Review

A systematic literature review on the conversion of plastic wastes into valuable 2D graphene-based materials

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ABSTRACT

Increasing the economic attractiveness of plastic waste reusing/recycling is expected to contribute towards reducing their input in the environment. The use of plastic wastes as feedstock for the synthesis of added-value carbon materials has been studied in this context. However, there is a lack of a systematic review of the published works on this topic. Bearing this in mind, a systematic review was carried out in this study, covering the available literature on the conversion of plastic wastes into carbon materials. Clearly defined methodologies/criteria were accordingly established. 142 studies were selected for qualitative/overall analysis, including type/condition of plastic (pristine or waste), and type of carbon materials obtained. It was found that most of the studies report the utilization of plastic wastes (75%); and that the most representative materials obtained are carbon nanotubes (CNTs; 47.1% of the studies under evaluation), activated carbons (ACs; 22.2%) and 2D graphene-based materials (9.2%). Nevertheless, despite already being the third most significant group of carbon materials produced from plastic wastes, none of the 12 review articles available in the literature is fully devoted to the conversion of plastic wastes into 2D graphene-based materials. Therefore, the literature available on this topic was thoroughly reviewed for the first time. These studies report the synthesis of monolayer, few-layer and multi-layer graphene (including flash graphene) obtained through 4 main synthesis methodologies: (i) thermal decomposition of the plastics directly over a metal substrate; (ii) prior thermal decomposition of the plastics, with the resulting hydrocarbon gases released being fed to a chemical vapour deposition (CVD) system containing the metal substrate; (iii) thermal decomposition followed by ball milling and microwave sintering; and (iv) flash Joule heating (FJH).

1. Introduction

The world's plastics production has been increasing stiffly over the years. Only in 2019, the global production of plastics reached nearly 370 million metric tonnes, with *ca.* 15.7% of those materials being produced in Europe [1]. Recent estimates suggest a cumulative plastic waste generation of over 25,000 million metric tonnes by 2050 [2]. Among these, 36.4% are expected to be discarded in landfills or in the environment; a similar fraction (36.4%) would be incinerated, and only 27.2% will be recycled [2]. These forecasts clearly point out that plastic wastes are being mismanaged, mainly because current technologies are unable to promote proper reusing/recycling of these materials [3]. As a consequence, soil, freshwater and oceans are becoming increasingly contaminated worldwide by plastic wastes, with subsequent impacts on

all living species and natural ecosystems [4]. As recently reviewed, both preventive strategies to reduce the input of plastic wastes to the environment (*i.e.*, upstream responses), and measures to mitigate their impact once released to the environment (*i.e.*, downstream responses), have been considered to tackle the global plastic waste crisis [4]. Proper waste management falls within the scope of upstream responses. However, several difficulties have been hindering the recycling of plastic wastes, such as the lack of economic attractiveness of the resulting products [4].

Using plastic wastes as feedstock for the production of added-value products and/or materials has been proposed as the boost needed to increase the attractiveness of plastics recycling. In particular, recent research efforts have been focused on proposing alternatives to the conventional inclusion of plastic residues in bitumen [5] and

construction products [6]. Among them are included (i) the production of gaseous and liquid fuels and chemicals [7,8], (ii) the production of composite materials with enhanced properties for high-end applications [9,10], and (iii) their conversion into carbon materials [11–22].

There has been a growing interest on carbon materials due to their unique properties, such as high surface area, porosity and electronic conductivity, rich/tailorable surface chemistry, and structural stability at high temperatures [23–25]. As a consequence, carbon materials have been increasingly employed in very different fields, including catalytic [26], environmental [27], biomedicine [28], energy [29], electronic [30] and analytical [31] applications, among others. Moreover, the latest inventory on consumer products with nanomaterials revealed that carbon materials are already the second most used class of nanomaterials, being surpassed only by metal nanoparticles [32].

Bearing this in mind, in the present study a systematic review was carried out to assess the literature currently available on the conversion of plastic wastes into valuable carbon materials. In the following of a qualitative analysis of all research articles published in this field, the literature focusing on the synthesis of 2D graphene-based materials from plastic wastes was thoroughly reviewed for the first time.

2. Methodology

The literature search was performed on the Scopus database (access on June 2021), using the search strings detailed in Table S1 (given as Supplementary material). Briefly, the literature search combined terms for different carbon materials (*e.g.*, graphite, graphene, carbon fiber, carbon nanotube, activated carbon, etc.) with the terms "plastic" and "waste". Titles and abstracts of the retrieved articles were screened for relevance considering the following eligibility criteria: use of plastics as feedstock for the synthesis of carbon materials, original studies and review articles. Exclusion criteria: full text in a language other than English and lack of access to the full article.

Studies appearing to meet the inclusion criteria were then screened for duplicates and reviewed in full. Data extraction was undertaken independently by O. Vieira and R.S. Ribeiro. Relevant information of each reviewed study was gathered and organized in the form of a table previously designed in an Excel spreadsheet, including publication year, first author, title, highlights, type of article (original or review), type and condition (pristine or waste) of plastic, synthesis conditions, carbon materials obtained, characterization techniques employed, application and novelty. The classification of carbon materials was performed according to our detailed analysis of the reported results, which not always agrees with the claims/conclusions made by the authors of the corresponding studies (as discussed in Sections 3.2 and 4). Review articles were excluded prior to qualitative analysis, but used as part of the criteria for study selection.

3. Results and discussion

3.1. Overall analysis and study selection

The literature search performed on the Scopus database using the strings detailed in Table S1 yielded 1115 results (*cf.* Fig. 1). The titles and abstracts of those articles were screened for relevance and duplicates removed, leading to the selection of 153 articles for full-text reading. Among these publications, 11 review articles were found and excluded from the qualitative analysis. The resulting 142 articles were analysed thoroughly. The type and condition of the plastics used as feedstock for the synthesis of carbon materials were analysed first. Although the search strings included "plastic AND waste" (*cf.* Table S1), 25% of the studies under analysis employed pristine plastics (*cf.* Fig. 2a). Polypropylene (PP), polyethylene terephthalate (PET), polyethylene (PE), polystyrene (PS) and polyvinyl chloride (PVC) are the type of plastics most commonly used in those studies (*cf.* Fig. 2a). These results agree quite well with the current plastics market demand [4].
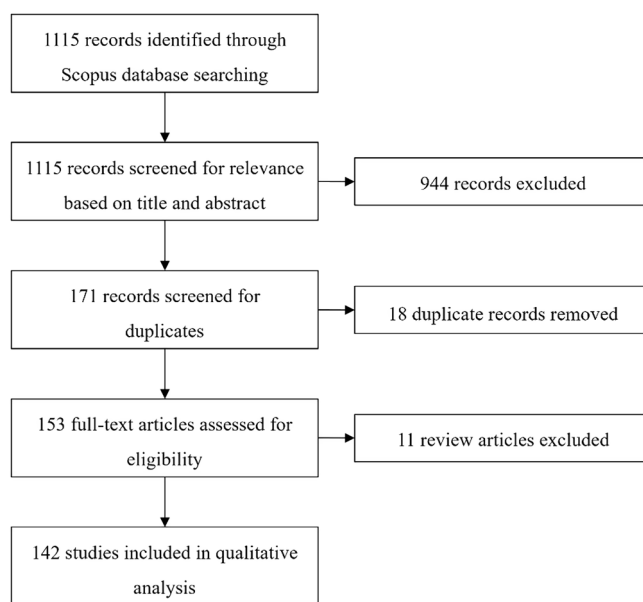

Fig. 1. Flowchart considered in the process of the study selection.

The conversion of plastic wastes into carbon materials was first reported in 2004, by Parra et al. [33]. Briefly, the authors prepared a series of activated carbons (ACs) upon pyrolysis of PET waste under inert atmosphere, followed by activation with CO_2, seeking to develop materials with high adsorption capacity for hydrogen [33]. Since 2004, several studies reported the synthesis of other carbon materials from plastics, including carbon nanotubes (CNTs), carbon microspheres, 2D graphene-based materials, carbon nanofibers (CNFs), graphite and fullerene. Indeed, the current research trend towards the conversion of plastic wastes into carbon materials is clearly shown in Fig. 2b. As observed, the amount of studies on this topic has been increasing continuously since 2004. Nearly half of those studies report the synthesis of CNTs, followed by ACs and 2D graphene-based materials (*cf.* Fig. 2c). On this regard, the materials denoted as "other" (*cf.* Fig. 2) refer mainly to composite materials, corresponding to mixtures of different carbon materials and/or carbon materials with other components, such as metals, polymers and/or fibres.

Most research articles on the conversion of plastic wastes into carbon materials are still focused on optimizing the synthesis conditions (53.5%, *cf.* Fig. 2d). Environmental applications represent 22.6% of the total number of articles, followed by electrochemical and energy field applications (21.2%). Among the articles aiming environmental applications, 73.5% are studies focused on the development of adsorbent materials for water treatment; while among the articles aiming electrochemical and energy field applications, 50.0% of the works are focused on the development of materials to be used in supercapacitors (*cf.* Fig. 2d).

The prevalence of studies on the synthesis of CNTs is clearly reflected in the number of review articles available in the literature. Indeed, 9 [11–17,21,22] out of the 11 review articles found through our systematic approach have addressed that topic (*cf.* Table S2). Regarding the synthesis of ACs from plastic wastes, 2 review articles were found [18,19]. However, although the studies on the production of 2D graphene-based materials from plastic wastes are already the third most significant group (*cf.* Fig. 2c) – with 14 original research articles published so far, no comprehensive review article on this topic was found in the literature. As observed in Table S2, the reviews performed by Din et al. [18] and Utetiwabo et al. [16] include some 2D graphene-based materials, but only those obtained under very specific conditions and/or prepared for a specific application. In the review of Din et al., only articles using PET as feedstock for the production of dye adsorbents were
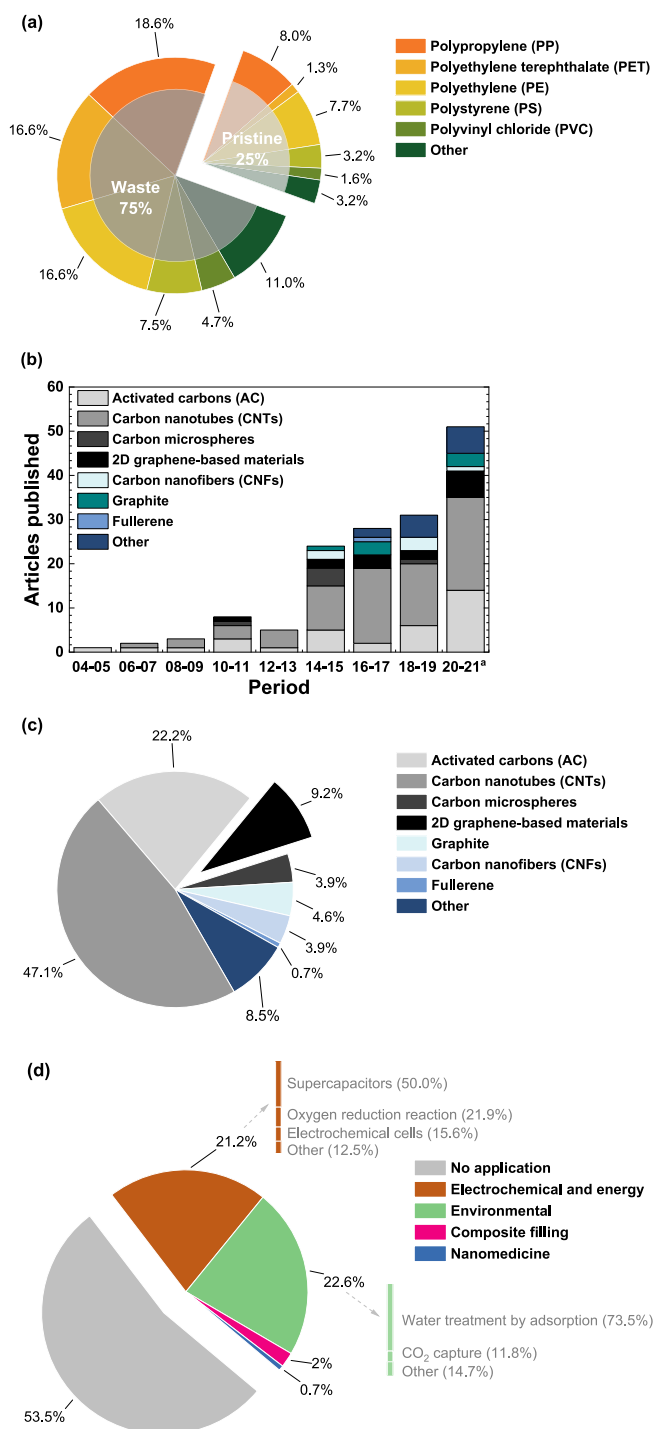
Fig. 2. (a) Type and condition of the plastics used as feedstock for the synthesis of carbon materials. (b) Evolution of the Scopus's indexed original research articles dealing with conversion of plastics into carbon materials. (c) Type of carbon materials produced from plastics. (d) Field of application of the carbon materials produced from plastics. Data collected from the 142 studies selected for qualitative analysis based on the criteria detailed in Section 2 and summarized in Fig. 1. Nevertheless, some of those studies report the synthesis of more than one carbon material, reason why the total amount of carbon materials displayed in (b) amounts to 153. [a] Data collected on June 29, 2021.

an emerging trend in the context of waste valorisation in Nigeria (the literature on the topic is not reviewed). It is noteworthy that the first review article in which the synthesis of 2D graphene-based materials from plastic wastes was addressed is that of Kwon et al. (not found using the search methodology described in Section 2, but herein included for the sake of clarity) [20]. The topics of this review included the synthesis of CNTs and 2D graphene-based materials from several carbon-rich residues, such as plastic wastes (*cf.* Table S2). Specifically, 3 original research articles on the conversion of plastic wastes into 2D graphene-based materials (published until 2017) were discussed in the review of Kwon et al. [20]. However, most of the studies on that topic were reported since then (*cf.* Fig. 2b). Bearing this in mind, the present review aims to overcome this gap, by conducting a literature review on the conversion of plastic wastes into 2D graphene-based materials, as summarized in Fig. 3 and detailed in the following Sections.
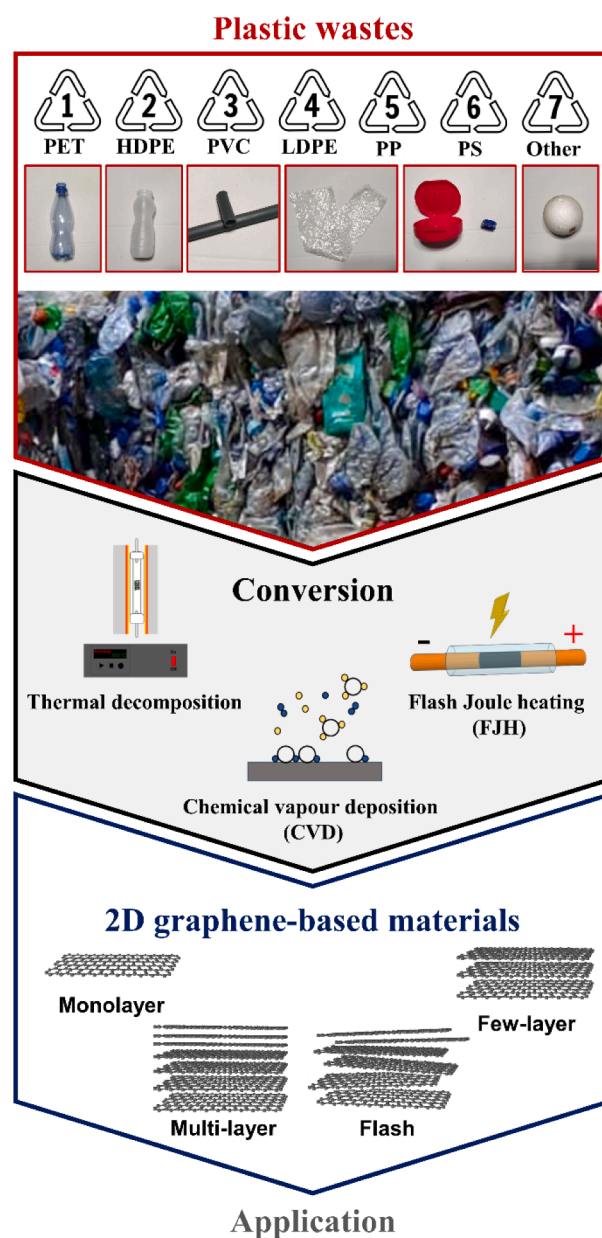
Fig. 3. Conversion of plastic wastes into valuable 2D graphene-based materials: schematic representation of the main topics covered in this review.

covered [18], while the review of Utetiwabo et al. included only carbon materials to be incorporated in electrodes for supercapacitors [16]. The conversion of plastic wastes into carbon materials (including graphene flakes) is discussed in the review of Nyakuma and Ivase [21], but only as

3.2. 2D graphene-based materials

Strictly speaking, graphene is a 2D monolayer of hexagonally arranged sp^2-bonded carbon atoms, whose thickness is that of a carbon atom, that is not an integral part of a carbon material, but is freely suspended or attached to a substrate, and characterized by a 2D gas of Dirac fermions [34–36]. 2D graphene-based materials are a broader class, which should not be limited by physical dimensions or the number of atomic layers, but rather defined by their properties - especially electronic properties [37]. Accordingly, these are highly anisotropic in electron mobility; with ballistic electron mobility in the x and y plane and, even when stacked graphene layers are present, with many orders of magnitude lower electron mobility in the z-axis [37]. The existence of such materials was predicted by the theoretical physicist Philip Wallace at the McGill University in Montreal, Canada, in 1947 [38]. However, the prove of the existence of graphene was obtained only in 2004, when A. Geim and K. Novoselov, from the University of Manchester, UK, isolated and identified graphene sheets for the first time [39], and for this pioneering work they were awarded the Nobel Prize in Physics, in 2010. Since then, graphene has received wide attention from the scientific community owing to its unique and remarkable properties [38]. Graphene has, for instance, astonishing electrical and thermal conductivity, with an electron mobility 100-fold higher than that of silicon and better heat conduction than that of diamond; it is the strongest material ever measured, with a mechanical strength 100-fold higher than that of steel (in spite of also being the thinnest material known to exist); it has good stretchability, allowing it to bend where other materials would snap [38]. Moreover, graphene has high specific surface area (theoretical value of 2630 m^2 g^{-1} for monolayer graphene), and high chemical and thermal stability [40,41].

2D graphene-based materials have been intensively used at the research level, including distinct applications such as electronics, electrodes for supercapacitors, electromagnetic interference shielding, preparation of polymer composites, sensors, catalysis and environmental protection, among others [40,42–44]. Nevertheless, although a considerable amount of commercial products is already available [32], the number of practical applications has been limited by the difficulty of producing high-quality 2D graphene-based materials in sufficient quantities and at an attractive cost [38]. Indeed, this is a common limitation of novel materials. For instance, decades of research were needed before establishing silicon in the technology market [38]. Therefore, research efforts should focus on enhancing current strategies for the production of 2D graphene-based materials.

Two different approaches can be considered to obtain 2D graphene-based materials: top-down and bottom-up [40]. In the top-down approach, graphene sheets are peeled from graphitic materials, while in the bottom-up approach graphene sheets are synthesized from a carbon source (cf. Fig. 4). The most commonly used top-down approach starts with a strong chemical oxidation of graphite, leading to the formation of graphite oxide [40,45]. Graphite oxide can be then thermally, mechanically or chemically exfoliated to graphene oxide (GO), followed by chemical, thermal, microwave, photo-chemical, photo-thermal or microbial/bacterial reduction to obtain the so-called reduced graphene oxide (rGO) – a nomenclature that allows distinguishing the materials obtained by this way [35,40,45]. This methodology results in high yields of 2D graphene-based materials at a relatively low cost; however, rGO usually possesses considerable amounts of structural defects and oxygen-containing functionalities, which affect the electronic properties and thus the quality of the material [45]. On the opposite, 2D graphene-based materials with high quality can be obtained through bottom-up methods, such as epitaxial growth on silicon carbide (SiC) and chemical vapour deposition (CVD) [40,45]; however, large-scale production is still a challenge with these bottom-up approaches [40,45]. Briefly, the formation of graphene sheets on the surface of SiC is accomplished upon sublimation of silicon in SiC, leaving behind carbon atoms that undergo graphitization [40,45]. This process is usually carried out under high
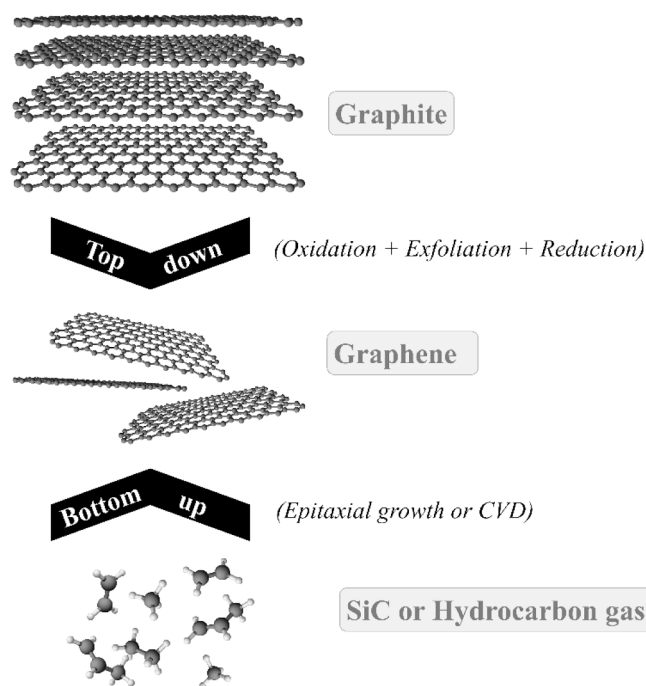
Fig. 4. Representation of top-down and bottom-up approaches for the synthesis of graphene sheets.

temperature (above 1000 °C) and ultra-high vacuum conditions [40,45]. In CVD, nucleation and growth of graphene sheets occur on the surface of transition metal and noble metal substrates (e.g., Fe, Ru, Co, Rh, Ir, Ni, Pd, Pt, Cu, Au and/or their alloys) upon exposure to a pure hydrocarbon gas (methane, acetylene, benzene are few examples considered within the literature) under high temperature (above 650 °C) and low pressure or ultra-high vacuum conditions [40,45]. 2D graphene-based materials can then be collected upon etching of the metal substrate [40,45]. Instead of adding a hydrocarbon gas to the CVD system, 2D graphene-based materials can also be obtained upon thermal decomposition of solid carbon sources placed directly on the top of the metal substrates [40]. Flash Joule heating (FJH) is an alternative bottom-up method, allowing the synthesis of bulk quantities of the so-called flash graphene, in theory from any carbon source [46]. 2D graphene-based materials obtained in this way are turbostratic, i.e., possess randomly oriented layering [35,46], rendering easy its exfoliation to monolayer graphene [46]. Briefly, the carbon source is graphitized to form flash graphene upon exposure to high-power FJH, promoted by the discharge of an electrical current that increases its temperature up to ca. 2750 °C in less than 100 ms, followed by cooling to room temperature in a few seconds [46]. Non-carbon – volatile, elements are rapidly outgassed from the carbon precursor during flashing [46].

Due to the high carbon content, plastic wastes can be used as feedstock for the production of 2D graphene-based materials through different methods, thus enabling innovative, promising and more environmentally sustainable routes for the production of these valuable materials [40]. The advancements made on this topic will be reviewed in the following sections, sorted by the type of 2D graphene-based material obtained. For the sake of clarity, the nomenclature recommended by the editorial team of the Carbon journal [35] will be followed instead of the nomenclature used in each research article under analysis. For that purpose, each specific material will be referred in a descriptive and scientifically accurate manner [35]. Monolayer graphene is a single-atom-thick sheet of hexagonally arranged sp^2-bonded carbon atoms of extended lateral dimension (ideally infinite in-plane) containing two atoms per unit cell (A and B, cf. Fig. 5a) [35,47]. Bilayer graphene is obtained when two graphene sheets are stacked (cf. Fig. 5b and c),
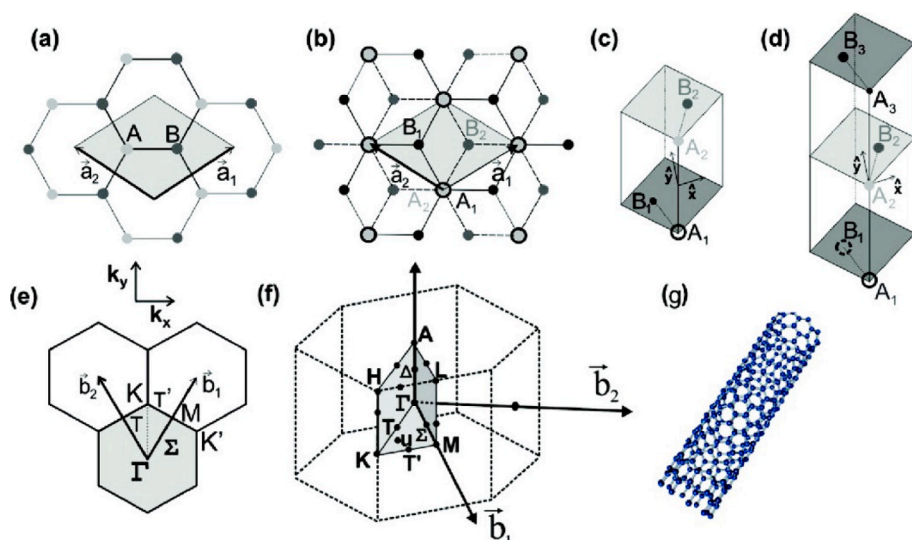
Fig. 5. Schematic representation of the structure of sp^2 nanocarbons. (a) Monolayer graphene. The two vectors $\overrightarrow{a_1}$ and $\overrightarrow{a_2}$ define the unit cell (gray rhombus) containing two atoms A and B. (b) Bilayer graphene. The unit vector, unit cell, and the four atoms (A_1 and B_1 from one layer, and A_2 and B_2 from the other) within the unit cell are displayed. (c) A 3D view of (b). (d) A 3D view of the unit cell for three-layer graphene. (e and f) The Brillouin zone for 2D graphene and 3D graphite, respectively, showing the high symmetry points and lines, and the reciprocal space wavevectors $\overrightarrow{b_1}$ and $\overrightarrow{b_2}$. (g) A single-walled carbon nanotube. Reprinted from [47]. Copyright 2010 American Chemical Society.

whereas three sheets give three-layer graphene (*cf.* Fig. 5d), and so on [47]. The staking order during the formation of multilayer 2D graphene-based materials is important. On this regard, the AB Bernal stacking (*cf.* Fig. 5b–d) leads to the lowest energy geometry (*i.e.*, ground state) and an equilibrium c-axis lattice constant (*i.e.*, interlayer spacing) of 0.335 nm [47]. When layering orientation is random (*i.e.*, without AB stacking), the resulting material (known as turbostratic graphene) will be comprised of graphene sheets with rotational mismatch between neighbouring layers [46]. Raman spectroscopy is the preferred technique to characterize the electronic structure of 2D graphene-based materials, allowing unambiguous, high-throughput, and non-destructive identification of graphene layers [48]. Moreover, Raman spectroscopy can also give important information on structural defects and other crystal disorder, among other properties such as layering orientation [37,47]. Additional information on Raman spectroscopy of 2D graphene-based materials can be found elsewhere [37,47,48]. Monolayer graphene is also the building block of all the other carbon materials [35,47]. For instance, it can be wrapped into (0D) fullerene, rolled up to become (1D) single-walled CNTs (*cf.* Fig. 5g) and stacked to form (3D) graphite (*cf.* Fig. 5f) [38,47]. The amount of stacked graphene layers, namely monolayer (1 layer), few-layer (2 – 5 layers) and multilayer (up to around 10 layers) graphene will be considered in this study; as well as its form (*e.g.*, film, foil or flake) and layering orientation (*i.e.*, with defined registry of the layers or turbostratic) [35].

3.2.1. Monolayer graphene

Monolayer graphene is challenging to obtain. Nevertheless, according to our results, monolayer graphene was indeed the first 2D graphene-based material reported in the literature as being produced from plastics. In this study, conducted by Ruan et al., in 2011, high-quality monolayer graphene was obtained using several low-value solid materials as carbon source, including pristine PS [49]. Briefly, PS without preliminary purification was placed on a Cu foil within a quartz boat and thermally annealed at 1050 °C in a horizontal tubular furnace under low pressure (1.24 kPa) in Ar/H$_2$ atmosphere, monolayer graphene being formed on the backside of the foil (*cf.* Fig. 6). After cooling, a spin-coated poly(methyl methacrylate) (PMMA) thin film was deposited on the backside of the foil, while non-volatile residues left on the top side were etched with an acidic solution. More details on the synthesis conditions are given in Table 1. The PMMA-coated monolayer graphene was then transferred to Si and quartz substrates, dried, washed with acetone and thoroughly characterized, including Raman spectroscopy, and X-ray photoelectron spectroscopy (XPS) [49]. The resulting Raman spectra revealed an intensity ratio of the D band relative to the G mode (I_D/I_G)
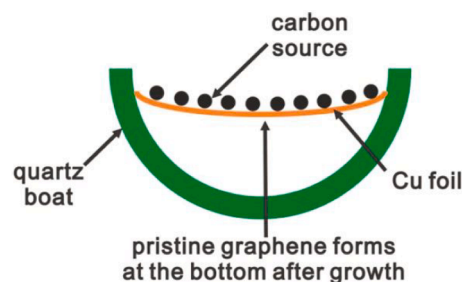
Fig. 6. Arrangement of the Cu foil within the quartz boat. The size of the Cu foil was $\sim 2 \times 3$ cm and the boat was 40 cm long and cut from a quartz tube with a 15 mm inside diameter. Reprinted from [49]. Copyright 2011 American Chemical Society.

below 0.1, indicating the existence of few structural defects; and a sharp 2D peak, corresponding to a ratio of the I_{2D}/I_G above 1.8, indicating its monolayer nature. Usually, $I_{2D}/I_G \sim 2 – 3$ indicates monolayer graphene, $2 > I_{2D}/I_G > 1$ is obtained for bilayer graphene and $I_{2D}/I_G < 1$ suggests multilayer graphene [50]. However, this approach based on the ratio I_{2D}/I_G should only be taken as an estimate. Indeed, the number of staked graphene layers should be determined through deconvolution of the 2D peak, *i.e.*, it should be split into its components [47,48]. This is particularly relevant to confirm the presence of either monolayer or bilayer graphene, which exhibit a single very intense Lorentzian peak or four Lorentzians peaks (two strong and two weak), respectively [47,48]. As the number of layers increases so do the components of the 2D peak, until the signal eventually converges to that of graphite, in which only two peaks are observed [47]. The underlaying physics of these observations are described elsewhere [47,48]. Nevertheless, Ruan et al. confirmed the hexagonal lattice structure of the monolayer graphene synthesized from PS by electron diffraction and the absence of heteroatoms by XPS [49]. Regarding the synthesis mechanism, Ruan et al. claimed that monolayer graphene was grown from the small portion of the carbon source that was able to diffuse to the backside of the Cu foil [49]. Although the authors were unable to demonstrate weather that diffusion was through the Cu foil or by its edges, they showed that only amorphous carbon is formed (on both sides of the Cu foil) when the carbon source is placed 5 cm above the Cu substrate.

A similar conclusion was withdrawn six years later, in 2017, by You et al. [51]. In this case, monolayer graphene was only grown when the plastic waste was placed directly on the substrate. Specifically, the

Table 1
Summary of the studies reporting the conversion of plastics into monolayer graphene.[a]

Authors	Synthesis		Method	Conditions	Yield	Application
	Substrate	Carbon source				
Ruan et al. [49]	Cu foil (annealed at 1050 °C under a Ar flow)	Pristine polystyrene (PS) Petri dishes (untreated)	Thermal decomposition of the carbon source directly on the top of the substrate, monolayer graphene growing on the backside	m_{PS} = 0.01 g; T = 1050 °C; P = 1.24 kPa; Atmosphere: Ar (500 cm³ min⁻¹) and H₂ (100 cm³ min⁻¹); t = 15 min	Not reported	Not reported
Sharma et al. [52]	Cu foil (annealed at 1020 °C under a H₂ flow)	Waste packaging material containing polyethylene (PE; 86 wt.%) and PS (14 wt.%) (cut into strips)	Thermal decomposition of the carbon source, the resulting hydrocarbon gas being continuously fed to a CVD system containing the substrate	Thermal decomposition: $m_{Plastic}$ = 0.003 g; T = 500 °C; R_T = 1.5 °C min⁻¹; t = 90 min; CVD: T = 1020 °C; P = 101.3 kPa; Atmosphere: Ar (98 cm³ min⁻¹) and H₂ (2.5 cm³ min⁻¹); t = 90 min, followed by cooling at 16 °C min⁻¹	Not reported	Not reported
You et al. [51]	Two Ni foils	Waste polyethylene terephthalate (PET) bottles (ground to powder)	Thermal decomposition of the carbon source placed between the substrate foils, monolayer graphene growing on the backside of the bottom foil	m_{PET} = 0.10 g; T = 900 °C; P = 101.3 kPa; Atmosphere: N₂ (1000 cm³ min⁻¹); t = 5 min, followed by cooling at 5.8 °C min⁻¹	Not reported	Not reported

[a] Data collected from the 14 studies reporting the synthesis of 2D graphene-based materials among the 142 studies selected for qualitative analysis based on the criteria detailed in Section 2 and summarized in Fig. 1.

authors developed a synthesis methodology in which waste PET bottles were ground to powder and placed between two Ni foils. Upon thermal annealing at 900 °C under atmospheric pressure in a N₂ flow (*cf.* Table 1), high-quality monolayer graphene (I_{2D}/I_G above 2.6) was formed on the backside of the bottom foil [51]. Moreover, the authors concluded that the purity of the resulting monolayer graphene was ensured by the condensation of gases generated from PET waste on the substrate during the thermal treatment, while leaving behind non-volatile residues on the top of the bottom substrate foil [51]. On the contrary, few- to multi-layer graphene (I_{2D}/I_G of 0.5) flakes with 2 to 3 μm were grown when a Cu-Si substrate (instead of Ni) was employed under similar operating conditions [51].

A different approach was considered by Sharma et al., in 2014 [52]. In this case, monolayer graphene was synthesized using a mixture of waste packaging material containing PE and PS. This waste was used as source of hydrocarbon gas in a furnace at 500 °C. The resulting gas is directed to a CVD system operated under atmospheric pressure with a Cu foil in a Ar/H₂ atmosphere at 1020 °C, as shown in Fig. 7 and detailed in Table 1. The resulting monolayer graphene was coated with a PMMA film, transferred to a SiO₂/Si substrate and characterized. As observed in Fig. 8a, large individual crystals with hexagonal shape were obtained. Raman spectroscopy allowed the authors to conclude about the low amount of structural defects (negligible D peak) and monolayer nature of the resulting material (high I_{2D} when compared to I_G, as depicted in Fig. 8b). Moreover, atomic force microscopy (AFM) allowed estimating the thickness of the graphene crystals (less than 0.5 nm; *cf.* Fig. 8c). Usually, the thickness of monolayer graphene is 0.335 nm [53]. Sharma et al. highlighted the heating rate (R_T) upon which the pyrolysis of plastic waste is conducted as a crucial parameter to obtain large crystals of monolayer graphene, as it influences the injection rate of decomposed polymeric components to the CVD system. Specifically, the larger crystals of monolayer graphene were obtained when R_T was increased up to 1.5 °C min⁻¹; whereas few-layer graphene were obtained for $R_T > 1.5$ °C min⁻¹ [52].

It is noteworthy that Ruan et al., You et al. and Sharma *et al.* have not reported the synthesis yield, nor the applicability of the obtained monolayer graphene materials (*cf.* Table 1).

3.2.2. Few-layer graphene

As mentioned above (Section 3.2.1), Sharma et al. prepared monolayer graphene but were also able to prepare few-layer graphene from a mixture of waste packaging material containing PE and PS [52]. This was accomplished by employing the synthesis procedure described in Table 1, but considering $R_T > 1.5$ °C min⁻¹ [52]. Therefore, in order to avoid duplication, the detailed synthesis conditions are not included in Table 2, which summarizes the studies reporting the conversion of plastics into few-layer graphene.

Another approach to obtain few-layer graphene was recently reported by Gu et al. [54]. In this case, waste PE bags were first ball milled to microscale particles and then carbonized at 1300 °C. Afterwards, the obtained block was ball milled and subjected to microwave sintering, high-quality (I_D/I_G = 0.06) few-layer graphene (I_{2D}/I_G = 1.3) being obtained [54]. More details on the synthesis conditions are given in Table 2. As observed by scanning (SEM) and transmission electron microscopy (TEM), the resulting material possesses wrinkled graphene sheets. Moreover, high-resolution TEM (HRTEM) allowed observing a 2D monolayer of hexagonally arranged sp^2-bonded carbon atoms, as well as confirming the few-layer (bilayer) configuration of this material. Although no application was reported in this study (*cf.* Table 2), the authors concluded that the easiness of preparation and the high quality of the few-layer graphene obtained in this way renders this material as potentially useful for electrochemical and energy applications [54].

3.2.3. Multi-layer graphene

Conversion of plastic wastes into multi-layer graphene has also been reported in the literature. In 2014, Gong et al. described the synthesis of
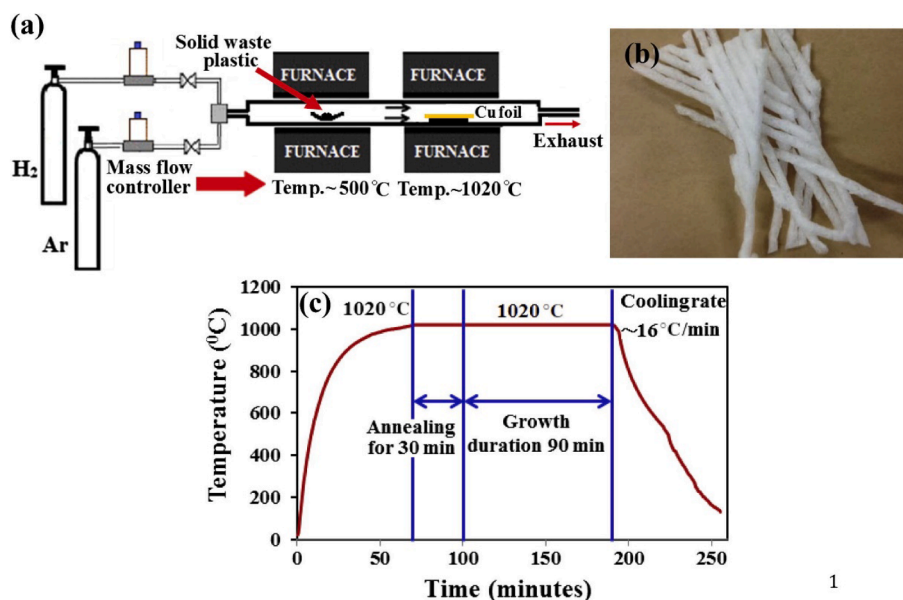
Fig. 7. (a) Representation of the chemical vapour deposition (CVD) process used for the synthesis of monolayer graphene from solid waste plastic. (b) Photograph of waste plastic used in these experiments. (c) Heating, annealing, growth duration and cooling rate of the synthesis process. Reproduced from [52], Copyright 2014, with permission from Elsevier.

multi-layer graphene flakes from thermal annealing of a mixture containing PP waste and organically modified montmorillonite (*cf.* Table 3) [55]. Briefly, the mixture was placed in a crucible and annealed at 700 °C, followed by a two-step purification with hydrofluoric and nitric acids. According to our search results, the study conducted by Gong et al. was the first one reporting the conversion efficiency of plastics into 2D graphene-based materials. Specifically, the authors studied the influence of the ratio between organically modified montmorillonite and

plastic waste. For ratios in the range 0.5 – 8, the yield of multi-layer graphene flakes increased gradually with the mass fraction of organically modified montmorillonite, values as high as 83.8% being obtained [55]. The resulting materials consisted of 12 – 20 stacked graphene layers with discontinuous and curved graphitic structure (*cf.* Fig. S1) containing significant structural defects (I_D/I_G ratios in the range 1.4 – 2.3) [55]. Nevertheless, the amount of both stacked graphene layers and structural defects decreased as the ratio between organically modified
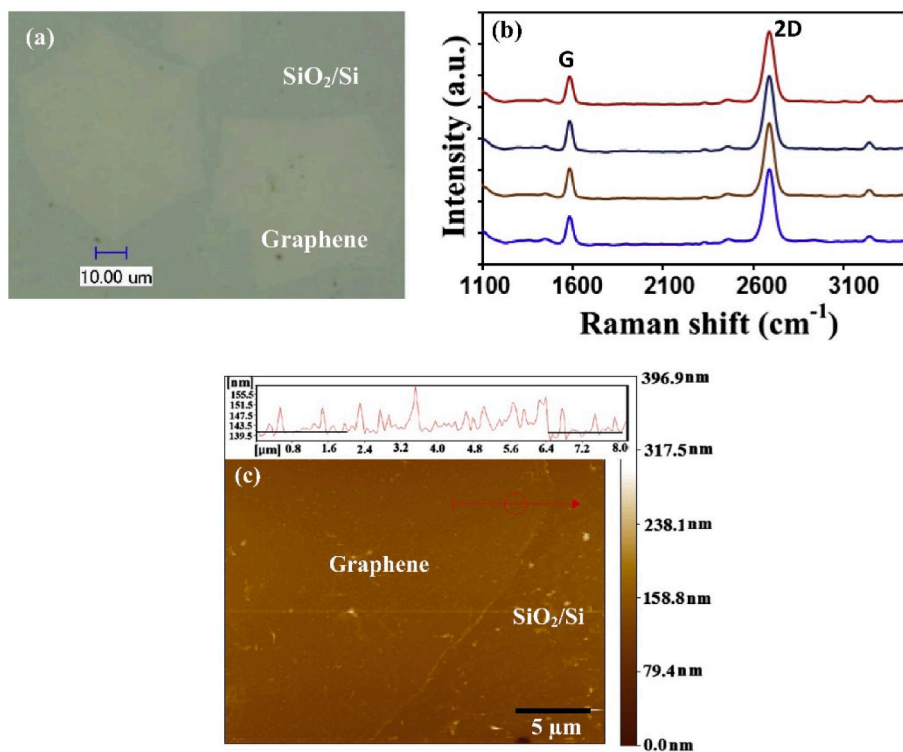
Fig. 8. (a) Optical microscope image of hexagonal graphene crystal after transferring to SiO$_2$/Si substrate. (b) Raman spectra of the transferred graphene crystal taken randomly at four different points. (c) Atomic force microscopy (AFM) image at an edge of the graphene crystal after transferring to SiO$_2$/Si substrate. Most of the edges of graphene crystal are folded; a thickness less than 0.5 nm is estimated. Reproduced from [52], Copyright 2014, with permission from Elsevier.

Table 2
Summary of the study reporting the conversion of plastics into few-layer grapheme.[a]

| Authors | Synthesis | | | | | Application |
	Substrate	Carbon source	Method	Conditions	Yield	
Gu et al. [54]	Not employed	Waste polyethylene (PE) bags (cut and ball milled)	Thermal decomposition of the carbon source in a vacuum oven, followed by ball milling and microwave sintering	Thermal decomposition: $m_{Plastic}$ = 2 g T = 1300 °C; P: Not reported t = 120 min Microwave sintering: Power: Not reported t = 6 min	Not reported	Not reported

[a] Data collected from the 14 studies reporting the synthesis of 2D graphene-based materials among the 142 studies selected for qualitative analysis based on the criteria detailed in Section 2 and summarized in Fig. 1.

montmorillonite and plastic waste increases [55]. The resulting materials consisted of over 12 stacked graphene layers with discontinuous and curved structure [55]. Organically modified montmorillonite participates as catalyst for the degradation of PP waste, leading to the formation of both gaseous (*e.g.*, hydrogen, methane, ethane, ethylene, propane, propylene and isobutene) and liquid (*e.g.*, monoaromatics such as benzene, diaromatics such as naphthalene, and polycyclic aromatic hydrocarbons such as phenanthrene) products, which were considered the effective carbon sources for the growth of multi-layer graphene flakes [55].

In 2017, Cui et al. employed 6 different plastic wastes as carbon sources for the preparation of freestanding graphene foil with microscale thickness [56], by employing a synthesis methodology similar to that previously reported by Sharma et al. [52]. Briefly, PMMA, PS, PP, PVC, PE and PET wastes were employed separately as source of

hydrocarbon gas to be added to a CVD system containing a Ni foil as substrate. After cooling, an etching solution of $FeCl_3$/HCl was used to remove Ni. More details on the synthesis conditions are given in Table 3. Despite the microscale thickness (in the range 1.7–2.8 μm), the resulting foil should be described as multi-layer graphene, since it consists of a defined number of countable graphene layers of extended lateral dimension [35]. Raman spectroscopy allowed concluding that, except for the graphene foil obtained from PMMA, which revealed a I_D/I_G ratio of 0.65, all the other materials possess negligible structural defects (I_D/I_G ratios in the range 0.03–0.11) [56]. Moreover, the authors concluded that both the compact interlayer stacking and highly ordered structure contributed to the very high electrical conductivity (up to 3824 S cm^{-1}) of the graphene foils produced from the different plastic wastes [56]. Taking this into account, the graphene foils were applied in the electrochemical and energy field, namely as anode materials for lithium-ion

Table 3
Summary of the studies reporting the conversion of plastics into multi-layer grapheme.[a]

| Authors | Synthesis | | | | | Application |
	Substrate	Carbon source	Method	Conditions	Yield	
Gong et al. [55]	Organically modified montmorillonite	Waste bumper composed mainly by polypropylene (PP; *ca.* 89 wt.%) and additives (*ca.* 11 wt.%) such as talcum (ground to 2 mm granules)	Thermal decomposition of the carbon source mixed with the substrate at 700 °C	$m_{Plastic}$ = 1.7 g T = 750 °C; P: Not reported Atmosphere: Not reported t = 15 min (cooling rate not reported)	83.8%	Not reported
Cui et al. [56]	Ni foil (annealed at 1050 °C under a Ar/H_2 flow)	Waste PP, polyethylene terephthalate (PET), polyethylene (PE), polystyrene (PS), polyvinylchloride (PVC) and poly(methyl methacrylate) (PMMA), used apart (cut into pieces)	Thermal decomposition of the carbon source, the resulting hydrocarbon gas being continuously fed to a CVD system containing the substrate	Thermal decomposition: $m_{Plastic}$: Not reported T: Not reported; R_T: Not reported t = 120 min CVD: T = 1050 °C; P: Not reported Atmosphere: Ar (150 cm^3 min^{-1}) and H_2 (25 cm^3 min^{-1}) t = 120 min (cooling rate not reported)	10 – 43%	Electrochemical cells
Nguyen et al. [61]	Ni mesh (treated with HNO_3)	Waste Parafilm® M, composed mainly by paraffin and PE (washed with water)	Thermal decomposition of the carbon source directly on the substrate, multi-layer graphene growing over the entire substrate	$m_{Parafilm® M}$: Not reported T = 900 °C; P = 0.13 x 10^{-3} kPa Atmosphere: Autogenous t = 8 min, followed by cooling at *ca.* 90 °C min^{-1}	Not reported	Supercapacitors and water treatment by adsorption
Mat Tahir et al. [57–60]	Cu foil (annealed at 1020 °C under a H_2 flow)	Waste packaging material of unknown composition (cut into strips)	Thermal decomposition of the carbon source, the resulting hydrocarbon gas being continuously fed to a CVD system containing the substrate	Thermal decomposition: $m_{Plastic}$: Not reported T: Not reported; R_T: Not reported t = 90 min CVD: T = 1020 °C; P = 101.3 kPa Atmosphere: Ar (100 – 150 cm^3 min^{-1}) and H_2 (0 – 50 cm^3 min^{-1}) t = 90 min, followed by cooling at *ca.* 2.8 °C min^{-1}	Not reported	Not reported

[a] Data collected from the 14 studies reporting the synthesis of 2D graphene-based materials among the 142 studies selected for qualitative analysis based on the criteria detailed in Section 2 and summarized in Fig. 1.

batteries. To the best of our knowledge, this was the first study reporting the application of 2D graphene-based materials produced from plastics.

Cui et al. [56] also reported the conversion efficiency of plastics into 2D graphene-based materials. As shown in Table 3, yields in the range 10 – 43% were obtained depending on the plastic waste used as carbon source. The highest yields were obtained with PE, PP and PS, which are the polymers with higher thermal resistance. Therefore, the authors concluded that the yield increases with the decomposition temperature of the plastic wastes, since a higher thermal stability of this carbon source leads to a lower injection rate of decomposed polymeric components to the CVD system [56]. This conclusion is in agreement with that previously withdrawn by Sharma et al. [52] on the importance of maintaining a low injection rate of hydrocarbon gases to the CVD system (*cf.* Section 3.2.1).

Mat Tahir et al. performed a series of 4 studies reporting the synthesis of 2D graphene-based materials prepared from packaging waste resulting from fruit cover plastics [57–60]. Although some operating conditions used in those studies are not given (*cf.* Table 3), the synthesis methodology was similar to that previously reported by Sharma et al. [52], as discussed above (*cf.* Fig. 7), some variations in the synthesis conditions being considered, such as the flow of Ar and H_2 in the CVD system (*cf.* Table 3). In the first of this set of studies, Mat Tahir et al. reported that few-layer graphene was obtained [57]; however, this conclusion was only supported by the I_{2D}/I_G ratio (0.56) determined by Raman spectroscopy. Although the number of layers of the materials obtained in the other studies were not discussed by the authors, the broad 2D peaks (*i.e.*, divisible in several components) observed in the Raman spectra (made available in [58;60]) suggest the presence of multi-layer graphene and/or graphite [48].

Multi-layer graphene can also be obtained by thermal decomposition of plastic wastes directly over a substrate. In 2017, Nguyen et al. reported the use of waste Parafilm® M as carbon source for the synthesis of freestanding, flexible and transparent multi-layer graphene films with nanoscale thickness, via a single-step thermal annealing process under vacuum [61]. Briefly, waste Parafilm® M was washed with water, attached to a Ni mesh, placed in a quartz chamber and thermally annealed under vacuum at different temperatures in the range of 600 – 980 °C. More details on the synthesis conditions are given in Table 3. After cooling, the resulting materials were coated with a PMMA film. An etching solution of $FeCl_3$/HCl was then used to remove Ni from the PMMA-coated materials, followed by a thermal treatment at 450 °C under vacuum to remove PMMA, thereby obtaining freestanding carbon-based materials [61]. The authors concluded that the temperature of thermal annealing is the operating parameter dictating the type/quality of carbon material obtained from the conversion of Parafilm® M [61]. The Raman spectra revealed that waste Parafilm® M was converted regardless of the temperature employed (*cf.* Fig. S2a). Nevertheless, the two broad and low-intensity peaks observed in the range 1200 – 1700 cm^{-1} indicated that only amorphous carbon was obtained when the carbon source was annealed at 600 °C; whereas samples annealed at higher temperatures (700–980 °C) revealed the Raman fingerprints for graphene/graphite, namely the peaks D, G and 2D. I_D/I_G ratios of 0.54, 0.41, 0.28 and 0.14 were observed with the materials obtained at 700, 800, 900 and 980 °C, respectively, indicating that the amount of structural defects decreases as the annealing temperature increases. However, the Raman spectrum of the sample obtained at 980 °C revealed a change in the shape, and a shift of *ca.* 10 cm^{-1}, of the 2D peak, accompanied by a marked decrease of the I_{2D}/I_G ratio, indicating the formation of a thin graphite film (*cf.* Fig. S2a). Bearing this in mind, the authors selected 900 °C as the optimum temperature for the synthesis process to be carried out [61]. Therefore, the sample obtained at that temperature was thoroughly characterized, including morphological characterization. Accordingly, SEM micrographs were taken before (Fig. S2b) and after (Fig. S2c) removing the Ni substrate, revealing the typical morphology of graphene as well as its 2D nature (thickness in the range 2.5–3.5 nm, as determined by AFM). Moreover,

the multi-layer nature (6–8 layers) was confirmed by HRTEM (inset of Fig. S2d). The materials prepared by Nguyen et al. were applied in the electrochemical and energy (supercapacitors), and environmental (water treatment by adsorption) fields [61].

3.2.4. Flash graphene

Flash graphene can also be prepared from plastic wastes. According to our results, the first example of such approach was reported in 2020 only, by Luong et al. [37]. In that study, flash graphene was obtained using several low-value solid materials as carbon source, including waste and pristine plastics employed separately (*cf.* Table 4). Briefly, 5 wt% of carbon black was added to the carbon source to increase its conductivity, the resulting mixture being placed in a flashing chamber designed for that purpose (*cf.* Fig. 9). Graphitization to form flash graphene was then accomplished in the absence of a catalyst, upon exposure to high-power FJH promoted by a direct current (DC) pulse discharge. The non-carbon atoms sublime out as small molecules during this ultra-fast procedure (the temperature increases up to *ca.* 2750 °C in less than 100 ms), leading to a product with very high carbon content [37]. More details on the synthesis conditions are given in Table 4. The turbostratic stacking of flash graphene was confirmed by Raman spectroscopy [37]. The random layering orientation (*i.e.*, turbostratic nature) is the main difference between flash graphene and the other forms of graphene discussed in this review, which display a defined registry of the layers with constant interlayer spacing of 0.335 nm [35,47].

The flash graphene yield obtained when using plastics as carbon source was not reported by Luong et al., but it can be as high as 80–90% when carbon black, calcine coke and anthracite coal are used as carbon source, corresponding to an electric input of *ca.* 7.2 kJ g^{-1} [37]. In this study, the synthesis processes employing calcine coke, anthracite coal and plastics were not optimized. Optimization was performed only when employing carbon black as feedstock for the production of flash graphene. In this case, flash graphene samples with ultra-high purity and low defects were obtained, as demonstrated by the exceptional Raman signatures observed (negligible D peaks and I_{2D}/I_G ratios up to 17) [37]. Moreover, the authors demonstrated the (possible) scalability of the FJH synthesis methodology. Through increasing the diameter of the quartz tube used as flashing chamber from 8 to 15 mm, the system enabled the synthesis of 1 g of flash graphene per batch [37].

Immediately after the publication made by Luong et al. [37], the same research group reported 2 additional studies focusing on the conversion of plastic wastes into flash graphene [62,63]. The first of those studies, conducted by Algozeeb et al., was focused on optimizing the quality and yield of the flash graphene obtained from several plastic wastes separately and in mixture [62]. The main experimental novelty introduced by Algozeeb et al. was the application of an alternating current (AC) pulse discharge (*cf.* Table 4). On this regard, the effect of particle size (in the range 0.04 – 2 mm), resistivity (110–163 Ω) and thermal stability of the plastic wastes on the yield of flash graphene was studied first. It was found that the yield increases as the particle size and thermal stability increases, and the resistivity decreases [62]. Nevertheless, the most significant finding made by Algozeeb et al. was that the sequential application of AC and DC-FJH allows obtaining higher quality flash graphene regardless of the plastic waste used as carbon source. Indeed, the Raman signatures of the resulting materials (I_D/I_G ratios in the range 0.03–0.14; and I_{2D}/I_G ratios up to 6) were far superior to those obtained when AC-FJH was employed independently [62]. This beneficial effect was ascribed to the additional lateral growth promoted by the DC-FJH step, which leads to an increase in the size of the graphene sheets [62]. For instance, the average sheet size of flash graphene obtained from high-density polyethylene (HDPE) increased from 16 to 27 nm upon DC-FJH (as determined by TEM). These results agree with the decrease observed in the D band of the Raman spectra (usually correlated to the surface to edges density) of the resulting materials, corresponding to a decrease of the I_D/I_G ratio from 0.77 to 0.09 [62].

Algozeeb *et al.* have also conducted a detailed study on the energy

Table 4

Summary of the studies reporting the conversion of plastics into flash graphene.[a]

Authors	Synthesis					Application
	Substrate	Carbon source	Method	Conditions	Yield	
Luong et al. [37]	Not employed	Waste polypropylene (PP), polyethylene terephthalate (PET), polyethylene (PE) and polyvinylchloride (PVC), and pristine polyacrylonitrile (PAN), used apart and mixed (sanded/cut to powder and mixed with 5 wt.% of carbon black)	The carbon source is compressed between two electrodes in a quartz tube placed inside a vacuum desiccator, through copper-wool plugs or graphite spacers to allow degassing of the volatiles. Flash Joule heating (FJH) is performed upon a direct current (DC) pulse discharge made by a capacitor bank	$m_{Plastic} = 0.10$ g; P: Not reported Compression regulated to minimize sample resistivity (1 – 1,000 Ω) Capacitor bank: 20 capacitors with a total capacitance of 0.22 F $E = 400$ V; $I = 1,000$ A $t = 100$ ms (cooling rate >7000 °C s^{-1})	Not reported	Not reported
Algozeeb et al. [62]	Not employed	Waste polypropylene (PP), polyethylene terephthalate (PET), polyethylene (PE), polystyrene (PS) and polyvinylchloride (PVC), used apart and mixed (sanded/cut to powder and mixed with 5 wt.% of carbon black)	The carbon source is compressed between two electrodes in a quartz tube placed inside a vacuum desiccator, through copper-wool plugs or graphite spacers to allow degassing of volatiles. FJH is performed upon sequential alternating current (AC) and DC pulse discharges	$m_{Plastic} = 0.50$ g; $P = 1.3$ kPa Compression regulated to obtain sample resistivity of 110 – 163 Ω AC: $E = 120$ V; $f = 60$ Hz $t = 8$ s (cooling rate: Not reported) DC: Capacitor bank: 10 capacitors with a total capacitance of 0.06 F $E = 120$ V; $I = 180$ A $t = 100$ ms (cooling rate: Not reported)	18%	Not reported
Wyss et al. [63]	Not employed	Ashes resulting from the industrial pyrolysis of waste PP at 450 °C (ground to powder)	The carbon source is compressed between two electrodes in a quartz tube placed inside a vacuum desiccator, through copper-wool plugs or graphite spacers to allow degassing of volatiles. FJH is performed upon a DC pulse discharge made by a capacitor bank	$m_{Pyrolysis\ ashes} = 0.40$ g (obtained by pyrolysis of 2 g of PP); P: Not reported Compression regulated to obtain sample resistivity of 15 Ω Total capacitance = 0.06 F $E = 160$ V; I: Not reported $t = 450$ ms (cooling rate: Not reported)	17%[b]	Composite filling

[a] Data collected from the 14 studies reporting the synthesis of 2D graphene-based materials among the 142 studies selected for qualitative analysis based on the criteria detailed in Section 2 and summarized in Fig. 1;

[b] Value determined considering the yields reported by Wyss *et al.* on the conversion of plastic wastes into pyrolysis ashes (20%) and on the conversion of pyrolysis ashes into flash graphene (85%).

input required to obtain flash graphene from mixed plastic wastes (40% HDPE, 20% PP, 20% PET, 10% low-density PE, 8% PS and 2% PVC). In total, the combined AC-DC-FJH synthesis procedure requires 23 kJ g^{-1} of plastic waste [62], which, at current Portuguese household electricity prices (0.1441 € kWh^{-1}), corresponds to *ca.* 920 € Mg^{-1} of flash graphene produced. This value is very competitive when compared to the average prices for the virgin (in the range 580–1110 € Mg^{-1}) and recycled (580–1810 € Mg^{-1}) plastics involved in the study of Algozeeb et al. [62].

In the third study on this topic made by the research group of Luong et al. [37] and Algozeeb et al. [62], Wyss et al. showed that flash graphene can also be obtained from FJH of the ashes resulting from the pyrolysis of plastic wastes [63]. However, even under the optimum conditions, the quality of resulting materials (I_D/I_G ratio of 0.37; and I_{2D}/I_G ratio of 0.83 [63]) is still worse than that previously reported by Algozeeb et al. [62].

4. Final remarks and conclusions

The 142 studies discussed in Section 3.1 demonstrate unequivocally the current research trend towards the conversion of plastic wastes into valuable carbon materials. In addition to the classical carbonization of plastic wastes into ACs, the interest regarding the synthesis of valuable CNTs and 2D graphene-based materials has been growing in recent years. Nearly half of the studies analysed in this systematic review addresses the synthesis of CNTs - a trend confirmed by the amount of review articles available in the literature (10 out of 12, according to our findings). Nevertheless, the amount and quality of the publications on

the synthesis of 2D graphene-based materials have been growing since the first publication on the topic, in 2011. These studies were herein reviewed for the first time.

Overall, we have found it very challenging to select which studies, among those reporting the synthesis of 2D graphene-based materials from plastic wastes, should be included in the discussion undertaken in Section 3.2. This difficulty arose mainly from the misclassification of the obtained materials as "graphene". Accordingly, some studies claiming the synthesis of "graphene" were not discussed in Section 3.2, since a detailed analysis of the reported results allowed concluding that those materials were actually mixtures containing large amounts of amorphous carbon and/or graphite, and/or metal composites containing small fragments of graphene sheets [64–68], rather than well-defined, countable, stacked graphene layers of extended lateral dimension [35]. Instead, these materials were classified as "other" type of carbon materials (*cf.* Fig. 2b and c). For instance, in some cases is claimed that few-layer graphene is obtained, but the average number of layers is not determined experimentally (*e.g.*, through deconvolution of the 2D peak obtained by Raman spectroscopy and/or HRTEM measurements), this conclusion being solely based on the broad 2D peak observed in the Raman spectrum of the resulting materials. In any case, a broad 2D peak in the Raman spectrum may suggest the presence of multi-layer graphene and/or graphite, rather than few-layer graphene [48]. Taking this into account, we suggest all authors to always characterize the obtained materials properly (including at least Raman spectroscopy and HRTEM observations) and refer to those materials in a scientifically accurate manner - preferably following the nomenclature recommended by the editorial team of the Carbon journal [35]. Accordingly, the authors are
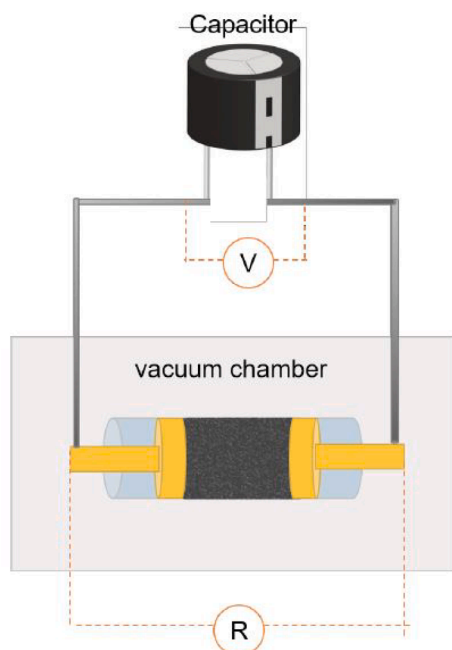
Fig. 9. Simplified scheme of the flash Joule heating (FJH) system. Reprinted with permission from [62], Copyright 2020 American Chemical Society. Detailed description of the circuit can be found in [37].

urged to take full advantage of Raman spectroscopy. Specifically, the dependence between the 2D peak and the number of stacked graphene layers [47,48] was not explored in any of the 11 studies reporting the conversion of waste plastics into monolayer, few-layer, or multi-layer graphene. On the contrary, the potential of Raman spectroscopy was fully used to confirm the presence of turbostratic graphene in the 3 studies reporting the synthesis of flash graphene from plastic wastes [37,62,63].

Regarding the results itself, the studies under revision report the synthesis of monolayer, few-layer and multi-layer graphene materials (including flash graphene) through 4 main synthesis methodologies:

1) Thermal decomposition of the plastics directly over a metal substrate;
2) Prior thermal decomposition of the plastics, with the resulting hydrocarbon gases released being fed to a CVD system containing the metal substrate;
3) Thermal decomposition followed by ball milling and microwave sintering;
4) Flash Joule heating (FJH).

Most of these studies (11 out of 14) are solely focused on optimizing the synthesis conditions. Nevertheless, the remaining studies also feature important contributions on this regard. As consequence, high-quality 2D graphene-based materials have been obtained through all the reported synthesis methodologies-regardless of their own specifications (*cf.* Table 5). However, both the yield and the cost associated to

Table 5

Summary of the 4 main synthesis methodologies used for conversion of plastic wastes into 2D graphene-based materials.

Synthesis methodology	Plastics converted	2D graphene-based materials obtained	Key advantages	Main constraints/ challenges
(i) Thermal decomposition of the plastics directly over a metal substrate	PP, PET, PE, PS, paraffin	Monolayer [49,51] Multi-layer [55,61]	Single-step process High yields (up to 83.4%) Relatively high material output (up to 1.42 g per synthesis batch)	Relatively lower quality product (I_D/I_G ratio in the range of 0.1–2.3) Very sensitive to the temperature employed (*e.g.*, amorphous carbon or graphite are formed when the temperature is below or above the optimum value, respectively) Large-scale production (reported plastic feedstock in the range of 0.01–1.7 g, corresponding to a maximum output of 1.42 g per synthesis batch) Purification of the final product is needed (remove metal substrate and non-volatile residues)
(ii) Prior thermal decomposition of the plastics, with the resulting hydrocarbon gases released being fed to a CVD system containing the metal substrate	PP, PET, PE, PS, PVC, PMMA	Monolayer [52] Multi-layer [56–60]	CVD is a well-established synthesis methodology (upon exposure of metal substrates to pure hydrocarbon gases) High-quality product (I_D/I_G below 0.65 in all the reported studies) The properties of the resulting materials (*e.g.*, crystal size and number of graphene layers) can be tailored by adjusting the injection rate of decomposed polymeric components to the CVD system	Two-step process Low yields (10–43%) Large-scale production (only one study reports the plastic feedstock: 0.003 g) Purification of the final product is needed (remove metal substrate)
(iii) Thermal decomposition followed by ball milling and microwave sintering	PE	Few-layer [54]	No metal substrate is needed High-quality product (I_D/I_G of 0.06 in the only reported study) The three steps involved in this synthesis methodology are well-established for the preparation of other carbon materials	Three-step process High temperature needed (up to 1300 °C) Unknown yield Large-scale production (the plastic feedstock in the only reported study is 2 g) Purification of the final product is needed (remove non-volatile residues)
(iv) Flash Joule heating (FJH)	PP, PET, PE, PS, PVC, PAN	Flash [37,62,63]	Single-step process No metal substrate is needed No purification is needed High-quality product (I_D/I_G in the range of 0.03–0.37) Competitive estimated costs (920 € Mg^{-1}) No furnace, solvents or reactive gases are needed	The conductivity of the plastic feedstock must be increased, either through an additive (*e.g.*, carbon black) or a preliminary thermal treatment Low yields (17–18%) Large-scale production (reported plastic feedstock in the range of 0.1–2.0 g, corresponding to a maximum output of 0.34 g per synthesis batch)

the synthesis of these materials have not been considered in most studies on this topic. Indeed, cost estimations were only performed for flash graphene obtained by FJH (*cf.* Table 5).

The key features and operating parameters affecting the quality of the materials obtained through the synthesis approaches, that are already reported for the conversion of plastic wastes into 2D graphene-based materials, are summarized in Table 5. Overall, the (i) thermal decomposition of the plastics directly over a metal substrate enables the highest yield of 2D-graphene-based materials; however, these materials are more defective than those obtained when the other synthesis methodologies are employed. Regarding the two-step approach involving (ii) prior thermal decomposition of the plastics, with the resulting hydrocarbon gases released being fed to a CVD system containing the metal substrate, has as advantage the replacement of the pure hydrocarbon gas typically feed to CVD processes by the decomposed polymeric components; therefore, the benefits and constraints of this approach are like those of the classic CVD processes. In fact, the 2D graphene-based materials with highest quality have been obtained through the synthesis methodologies carried out in the absence of a metal substrate, namely (iii) thermal decomposition followed by ball milling and microwave sintering, and (iv) FJH. These are also the methodologies in which the plastic wastes are subjected to higher temperatures, namely (iii) in vacuum (1300 °C) and microwave (unknown temperature) ovens, or (iv) as result of an electrical discharge (*ca.* 2750 °C), thereby suggesting that the operating temperature is the crucial parameter to obtain high-quality materials. Moreover, flash-graphene obtained by FJH does not need to undergo a purification step. This is an additional advantage resulting from the combination of the extreme temperature reached in the flash process, which outgasses non-carbon elements from the carbon precursor, and where a metal substrate is not required. On the contrary, the materials obtained through the other synthesis methodologies must be subjected (at least) to a final purification step to remove the metal substrate and/or non-volatile residues (*cf.* Table 5). At the laboratory scale, this has been accomplished using careful washing and/or rinsing procedures employing acidic solutions (to remove the metal substrate and, in some cases, amorphous carbon) [49,52,55,56,61] and organic solvents, mainly acetone (to remove non-volatile residues) [49,52,61]. Therefore, the industrial viability of such purification processes should be studied, and improvements made as needed. Moreover, the possible environmental impacts of the resulting aqueous effluents should also be considered. This is particularly relevant since using plastic wastes as feedstock to produce 2D graphene-based materials aims to increase the attractiveness of plastics recycling and thus mitigate environmental impacts, and thus life cycle assessment (LCA) could be an important tool in this domain. In addition, it is noteworthy that some studies do not provide details on the procedure (and solutions) used in the purification step(s), a situation that should be addressed more carefully in future studies.

As recognized by Algozeeb et al., the effective environmental impact assessment of the synthesis approaches used for the conversion of plastic wastes into 2D graphene-based materials remains to be done [62]. Indeed, LCA is an important tool to help with decision and policy making. It should take into account the environmental impacts arising during the full life cycle of these processes, but also other relevant variables such as related costs. Accordingly, possible negative contributions (*e.g.,* release of greenhouse gases and generation of aqueous effluents due to etching/washing steps) should be minimized, and the associated costs compared to those of the other options (*e.g.,* synthesis of 2D graphene-based materials employing other feedstocks, and other options for the management of plastic wastes). FJH has an apparently important advantage on this regard, as it uses no furnace, metals, harmful purification solutions or reactive gases [37].

In conclusion, despite the significant findings and improvements on the synthesis of 2D graphene-based materials from plastic wastes discussed in this review, much is yet to be done before establishing any of

such processes as part of practical options for the management of plastic wastes. In that sense, the lack of scalability of the available synthesis methodologies can be pointed as the main constraint hindering real-scale applications. As summarized in Table 5, the maximum output per synthesis batch does not exceeds 1.42 g regardless of the approach taken. Nevertheless, some improvements have been made regarding the output [37] of flash graphene by FJH using carbon-rich residues other than plastic wastes, opening future prospects for the possible scalability of this promising technology. Nevertheless, despite enabling a much higher output than that of the CVD-based processes, the amount of flash graphene obtained through this optimized system is still very low (1 g per batch) for industrial-scale applications. However, it is noteworthy that the conversion of plastic wastes into flash graphene occurs in less than 1 s [37], rendering FJH incomparably faster than the other synthesis methodologies. Moreover, the competitive costs already estimated to produce flash graphene from plastic wastes at laboratory scale (920 € Mg^{-1}) render this synthesis methodology economically attractive for scale-up [62]. Accordingly, additional research efforts should focus on the development of reliable and scalable synthesis methodologies, since enabling large-scale production of high-quality 2D graphene-based materials is the only possible route towards increasing the economic - and, possibly, environmental - attractiveness of the conversion of plastic wastes. Nevertheless, efforts should also be made to minimize the environmental impacts arising during the full life cycle of the synthesis processes themselves.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2021.131399.

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