

Direct Conversion of Plastic Waste to Graphite at Room Temperature under Air via Solvent-Free Mechanochemistry

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Article

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Direct Conversion of Plastic Waste to Graphite at Room Temperature under Air via Solvent-Free Mechanochemistry

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Abstract

Owing to the chemical inertness of sp^3 C–C bonds in polymers, their activation under mild conditions poses a significant barrier to both plastic upcycling and graphitization. Here, we demonstrate a straightforward, solvent-free, one-step mechanochemical method that directly transforms various plastic waste into a critical material, graphite, under ambient conditions ($T < 30^\circ\text{C}$). Facilitated by reactive metals such as Mg, this process requires only $\sim 1.8\%$ of the energy input used in traditional methods while delivering graphite with 92% sp^2 carbon content, 80.4% reaction yield, and 15.3% C/O by mol. Both experimental data and theoretical calculations support the formation of carbides and their conversion into graphite, collectively revealing a carbide–graphite pathway that operates without heat, harsh solvents, extreme temperatures, or inert atmospheres, which are traditionally essential. Experiments further reveal that metals more reactive than carbon (Al, Mg, and Ca) can promote the breakdown and graphitization of a wide range of materials, including non-graphitizable, contaminated, and unsorted plastic waste, as well as biomass. This process dramatically reduces thermal energy demands, requiring less than 8% of the input used in pyrolysis and only 1.5% of that required for conventional graphitization. Beyond scalable and safer waste valorization, the resulting graphite functions effectively in Zn-ion batteries and pencils, and establishes a broadly applicable, ambient-temperature platform for carbon waste conversion, opening new directions in solid-state radical chemistry and circular carbon technologies.

Keywords: graphitization, plastic, upcycling, mechanochemistry, polymer, DFT

Introduction

The increasing demand for plastics in society, along with the waste it generates, is becoming a serious concern, with projections indicating that landfill rates could rise by over 400% by 2050 ¹. Due to their inert nature, plastics predominantly end up in landfills or oceans after use, exacerbating the global plastic waste crisis ². Short-term use of plastics, with a low global recycling rate of only around 9% and the energy-intensive synthesis process, poses significant threats to the environment and ecosystems ³. This challenge is especially pronounced for non-biodegradable plastics, such as highly crystalline low-density polyethylene, which are difficult to manage due to issues like radical recombination, copolymerization, and side reactions during various recycling methods ³.

Numerous great approaches, including thermal, chemical, catalytic, photothermal, and biodegradable recycling, have been proposed to convert polyolefins into products like light olefins, surfactants, wax, benzene-toluene-xylene (BTX), or oils ⁴. However, these methods often face challenges, such as low selectivity, complex processes, the need for expensive catalysts, and extremely high temperatures (Fig. 1a). For instance, despite the remarkable innovation, bacterial degradation of plastic requires nearly two months to break down just 50 mg of it ⁵, while faster degradation (3–6 hours per batch) requires harsh conditions involving strong acids (e.g., HNO_3) and strong oxidizing agents (e.g., KMnO_4 , H_2O_2), with limited control over selectivity. Similarly, traditional pyrolysis at temperatures above 400 °C produces complex mixtures of low-value gases, liquid hydrocarbons, and char ⁴.

Despite these impressive technological advances, many reported methods require extremely high temperatures, stringent reaction environments (such as exclusion of oxygen, carbon dioxide, and water), and significant amounts of co-reactants such as hydrogen.⁴ The harsh conditions accelerate catalyst degradation and corrosion, which can limit the adaptability of these innovations ⁶. Additionally, the lack of proper sorting and cleaning of collected plastics, which leads to contamination and inefficiencies, further exacerbates challenges in traditional recycling chemistry and technologies ⁶.

Upcycling plastic waste into high-demand products such as graphite or graphene offers a promising

solution, given the growing demand across industries and the higher market value compared to traditionally upcycled plastic products, such as oils, wax, BTX, and surfactants ⁷⁻⁹. However, graphitization (the synthesis of graphite) also faces significant challenges related to substrate sorting, catalysts, hazardous outputs, and operating conditions (Fig. 1b). Conventional graphitization methods, such as chemical vapor deposition (CVD), typically require temperatures above 3000 °C. Even when catalysts are used, allowing growth around 1000 °C, CVD still demands an ultra-clean environment, with oxygen pressures in the microtorr (μ Torr) range ¹⁰. Metal catalysts such as Cu or Fe can lower graphitization temperatures through either dissolution-precipitation or carbide formation mechanisms, although only the latter is relevant for plastic upcycling. The dissolution-precipitation route still requires high temperatures to dissolve carbon into the metal and introduces additional constraints ¹¹. Moreover, CVD processing is time-intensive. Nonetheless, as highlighted in red in Fig. 1b, its key advantage lies in producing large-area, wrinkle-free graphite with high structural uniformity ¹². In contrast, mechanochemistry, particularly ball milling, has recently again emerged as a promising and efficient solvent-free approach, effectively enabling chemical transformations and solid-state reactions beyond those achievable by traditional methods ¹³. Its simplicity and scalability make mechanochemistry a compelling alternative for sustainable synthesis.

Here, we demonstrate a room-temperature, solid-state reaction pathway for graphitizing plastic waste via solvent-free ball milling using Mg. Mechanical force destabilizes polymeric C–C bonds, making them more susceptible to reaction with mechanically activated Mg, leading to the formation of transient carbide intermediates that ultimately yield graphite through continuous milling. This approach addresses two major challenges: plastic waste valorization and the easy production of graphitic carbon for energy and material applications. In contrast to conventional high-temperature or multi-step complex processes, this method is operationally simple, low-temperature, and inherently safer, offering a new paradigm for both recycling and graphite synthesis.

This process utilizes the SPEX 8000D model miller with stainless steel balls, with polyethylene (PE), the most widely used and inert plastic (comprising ~36% of plastic waste by mass), serving as the

carbon source. Magnesium (Mg) promotes the efficient scission of C–C bonds within the polymer backbone, while aluminum (Al) serves as an effective graphitization agent. Together, these components enable the conversion of a wide range of plastics and biowastes into ordered graphite (Fig. 1c) characterized by a high sp^2 carbon content (~92%), excellent thermal stability, electrical conductivity, and elemental analysis on par with commercial highly ordered pyrolytic graphite (HOPG). This versatile method has been validated on both reagent-grade and commercial polymers, including polyethylene bags, coffee cups, Styrofoam, bio-waste, laboratory mixed waste, paper waste, and polyvinyl alcohol (PVA). Remarkably, it also enables the graphitization of traditionally non-graphitizable plastics such as polyacrylonitrile (PAN) and polystyrene (PS) via a stress-induced mechanism¹⁴

Despite the high-value end product, graphite, the main advantages of this upcycling approach include the ability to process mixed plastics, paper, and other contaminated carbon sources without requiring precise sorting or cleaning, which was a critical requirement in previous methods. This simplifies the overall workflow and reduces barriers to large-scale implementation.

The practical application of the resulting ball-milled graphite (BM-Graphite) demonstrates its utility in aqueous zinc-ion batteries (AZIBs). BM-Graphite eliminates the activation time typically needed and reduces dissolution issues in V_2O_5 cathodes. When composited with V_2O_5 , BM-Graphite significantly enhances AZIB performance, retaining 96% of its capacity after 500 cycles at 5 A g^{-1} . This study opens new avenues for room-temperature plastic waste upcycling and graphitization.

Results and discussion

Chemistry of upcycling plastic wastes into BM-Graphite

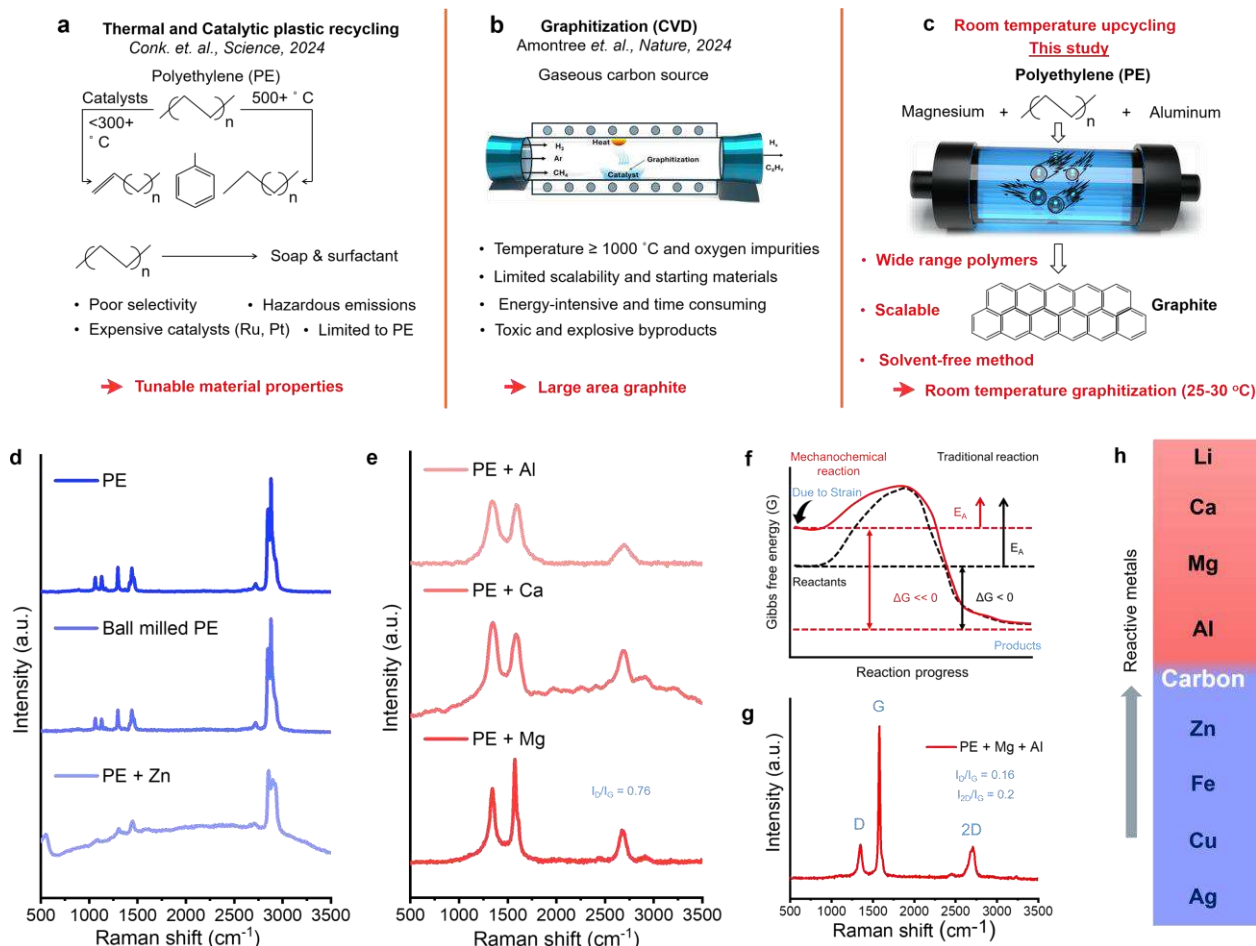


Figure 1 | Ambient-temperature mechanochemical graphitization of polyethylene (PE). (a) Challenges associated with previously reported thermal and catalytic plastic recycling approaches. (b) Limitations of traditional graphitization methods, such as chemical vapor deposition (CVD). (c) Schematic representation and advantages (in light red) of our ball-milling method for synthesizing graphite from plastics at 1060 rotations per minute. (d) Raman spectra comparing pristine PE, ball-milled PE, and PE with Zn demonstrate insufficient graphitization in the absence of highly reactive metals. (e) Introduction of reactive metals (Al, Ca, Mg) under ball milling triggers graphitization, shows characteristic D and G bands with an I_D/I_G of 0.76 for PE + Mg. (f) Comparison of reaction energy barriers across different reaction modes. Mechanochemical pathways uniquely lower energy requirements due to strain-induced bond activation, enabling solid-state transformations accessible at room temperature. (g) Co-milling PE with Mg and Al results in efficient graphitization (I_D/I_G from 0.76 to 0.16). (h) Reactivity series of various metals, highlighting the correlation between metal reactivity and graphitization efficiency under mechanochemical conditions.

The persistence of plastic waste in the environment is largely attributed to the inert nature of its polymeric backbone, composed predominantly of strong sp^3 -hybridized C–C bonds¹⁵. Consistent with this, PE subjected to ball milling without any additives exhibited no significant structural change, as confirmed by Raman spectroscopy and X-ray diffraction (XRD) (Fig. 1d, S2). Similarly, elements positioned lower in the reactivity series—such as copper (Cu), zinc (Zn), chromium (Cr), manganese (Mn), and nickel (Ni), also do not induce carbide formation under the same mechanochemical conditions (Fig. S3). For instance, when PE was ball-milled with Zn, which is just below carbon in the reactivity series,¹⁶ only modest structural disorder was observed, as suggested by peak broadening and reduced crystallinity in Raman (Fig. 1d). The overall Raman spectra, however, remained similar to pristine PE. These changes are indicative of increased amorphization or disrupted molecular packing, rather than carbide formation. Similar observations were made when PS and PE were ball-milled with other transition metals, which are less reactive than carbon (Fig. S3). In contrast, elements positioned above carbon in the reactivity series, such as Al, Mg, and Ca, can efficiently promote the breaking of C–C polymeric bonds to form carbides, which turn into graphite upon continuous ball milling. For instance, ball milling PE with Mg for 45 minutes resulted in its conversion to graphitic carbon, as evidenced by the appearance of characteristic D, G, and 2D bands in the Raman spectrum near 1350 cm^{-1} , 1580 cm^{-1} , and 2700 cm^{-1} , respectively (Figs. 1e, S1).

This observation is consistent when PE and PS were milled with metals positioned above carbon in the reactivity series, specifically Al, Mg, and Ca (Figs. S3–S6). Although when PE is ball milled with Mg, the presence of a relatively intense D band ($I_D/I_G = 0.76$) suggests moderate defect density (Fig. 1e)¹⁷, nevertheless, these results clearly demonstrate that reactive metals can drive polymer-to-graphite conversion under ambient conditions. We attribute this conversion to the stress and strain induced by ball milling, which lowers the activation energy (E_a) barrier for the transformation (Fig. 1f). Mechanochemical processes harness defect- and stress-induced reactions arising from repeated collisions, elevating reactants to high-energy, reactive states and thereby reducing the activation energy required for graphite formation^{18,19}. Consequently, this approach circumvents the high temperatures, harsh solvents, and long processing times.

To improve the quality of the resulting graphite, a series of graphitization reagents was systematically evaluated. Among them, Al emerged as the most effective (Figs. 1g, S4, S5). When Al is used in combination with Mg, the Raman spectrum of the product exhibited a markedly suppressed D band ($I_D/I_G = 0.16$) and an asymmetric 2D band (Figs. S4–S6), indicative of well-ordered graphitic domains (Fig. 1g). These findings suggest a synergistic effect wherein Mg acts as a bond-scission agent, promoting the disruption of the polymer backbone to form carbide, while Al promotes graphitic reorganization.

To further explore the generality of the reactivity series and validate the mechanochemical graphitization pathway, additional combinations such as Mg–Ca, Mg–Fe, and Al–Ca were tested. All were successful in converting polyethylene into graphitic carbon (Figs. S4–S6), reinforcing the versatility and robustness of the approach. Based on these observations, we believe that any reagent capable of generating radicals and stabilizing them more effectively than the polymeric carbon backbone can facilitate a radical-based reaction, as illustrated in Fig. S7.

The impacts of disruptive reagents, graphitization reagents, temperature fluctuations, and pretreatment were carefully analyzed (Fig. S1–S9). Results indicate that the quality of the resulting graphite is primarily influenced by the graphitization reagents (Al, Ca) along with the disruptive reagents (Mg, Al). Notably, optimizing the duration of ball milling and the number of balls used are critical factors affecting graphite quality, with these parameters varying based on the amount and nature of the substrate (for details, refer to the optimization section and the optimization tables (S1, S2) in the SI). Additionally, pretreatment enhances graphite quality due to its structural directing effect (Fig. S8)²⁰. However, in the absence of a reactive metal, pretreatment alone was insufficient to drive the transformation, as evidenced by the negligible changes observed in both Raman and XRD analyses for samples ball-milled without Mg (Fig. S2). These findings underscore the necessity of chemical reactivity of metals (Fig. 1h), beyond mechanical activation alone, for enabling sp^3 -to- sp^2 carbon conversion at room temperature.

A comparative quality test of BM-Graphite

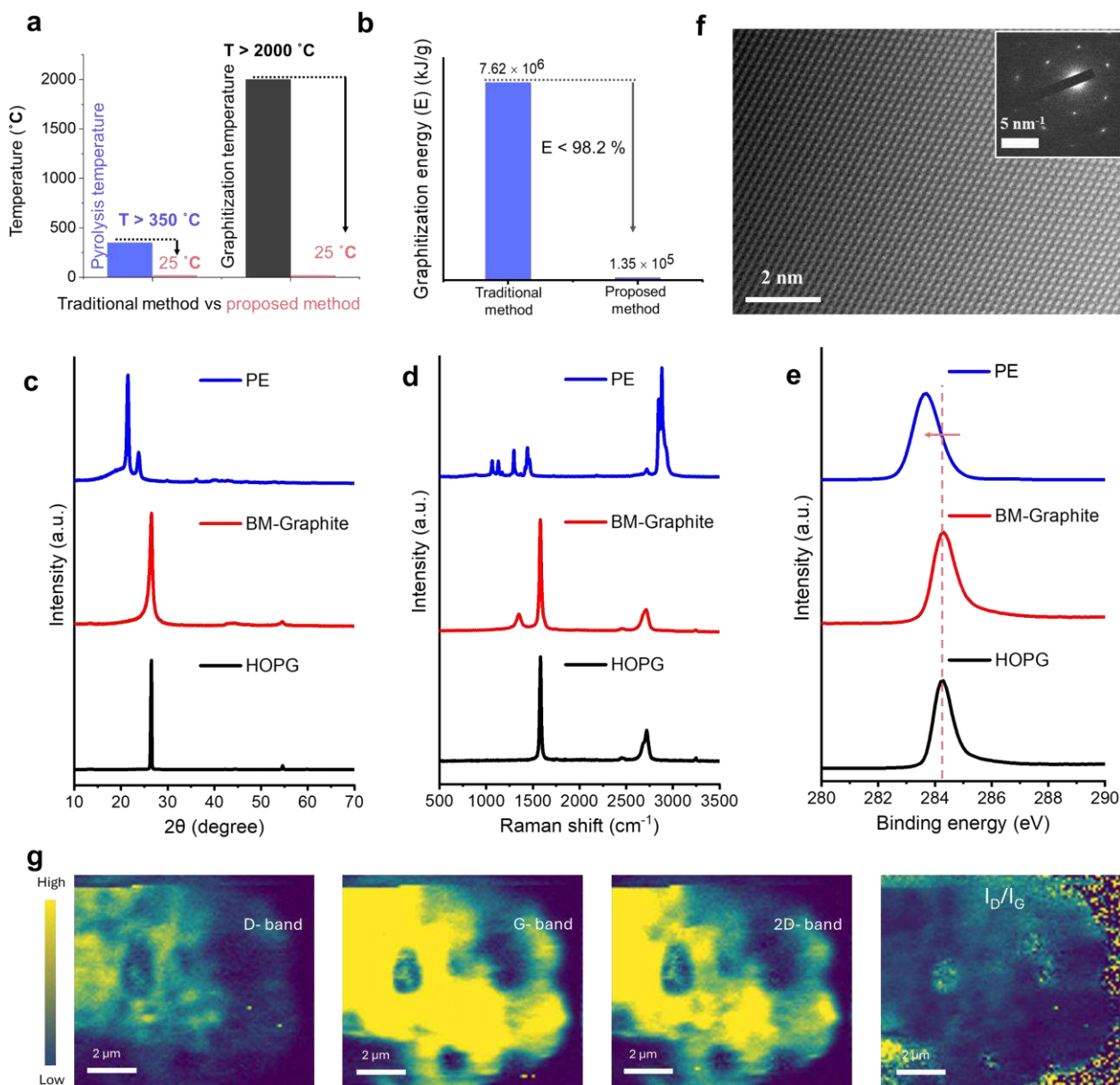


Figure 2 | Structural characterization of BM-Graphite synthesized via mechanochemistry.

(a) Comparison of the process temperature for ball milling (proposed method) with conventional pyrolysis routes and high-temperature graphitization. (b) Comparative energy consumption for graphite production on a gram scale. (c) X-ray diffraction patterns of PE, BM-Graphite, and HOPG. PE exhibits characteristic peaks at 21.4° and 23.76° , corresponding to the (110) and (200) planes of crystalline polyethylene. BM-Graphite displays a sharp peak at 26.56° , consistent with graphitic (002) stacking. (d) Raman spectra of PE, BM-Graphite, and HOPG. The emergence of distinct D, G, and 2D bands in BM-Graphite, with a 2D peak downshifted to 2703 cm^{-1} , indicates multilayer graphene formation with low disorder ($I_D/I_G = 0.16$). (e) High-resolution X-ray photoelectron spectroscopy of the C 1s region, showing a dominant sp^2 signal for BM-Graphite centered at 284.5 eV , similar to HOPG, and confirming the removal of sp^3 -rich features present in PE. (f) High-resolution transmission electron microscopy image of BM-Graphite, revealing well-ordered, defect-free graphitic layers. Selected area electron

diffraction pattern of BM-Graphite, showing sharp, discrete diffraction spots indicative of high crystallinity. (g) Raman mapping of BM-graphite showing the D-to-G intensity ratio, where yellow regions denote high signal intensities and green areas represent lower intensities.

Comparative analysis reveals a substantial reduction in processing temperature, exceeding 92% relative to pyrolysis (350 °C) and 98% compared with conventional graphitization (~2000 °C) (Figs. 2a). The present low-temperature route, conducted at 25–29 °C (Figs. S9), markedly simplifies the synthesis while mitigating industrial thermal emissions, thereby aligning with global net-zero objectives^{21,22}. Consequently, the graphitization energy requirement is reduced by up to 98.2%, dropping from the conventional $7.62 \times 10^6 \text{ kJ g}^{-1}$ to only $1.35 \times 10^5 \text{ kJ g}^{-1}$ (Fig. 2b) (energy calculation, page SI21)²³. Synthesized graphite was found to exhibit an interlayer distance of 0.3354 nm corresponding to the (002) planes, as determined by Bragg's equation (S – eq. i). The corresponding two-theta value for this peak was 26.56 degrees (Fig. 2c), consistent with theoretical values²⁴ and the HOPG structure (26.52 degrees). The average number of polyarene rings, determined from XRD analysis (S–eqs. ii and iii), was found to be 31, while Raman mapping of the 2D-to-G intensity ratio (I_{2D}/I_G) confirmed the uniformity of the layer structure (Fig. S10). The observed downshift in the 2D band of BM-Graphite (2703 cm^{-1}) in the Raman spectra (Fig. 2d), compared to HOPG (2715 cm^{-1}), can be attributed to a reduction in the number of graphene layers²⁵. This downshift results from the momentum conservation of two phonons, which is closely related to the electronic band structure and stacking of rings^{25,26}. The Lorentzian fitting of the 2D band shows a full width at half maximum (FWHM) and asymmetry comparable to those of highly ordered pyrolytic graphite (HOPG), indicating that the synthesized graphite possesses high structural quality (Fig. S10)^{25,26}. Based on the intensity ratio ($I_D/I_G = 0.16$), it can also be inferred that the sample consists of multilayer graphene, aligning with the XRD calculations. In a comparative high-resolution X-ray photoelectron spectroscopy (XPS) C1s spectrum (Fig. 2e), BM-Graphite exhibits a peak position (~284.5 eV) similar to that of HOPG (284.5 eV), and the comparable FWHM indicates a low distribution of energy levels.²⁷ This suggests a low content of foreign elements, such as oxygen, in the BM-Graphite, confirming a high percentage of sp^2 carbon (~92%) (Fig. S11).

The observed redshift in the XPS C1s peak for PE in Figure 1e is attributed to its sp^3 hybridization and the presence of hydrogen atoms bonded to the carbon atoms. Direct CHNSO elemental analysis verified that the sample consists exclusively of carbon and oxygen, exhibiting a weight ratio of 11.5 and a molar ratio of 15.3 (Fig. S12), confirming the high purity of BM-Graphite.

High-resolution transmission electron microscopy (HRTEM) provides strong evidence for the structural quality of the BM-Graphite, consistent with Raman spectroscopy results and the 92% sp^2 carbon fraction observed in XPS.

Furthermore, the HRTEM image reveals a largely defect-free surface (Fig. 2f), while the distinct dotted pattern in the selected-area electron diffraction (SAED) further confirms its crystallinity.²⁸ This observation is corroborated by wide-angle (2D) XRD and its corresponding 1D profile, which exhibit a pronounced in-plane diffraction intensity near $q = 43.5 \text{ \AA}^{-1}$, consistent with the surface uniformity observed in HRTEM (Fig. S13). These findings suggest that the polyarene rings are well-aligned with a high abundance of sp^2 -hybridized carbon atoms.

Raman mapping across a large sample area reveals the dominance of the G and 2D bands throughout the region, in agreement with HRTEM and complementary analyses, resulting in a uniformly low I_D/I_G ratio (Fig. 2g). Unlike HOPG²⁹, the origin of the D band in this sample can be attributed to minimal defects or grain boundaries. The integration of XPS analysis (confirming high sp^2 carbon content), CHNSO elemental composition ($C/O \approx 15.3$ mol ratio), and 1D XRD, alongside TEM imaging, shows good quality output with lower defects and well-aligned layers.

Mechanistic understanding of mechanochemical graphitization

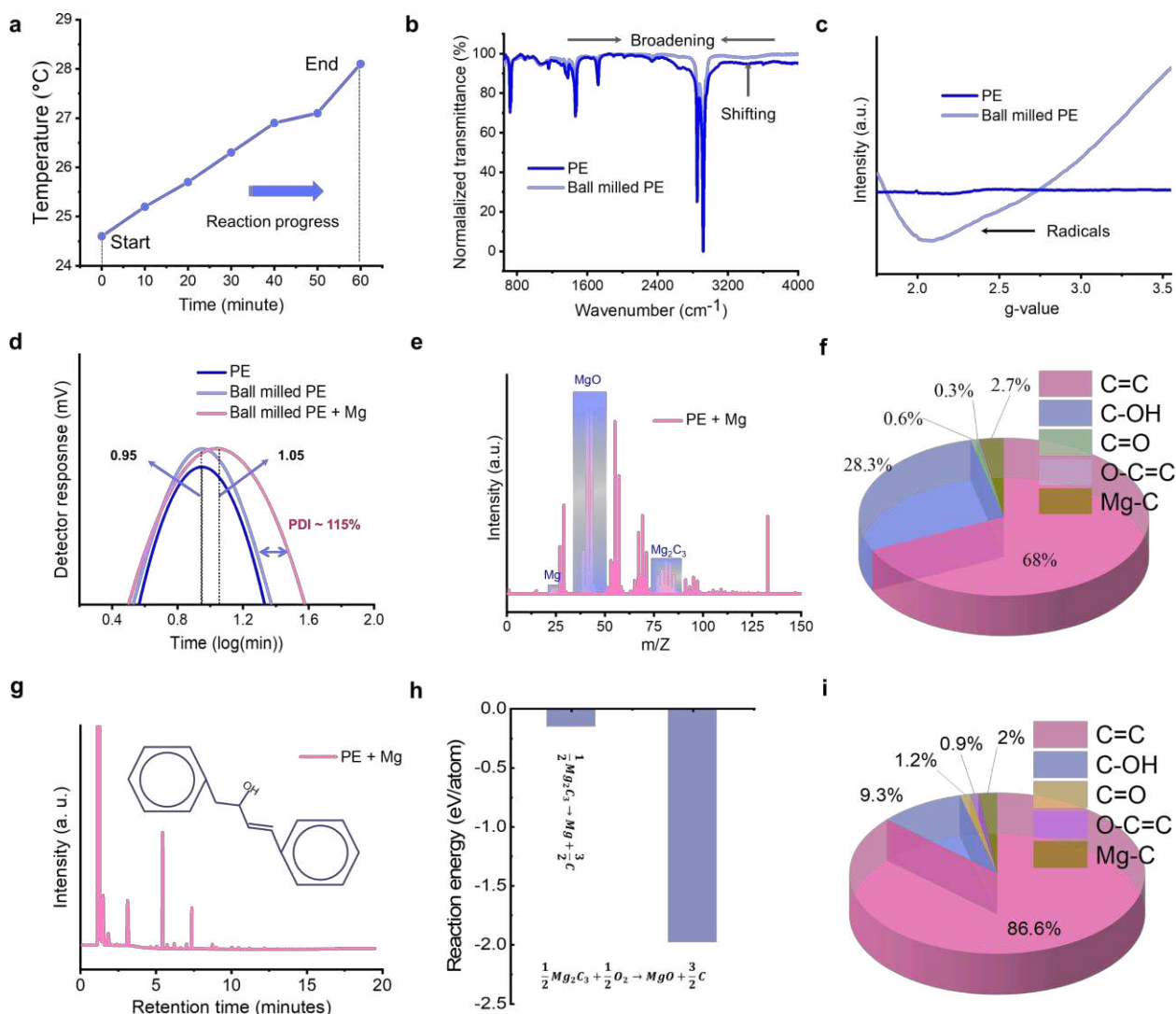


Figure 3 | Proposed mechanism for the conversion of PE into graphite via mechanochemical activation with Mg. (a) Average temperature variation of the ball milling system remains within ambient conditions (25–29 °C), confirming a heat-free transformation. (b) FTIR spectra of PE and ball-milled PE. (c) EPR spectra of PE and ball-milled PE. (d) GPC analysis shows that ball milling with Mg increases the PDI by 115.4% (red line). (e) TOF-SIMS analysis of PE+Mg (PEM) reveals the formation of Mg₂C₃ and MgO, alongside residual hydrocarbon fragments (non-shaded regions correspond to hydrocarbons). (f) Areal pie chart of the C 1s XPS analysis of PEM, showing 68% sp² carbon. (g) GC-MS analysis of gaseous products collected immediately after the reaction. (h) DFT-derived reaction pathway and corresponding energies, indicating favorable routes toward graphitization. (i) Areal pie chart of C 1s XPS spectra of PEM+Al, further confirming Al's role in promoting graphitization (86.6% sp² carbon).

Ball milling PE with Mg or other reactive metals swiftly promotes graphite formation, with D and G bands emerging and polymer peaks vanishing from the mixture in less than 30 minutes (see SI

optimization section). We believe under milling, activated C–C bonds and excited Mg atoms¹⁸ react to yield transient carbides, which further dissociate, producing graphite and MgO.

To determine whether the transformation occurs exclusively under ambient mechanochemical conditions, temperature evolution during ball milling was monitored using infrared (IR) thermography at regular 10-minute intervals. As shown in Fig. 3a and Supplementary Fig. 9, the temperature remained within a narrow range of 25–29 °C throughout the process for multiple samples. This negligible thermal fluctuation confirms that the transformation proceeds via a dominantly mechanochemical route, with no significant thermal contributions.

To probe the role of mechanical energy in activating PE, we first examined the effect of ball milling in the absence of any reagent. Fourier-transform infrared (FTIR) analysis (Fig. 3b) of pristine PE reveals broad C–H stretching bands centered at ~2917 and ~2848 cm⁻¹, characteristic of methylene units and associated with vibrational coupling and inter-chain interactions³⁰. After milling (BPE), these bands become sharper and more resolved, with increased transmittance in the high-frequency region, indicating altered crystallinity, reduced conformational disorder, and possible chain scission³¹.

Complementary electron paramagnetic resonance (EPR) measurements (Fig. 3c) show that while pristine PE is EPR-silent, BPE exhibits a clear radical signal. These findings are consistent with previous reports on mechanochemical degradation, indicating that mechanical activation promotes homolytic C–C bond cleavage and generates carbon-centered radicals (“mechanoradicals”)³². These radicals are likely stabilized within amorphous or disordered regions, as evidenced by peak broadening in the XRD data (Fig. S2), which reflects reduced crystallinity³³.

Despite radical generation, BPE retains the key vibrational and structural features of PE in FTIR (Fig. 3b), Raman, and XRD (Fig. S2), suggesting that in the absence of a reactive co-reagent, radicals either recombine or undergo minor rearrangement. Gel permeation chromatography (GPC, Fig. 3d) supports this interpretation: while the dispersity of BPE increases slightly relative to PE, the overall molecular weight distribution remains similar. These findings indicate that mechanical energy alone induces bond cleavage but does not drive significant chemical transformation, highlighting the necessity of a reactive

co-reagent to channel the radical-based reaction.

The introduction of Mg, a highly reactive metal, dramatically alters this behavior. GPC (Fig. 3d) reveals a 115.4% increase in dispersity (PDI from 1.3 to 2.8), signifying substantial molecular fragmentation. Raman and XRD analyses (Figs. 1e, S1) show characteristics of graphitic carbon, suggesting that Mg facilitates a radical-mediated transformation pathway culminating in graphitization. This hypothesis is further supported by Time-of-Flight Secondary Ion Mass Spectrometry (TOF-SIMS) (Fig. 3e) and XPS (Fig. 3f), which reveal the formation of Mg_2C_3 and sp^2 -rich carbon structures in ball-milled PE with Mg. Additionally, gas chromatography-tandem mass spectrometry analysis of the gaseous products collected from the vial reveals the presence of MgO (Fig. S14). Gas chromatography mass spectrometry (GC-MS) analysis of the gaseous products released during ball milling was performed over a molar mass range of 10–1000 Da. Upon expanding the retention time data (Fig. 3g), a variety of hydrocarbons were identified (Fig. S15). Interestingly, excluding a few linear compounds such as 1-hexene and n-heptane, most of the detected species exhibited ring structures, including 1-(2-benzylphenyl)-2-propyn-1-one, 1,4-diphenylbut-3-ene-2-ol, and 4-tert-butyl-2-tert-pentylphenol.

These findings suggest that the detected gases are intermediates in the graphitization process, as the predominance of cyclic species indicates a transitional state between polyethylene and fully graphitized carbon. Although our current instrumentation does not allow for the direct detection of H_2 , we postulate that hydrogen either participates in the formation of compounds such as CH_3OH or is released as H_2 , consistent with reported hydrogen evolution during polymer recycling³⁴.

Collectively, these results demonstrate that the addition of Mg promotes the cleavage of polymeric C–C bonds, generating carbon-centered mechanoradicals, which subsequently undergo transformation into carbides and, ultimately, graphitic carbon. Since Mg_2C_3 is a metastable carbide³⁵, it further dissociates into graphite upon continuous ball milling. The feasibility of Mg_2C_3 formation and its low stability are also verified by theoretical calculations (Fig. S17a), as well as experimental evidence (Fig. 3e). The presence of a small amount of carbide (low-intensity carbide peak) (Fig. 3e), along with a highly intense peak corresponding to MgO, can be interpreted as the carbide (Mg_2C_3) dissociating into graphite and

MgO upon continuous ball milling and reaction with oxygen, as supported by DFT calculations (Fig. 3h). However, the conversion of carbide to graphite appears favorable regardless of the presence of oxygen. The presence of MgO and Mg₂C₃ was confirmed by XRD (Fig. S1c) and TEM (Fig. S17d). The XRD data reveals peaks near 37° and 43.5°, corresponding to the (120) plane of Mg₂C₃ (PDF card #010810587) and the most stable (200) plane of MgO (PDF card #010855620), respectively. The measured interlayer distances via TEM also align with these peaks: 0.25 nm for the (120) plane of Mg₂C₃ and 0.21 nm for the (200) plane of MgO (Fig. S17d).

We performed DFT calculations using the Vienna Ab initio Simulation Package (VASP)^{36,37} to explore potential reaction paths. The generalized gradient approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) functional (35) was used for the exchange-correlation function, and all calculations were spin-polarized. In all cases, the plane-wave energy cutoff was set to 450 eV, the k-points mesh density to $2\pi \times 0.04 \text{ \AA}^{-1}$, and the self-consistent field energy convergence criterion to 10^{-6} eV .

Two reaction paths were considered:



The reaction energies, E_{path1} and E_{path2} for path 1 and path 2, respectively, were calculated using the following equations:

$$E_{\text{path1}} = \frac{[(E_{\text{Mg}} + \frac{3}{2}E_{\text{C}}) - \frac{1}{2}E_{\text{Mg}_2\text{C}_3}]}{N_{\text{Mg}} + N_{\text{C}}} \quad (1)$$

$$E_{\text{path2}} = \frac{[(E_{\text{MgO}} + \frac{3}{2}E_{\text{C}}) - (\frac{1}{2}E_{\text{Mg}_2\text{C}_3} - \frac{1}{2}E_{\text{O}_2})]}{N_{\text{Mg}} + N_{\text{O}} + N_{\text{C}}} \quad (2)$$

Where E_{Mg} , E_{C} , $E_{\text{Mg}_2\text{C}_3}$, E_{MgO} , and E_{O_2} represent the energy of a Mg atom in bulk (hcp), a C atom in bulk (graphite), Mg₂C₃, MgO, and an O₂ molecule, respectively. N_{Mg} , N_{C} , and N_{O} represent the number of Mg, C, and O atoms in the reactants. The calculated reaction energy for Path 1 is -0.153 eV/atom , indicating an exothermic reaction. However, the reaction energy for Path 2 is -1.986 eV/atom , which

is highly exothermic and significantly lower than that of Path 1. The calculations suggest that Path 2 is the more favorable reaction process (Fig. 3h). Thus, MgO and graphite were formed through the reaction between Mg_2C_3 and oxygen molecules during ball milling under ambient conditions, consistent with our experimental observations. This approach enables the synthesis of graphite while avoiding the need for harsh or inert environments, high temperatures, etc (See Mechanism section in the SI (page 29)).

Based on combined experimental observations and DFT analysis, we propose a three-step mechanistic model for the ambient-temperature graphitization of PE, (i) The formation of carbon radicals and the excitation of Mg due to stress and strain (Mg^*), which enhance reactivity by making Mg more prone to losing electrons considering its low ionization energy; (ii) The reaction between carbon radicals and Mg^* to form Mg_2C_3 (as an intermediate product) while releasing hydrogen; (iii) The reaction of Mg_2C_3 with oxygen to form graphite and MgO.

Remarkably, stoichiometric calculations indicate a conversion yield exceeding 80% (see SI: mechanism section). This pathway stands in contrast to the conventional fate of radicals in polymers, crosslinking, oxidative degradation, or amorphization, which predominates in the absence of a reactive metal. The ability of Mg to trap carbon fragments as metastable carbides thus plays a pivotal role in steering the transformation toward graphitic products.

To further understand the role of Al, a two-step ball milling process was performed. First, PE was ball-milled with Mg, as previously described. Subsequently, Al was introduced into the system. Since Al is known to enhance the degree of graphitization^{38–41}, Raman spectroscopy (Fig. 1g) reveals a sharper and asymmetric 2D band and a suppressed D band ($I_D/I_G = 0.16$) in the Mg–Al system (PEMA), compared to PEM ($I_D/I_G = 0.76$), indicating a more ordered graphitic structure.

XPS analyses of the PEM (Fig. 3f) and PEMA (Fig. 3i) samples provide further evidence of the role of Al by highlighting differences in their graphitization degrees. The high intensity of metal-carbon (M–C) bonds and the low intensity of $\text{C}=\text{C}$ (sp^2) bonds in the PEM sample (Fig. S17b) indicate a lower degree of graphitization compared to the PEMA sample (Fig. S17c), which exhibits a lower intensity

of M–C bonds and a significantly higher intensity of C=C (sp^2) bonds. This difference is quantified by the areal pie chart from the XPS analysis, showing that the PEMA sample contains 86.6% sp^2 carbon, while the PEM sample contains only 68.07%, confirming the role of Al as a graphitization promoter in the PEMA sample (Fig. 3i).

Properties and applications of upcycled BM-Graphite

After purification, BM-Graphite exhibits an aggregated, sheet-like morphology (Fig. S18), characterized by a dominant sp^2 carbon content (91.8%) and minimal oxygen presence (Fig. S11). However, the slightly higher FWHM in the XPS of BM-Graphite (0.89 eV) compared to HOPG (0.73 eV) suggests a higher presence of oxygen or energy states than in HOPG (Fig. S11)²⁷. TEM Energy-dispersive X-ray spectroscopy (EDS) analysis of BM-Graphite further supports the findings from XPS, showing negligible amounts of Mg and Al, with over 97% carbon and nearly 2% oxygen (Fig. S19). The FTIR spectra (Fig. S20) show only one vibration corresponding to aromatic C=C bonds. The absence of C–O bonds in solid-state ^{13}C nuclear magnetic resonance (^{13}C -NMR) (Fig. S20) and FTIR further supports the XPS findings. The stability of the graphite and the absence of functional groups were analyzed using thermogravimetric analysis (TGA). Figure 4a confirms the comparable thermal stability of BM-Graphite to that of HOPG, differing significantly from PE, which does not maintain stability above 500 °C. The TGA curve indicates a weight loss of only 8.88% at high temperatures (900 °C) for BM-Graphite, demonstrating its high thermal stability. For PE, 99.8% of the weight loss occurs before reaching 500 °C, while for HOPG, weight loss is less than 1%, all under an inert (N_2) atmosphere.

Beyond thermal stability, electrical conductivity is a key indicator of graphite quality. As shown in Figure 4b, the average sheet resistance of BM-Graphite is $74.83 \text{ m}\Omega \text{ sq}^{-1}$, closely matching that of HOPG, which exhibits a sheet resistance of $74.65 \text{ m}\Omega \text{ sq}^{-1}$ (measured via four-probe resistivity, Fig. S22). Comparable resistivity reflects the long-range electronic conjugation enabled by graphitic domains. Additionally, nitrogen adsorption–desorption isotherms reveal that BM-Graphite possesses a

significantly larger surface area than HOPG, which is attributed to its mesoporous nature (Fig. S21).

The high surface area and comparable electrical conductivity enable BM-Graphite to outperform HOPG in electrochemical performance (Fig. S23). We used BM-Graphite as a supporting material for the cathode (V_2O_5) in a zinc-ion battery (ZIB)⁴². V_2O_5 (VO) is considered a promising candidate for ZIB cathodes⁴³, however, a commercial VO cathode requires prolonged activation (Fig. S23), requiring 30 cycles to reach full capacity, and its cycling stability is also poor (Fig. S23, S24). To address these issues, we explored graphite as a potential composite material due to its ability to enhance conductivity and stability in VO cathodes in aqueous media (Fig. S24).^{8,44}

The BM-Graphite/VO composite exhibits a sheet-like morphology (Fig. S25) and delivers a high specific capacity of 225 mAh g^{-1} at 0.5 A g^{-1} , outperforming both the HOPG composite (150 mAh g^{-1}) and V_2O_5 alone (75 mAh g^{-1}) (Fig. S23). While the HOPG composite initially performs better at 5 A g^{-1} , the BM-Graphite composite shows superior long-term stability, retaining 96% of its capacity after 500 cycles (vs. 79.2% for HOPG and 51.1% for V_2O_5).

The enhanced performance of the BM-Graphite composite cathode is primarily attributed to its low charge-transfer resistance, which facilitates improved kinetics and mitigates cathode degradation (Fig. S23). In addition, the larger surface area and more favorable composite structure of BM-Graphite contribute to better compatibility and interfacial uniformity with VO compared to HOPG, as confirmed by XRD, Raman, and SEM analyses (Fig. S25). Together, these factors synergistically enhance the overall electrochemical performance.

Through the mechanical exfoliation of BM-Graphite, graphene can also be produced (Fig. S26), characterized by a Raman shift and Lorentz fit for the 2D band and an I_{2D}/I_G ratio approaching 1.

Furthermore, the straightforward application of recycled graphite produced through this process, without additional refinement, is demonstrated in the fabrication of writing and coloring pencils (Fig. 4c).

Applicability of proposed chemistry for other polymers/wastes

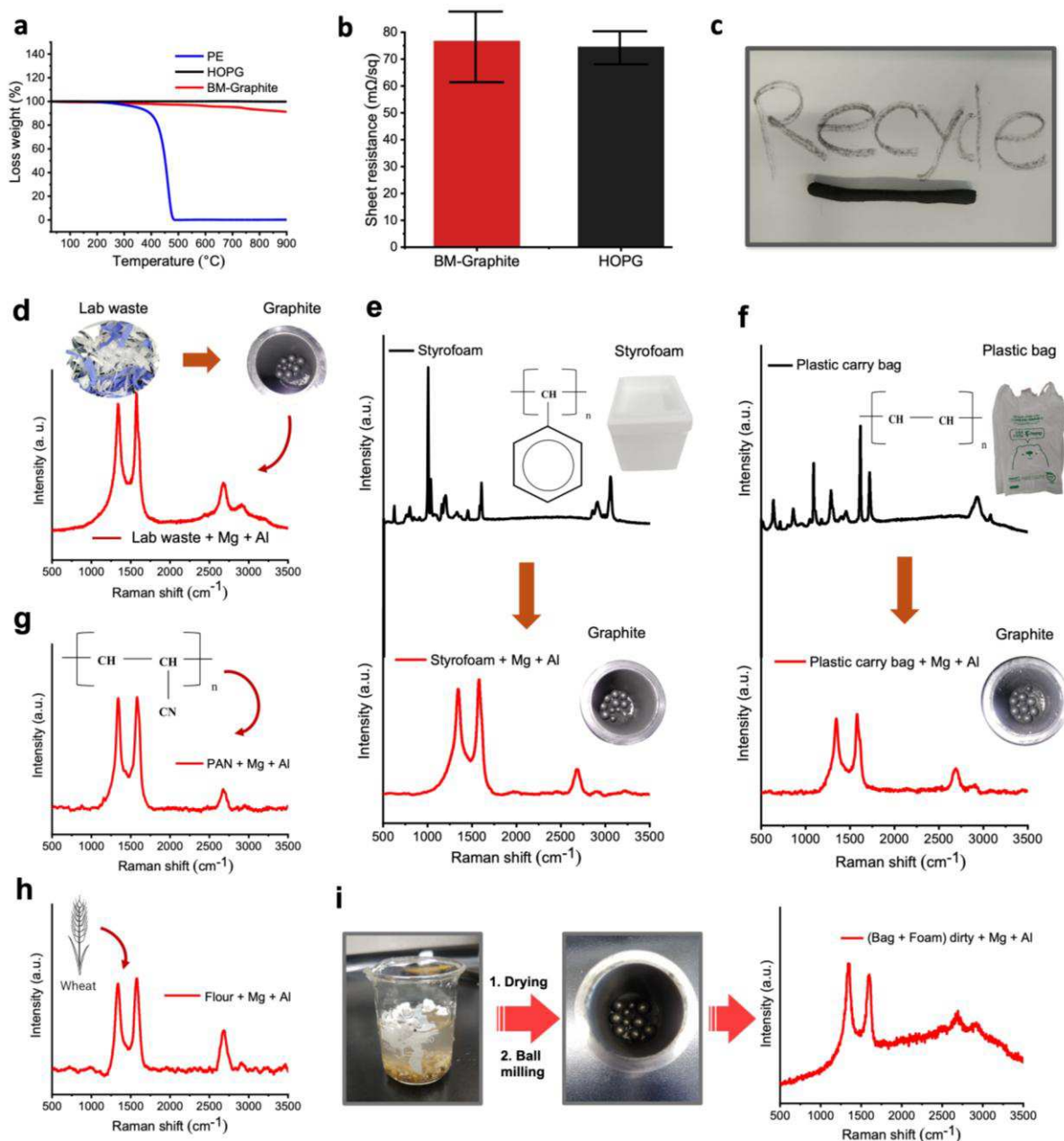


Figure 4 | Properties of BM-Graphite and applications of our chemistry on commercial waste. (a) TGA shows high thermal stability of BM-Graphite, comparable to HOPG and significantly higher than PE. (b) Four-point probe measurements indicate that the average sheet resistance of BM-Graphite is comparable to that of HOPG. (c) Pencil made from an upcycled mixture of carry bags and Styrofoam products, without additional processing such as purification, demonstrating real-world utility. (d) Graphitization of a mixture of laboratory waste. (e) Conversion of Styrofoam waste into graphite. (f) Graphitization of plastic carry bags. (g) Graphitization of traditionally non-graphitizable polymer PAN. (h) Graphitization of bio-waste (wheat flour). (i) Graphitization of a contaminated mixture of polyethylene and polystyrene.

Extending this methodology to other major polymers without pretreatment, such as PS, PAN, and PVA, reveals that polymer crystallinity plays a crucial role in determining the extent of graphitization (Fig. S27). Polymers with higher degrees of crystallinity^{45,46} tend to exhibit greater degrees of graphitization. For instance, PS, an amorphous polymer, displays limited graphitization due to its low crystallinity. In contrast, PVA and PAN, both semi-crystalline polymers, demonstrate improved graphitization, while PE, a crystalline polymer, exhibits the highest degree of graphitization (Fig. S28, S29). Remarkably, even non-graphitizable polymers, such as polyacrylonitrile (PAN)¹⁴, undergo graphitization under ambient conditions due to their semi-crystalline nature. Based on these observations, we hypothesize that increased amorphous character leads to the formation of a rubbery structure upon pressurization, which enhances tensile tolerance and reduces susceptibility to graphitization.

The versatility of the method is demonstrated by its direct application to an unsorted mixture of common laboratory waste, including hand gloves, tissue paper, weighing boats, and pipettes, without pretreatment or material separation. Remarkably, Raman spectroscopy confirmed the successful conversion of this heterogeneous blend, composed of plastics, rubbers, paper, and hybrid materials, into graphitic carbon (Fig. 4c). This highlights the method's tolerance to real-world contamination and material variability.

Furthermore, individual components were tested separately, each yielding graphitized products (Fig. S30), thereby validating the process across a broad spectrum of feedstocks. Widely used consumer plastics, such as Styrofoam (polystyrene) and polyethylene carry bags, also underwent effective graphitization under identical conditions (Fig. 4e, 4f). Notably, polystyrene, typically considered non-graphitizable due to its tendency to crosslink via Friedel–Crafts chemistry, which inhibits graphitic layer formation⁴⁷, was graphitized under mechanochemical conditions. Non-graphitizability is typically associated with polymers having low helium densities, generally below 2.0 g cm⁻³. For instance, polyacrylonitrile (PAN), with a density of around 1.57 g cm⁻³, is considered non-graphitizable⁴⁸. However, due to its semicrystalline structure, PAN also undergoes graphitization (Fig. 4g).

We further validated the method using natural biowaste, such as wheat flour (Fig. 4h), rice, cabbage,

and potato peels, and observed successful graphitization (Fig. S29). These results indicate that ball milling with reactive metal reagents can efficiently convert most of the carbon-based waste into graphitic carbon. These findings demonstrate that mechanochemical approach effectively overcomes the long-standing challenges associated with graphitizing polymeric waste materials.

To simulate practical, real-world waste scenarios, we investigated two types of plastic contamination: food-related residues and soil-/sand-based contaminants (Figs. 4i, S29). Tests included common paper products (e.g., writing paper, coffee cups) as well as plastics intentionally contaminated with soil or food residues. To simulate dirty waste, Styrofoam and plastic carry bags were immersed in sand water to allow dirt to adhere to the surfaces. After drying at 70 °C for 12 hours, the sample mass increased from 100 mg to 135 mg due to adhered dirt (Fig. 4i). Raman analysis revealed that ball milling with Mg and Al successfully converted the contaminated plastics into graphite. This result demonstrates that the method is effective even without the sorting, purification, or cleaning steps that typically complicate conventional recycling processes.

It is worth noting that the feasibility of thermal graphitization diminishes in polymers with prolonged liquid-phase transitions ⁴⁹. However, mechanochemical graphite synthesis primarily depends on the degree of crystallinity and is relatively unaffected by liquid-phase transitions. This attribute broadens the applicability of the method to a wide range of plastics.

Conclusions

We propose a versatile, eco-friendly strategy to upcycle diverse carbonaceous wastes, including plastics, biomass-derived waste, and paper, into graphite, a modern critical material. Conducted under ambient conditions, this approach offers a sustainable alternative to conventional energy-intensive and solvent-based graphitization and waste recycling methods. This process achieves temperature reductions of 92% compared to pyrolysis and over 98% versus traditional graphitization, resulting in a 98.2% decrease in energy consumption per gram. By eliminating the need for external heat and solvents, while demonstrating high tolerance to feedstock variability and contamination, this method provides a direct, sustainable pathway for carbon waste valorization. By integrating mechanochemical activities of elements with DFT calculations, this study establishes a novel room-temperature graphitization route with deep mechanistic insights. The demonstrated applicability to both laboratory reagents and commercial waste mixtures highlights its potential to scale the upcycling of diverse carbon-based materials. Beyond its environmental benefits and contribution to understanding solid-state radical-based chemistry, BM-graphite demonstrated electrochemical performance in zinc-ion batteries comparable to that of commercial graphite. Ultimately, catalytic mechanochemistry will provide a compelling alternative to conventional recycling protocols, enabling a cleaner, simpler and potentially scalable route for the sustainable valorization of carbonaceous waste.

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