvolytic)	Variable (often hazardous catalysts/solvents)	1.5–5.0	0.5–4.0
This work (size-coded dissolution)	Excellent (high-purity sorting <i>via</i> sieving)	Virgin-like (upcycling verified)	Room temp. (dissolution) 56 °C (distillation)	Recommended (acetone)	3.62 (lab actual) 1.28 (industrial intended)	2.62 (lab Actual) 0.28 (industrial intended)

4. Conclusions

In conclusion, we have established a transformative “recyclable-by-design” platform that overcomes the intrinsic limitations of conventional colored plastic recycling. By hermetically embedding diverse colorants, including C-dots and industrial organic pigments, within dense and thermally robust silica microspheres, we successfully decoupled optical functionality from the polymer matrix. This structural innovation prevents the irreversible color contamination that typically leads to low-value downcycling, offering a new paradigm for high-value material circulation.

Our results demonstrate that this strategy is not only chemically versatile across multiple resin systems, such as PMMA and PS, but also industrially viable. The silica shell acts as a critical thermal barrier, allowing embedded colorants to withstand high-temperature industrial processing (up to 250 °C) without degradation. Furthermore, we demonstrated that both the polymer and the colorant-embedded microspheres can be recovered in their near-virgin states over multiple recycling generations, maintaining identical molecular weight distributions and optical profiles. While this study focused on PMMA and PS, this platform is fundamentally applicable to other major resins such as polyesters and polyolefins through appropriate solvent handling or depolymerization processes. However, for specific high-aspect-ratio applications like textile fibers, further downscaling of the silica carriers below the micrometer range will be necessary to preserve fiber spinnability and mechanical integrity. Moreover, from a broader sustainability perspective, the practical scaling of such solvent-based plastic recycling platforms requires meticulous management of the recycling solvents. The substantial energy footprint associated with thermal solvent reclamation (*e.g.*, distillation or evaporation) and the chemical risks regarding the toxicity and flammability of organic solvents represent critical challenges. Therefore, the implementation of closed-loop solvent recovery systems, the minimization of volatile emissions, and the strategic adoption of safer, less hazardous solvent alternatives are essential requirements for translating this proof-of-concept separation strategy into commercial upcycling operations.

The most significant advancement of this work is the introduction of size-coded colorant sorting. By leveraging the physical dimensions of the silica carriers as information carriers, we demonstrated that multi-colored waste streams can be precisely sorted into their constituent colors *via* simple mechanical sieving. This approach transforms plastic waste into high-purity and reusable resources rather than inseparable mixtures. Because spray-drying is already a mature, high-throughput technology widely utilized in industrial manufacturing, the synthesis of these silica carriers can be readily scaled up without fundamental processing hurdles. As a scalable and economically feasible solution, this closed-loop upcycling platform (Fig. 7) provides a practical pathway toward a circular plastic economy. Beyond its recycling potential, the system inherently ensures environmental safety, as the organic com-

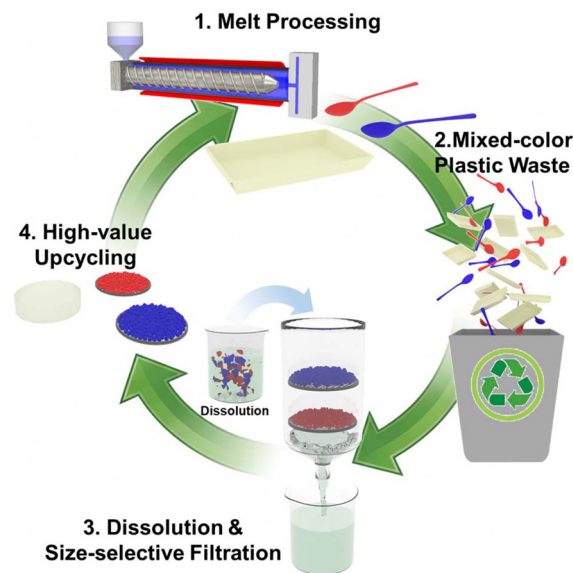


Fig. 7 Overview of the “recyclable-by-design” closed-loop system. (1) Fabrication: production of functional plastics integrated with color-coded SiO₂ microspheres *via* melt processing. (2) Collection: gathering mixed-color plastic waste. (3) Recovery: solvent-based dissolution followed by size-selective filtration to isolate specific colorants and the polymer matrix. (4) Upcycling: re-manufacturing of high-purity, virgin-quality products, establishing a zero-loss circular economy.

ponents can be thermally decomposed to revert the material to a benign mineral state. This holistic design directly addresses global sustainability imperatives and aligns with the principles of green chemistry to realize a truly sustainable future for functional polymer composites.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the supplementary information (SI). Supplementary information is available. The supplementary information (SI) contains quantitative sustainability metrics (E-factor, Process Mass Intensity [PMI], and material yields) for both the microsphere synthesis and the closed-loop polymer recycling process. It also includes characterization data such as SEM micrographs, tensile testing curves, nitrogen adsorption-desorption isotherms, aqueous leaching test results, FT-IR and photoluminescence spectra, GPC molecular weight distributions, UV-Vis absorption profiles, and DSC/TGA thermal analysis profiles (Fig. S1–S10). See DOI: <https://doi.org/10.1039/d6gc02002j>.

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