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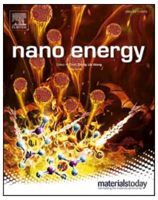

Review

Carbon-enabled microwave chemistry: From interaction mechanisms to nanomaterial manufacturing

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ABSTRACT

The multiple interaction effects between external microwave fields and carbon materials trigger the thermocatalysis and conversion reactions by constructing the uniformly thermal room or localized heat microenvironment in confined space, providing an alternative and promising technology for functional materials engineering and manufacturing. This review summarizes the state-of-the-art microwave chemistry techniques involved in or enabled by the carbon additives, with a focus on the introduction of the underlying mechanism, influence factors/key parameters, and the technologies of advanced material preparation from nano to bulk scales. Specifically, it is reviewed in detail that the carbon materials are used as microwave heating media and hotspots/arc plasmas generators to drive and boost the thermochemical reactions. The carbon materials as precursors in microwave are also summarized, which can be helpful for producing the advanced materials with tailored properties and multi-functionalities that are not far realized by conventional techniques/methods. Moreover, the expectation for the future of carbon-participated microwave processes and some approaches to optimizing the corresponding reactions in terms of the technical and industrial levels are proposed, which will help to jump out of gap from lab to industrial application and finally inspire many more endeavors for the manufacturing of advanced/innovative nanomaterials in future.

1. Introduction

Carbon materials formed by covalently bonded carbon atoms in diverse hybridization states (sp, sp^2, sp^3) deliver a wide range of structures, as well as versatile physicochemical characteristics, which underpin the immense development of material science [1–4]. Throughout the preparation technologies of carbon materials, the commonly used heat treatment, which operates at high temperatures by consuming electric energy, is a crucial thermochemical process for pyrolysis/carbonization, purification, modification, and even transformation [5–8]. Microwave-driven/assisted schemes, by contrast, can provide rapid and affordable approaches to overcoming the high energy-consuming problems in efficiently controlling the synthesis, composition, and morphology of materials, which may have great significant in the high-quality and scale-up production [9–13]. Up to now, microwave chemistry has gained a plenty of popularity for the synthesis of carbon materials with small molecules to long chain structures. For instance, the carbon allotropes, including sp^2-dominated graphene [14–18] and carbon nanotubes (CNTs) [19–22], sp^3-dominated diamond [23,24], sp^2 and sp^3-cohybridized fullerene [25], and graphdiyne with sp^2 and sp-mixed hybridization [26], as well as carbon materials constructed by atoms in complex hybridization states such as carbon fibers [27,28], carbon dots (C-dots) [29–33], porous carbons [34,35], and activated carbons (ACs) [11,36–38] are nowadays effectively prepared by microwave approaches.

Generally, the close conduction and valence bands for most of carbon materials determine that the π-electrons move freely and their movement can be accelerated by the electric field component of microwave [39–41]. The prevailing model of *Joule* heating depends on a classical theory in which the free electrons collide with the carbon atoms to generate heat locally, while this model has been supported by most of experiments and used to explain the heating process involving carbon

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absorbers. In addition, as the best of we know, the carbon materials have a big family and most of them demonstrate the complex surfaces and structures. It is noteworthy that the functional groups and defects on surface, as well as the adjustable porous structures can attenuate the incident microwaves and transform them into heat energy. Further, the state-of-the-art carbon materials with multiple function/effects were also presented by tuning the component, structure and morphology [42,43]. More meaningfully, the energy conversion efficiency from microwave to heat can be further enhanced in these carbon-participated/enabled microwave reactions.

Consequently, the carbon materials, as typical heating media, hotspots/arc plasmas generators and precursors, are capable of being used for the microwave-driven/assisted materials configuration, which are elaborately explored and developed by many previous studies (Fig. 1a). Benefitting from the excellent thermal, mechanical, and electrical properties, these carbon materials deliver the diverse microwave absorption (MA) properties, thus endowing them with the capability to participate the reactions in the forms of heating media and/or hotspots/arc plasmas generators [42,44]. Typically, the ultra-high vaporization temperature and thermal conductivity of carbon heating media guarantee the harsh reaction environment for microwave reactions. For example, the purified carbon nanotubes can be microwave-heated to 500–650 °C in a couple of seconds due to the high dielectric losses [45]. The visualized red hot also proves the elevated temperature of carbon bed, as shown in many of the previous reports [46,47]. After employing carbon absorbers, the hotspots and arc plasmas with the characteristics of localized super-heating would be also produced and play the fantastic effects on processing material in micro/nano-scale, such as surface heating, doping, exfoliation, reduction, and transformation. Another branch for the applications of carbon additives is that carbon can serve as precursors. The self-configuration of carbon materials can be aroused by microwave irradiations, which offers more opportunities in effectively regulating the structures and components of carbon themselves. Of course, the microwave treatment in these cases is the result of coupling the diverse effects of carbon additives, for instance, the carbon species simultaneously function as both heating media, hotspots/arc plasmas generators and precursor. Moreover, there are some carbon precursors that fail to achieve such a self-configuration due to the mismatching between the absorbed microwave and the required energy of structural reconstruction. In order to tackle this problem, additional pre-treatment [14], non-carbon absorbers [48–50], liquid phase [51] as well as microwave plasmas-induced solid-vapor-solid reactions [23,52] have also been applied for preparing carbon materials with novel nanostructures and properties, enabled by modulating the MA property of precursors or constructing high-energy-density environment.

As witnessed by us, the development and application of microwave chemistry and technique has shown its promising potential towards the preparation of advanced carbon-related materials. In return, the carbon-based materials with the different electromagnetic absorption properties bring in both opportunities and challenges in the fields of materials engineering and microwave chemistry. The published articles about reactions driven by microwave, involved in different carbon additives for processing the advanced materials rapidly increase in recent years. The statistical data from Web of Science are shown in Fig. 1b. In this review, the multiple effects of carbon additives in microwave systems were summarized. In parallel, the mechanism and influence factors/key parameters have also been explicitly introduced. Furthermore, driven by microwave field, the corresponding conversion reactions ranging from microscopic to macroscopic level are also summed up. The challenges and perspectives in this field are presented and commented, with an aim to help to broaden the horizon for the design and preparation of advanced materials and serve as a guideline for building the techniques related to microwave chemistry.

2. Fundamental for carbon-enabled microwave chemistry

Microwave is a kind of electromagnetic wave that exhibits a sinusoidal variation of electric and magnetic fields. The frequency of microwave is from 300 MHz to 300 GHz, in which the 2.45 GHz frequency is most often used and has been applied in the commercial microwave ovens. Microwave treatment, as the emerging strategy can dramatically reduce the chemical wastes and shorten reaction time in chemical synthesis and transformations in comparison to the traditional one [53]. In fact, in addition to fast heating effects, selective and volumetric heating is achievable without direct interaction between the microwaves and their microwave-transparent surroundings. Such a heating mode also leads to reducing the thermal gradients throughout the bulk media, thus producing the uniform and efficient reactions (Fig. 2a) [9,54,55]. Generally speaking, the thermocatalysis and conversion reactions are driven by interaction among microwaves and materials with appropriate electromagnetic loss abilities. There are three interactions when the microwaves radiate on the materials, namely penetration, reflection and

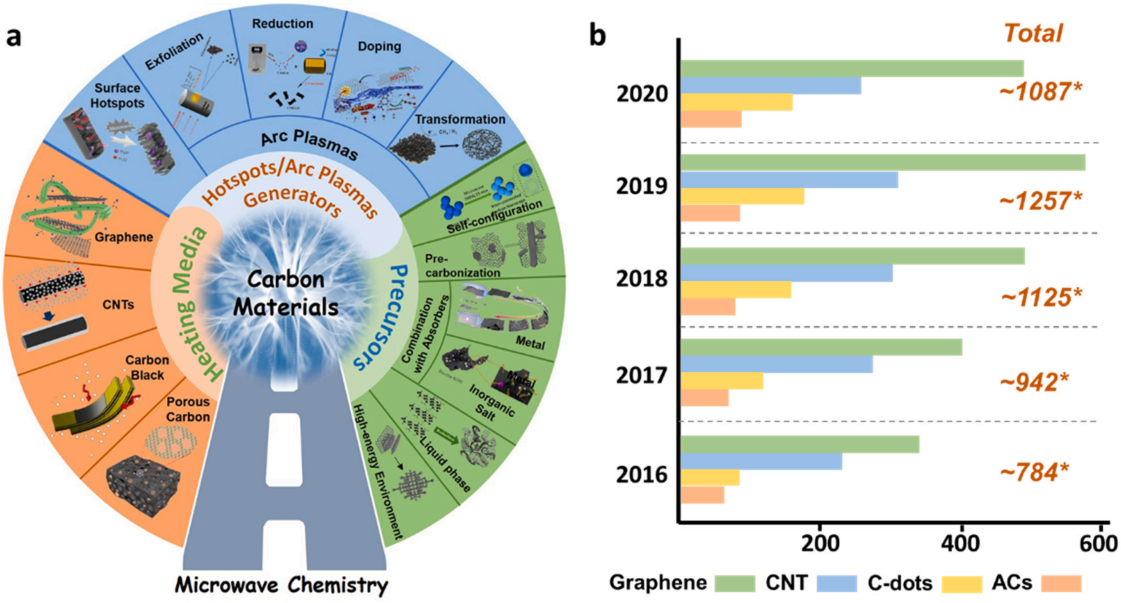

Fig. 1. (a) The application of carbon additives in microwave chemistry. (b) The statistical data of published papers in recent five years (* data from Web of Science).

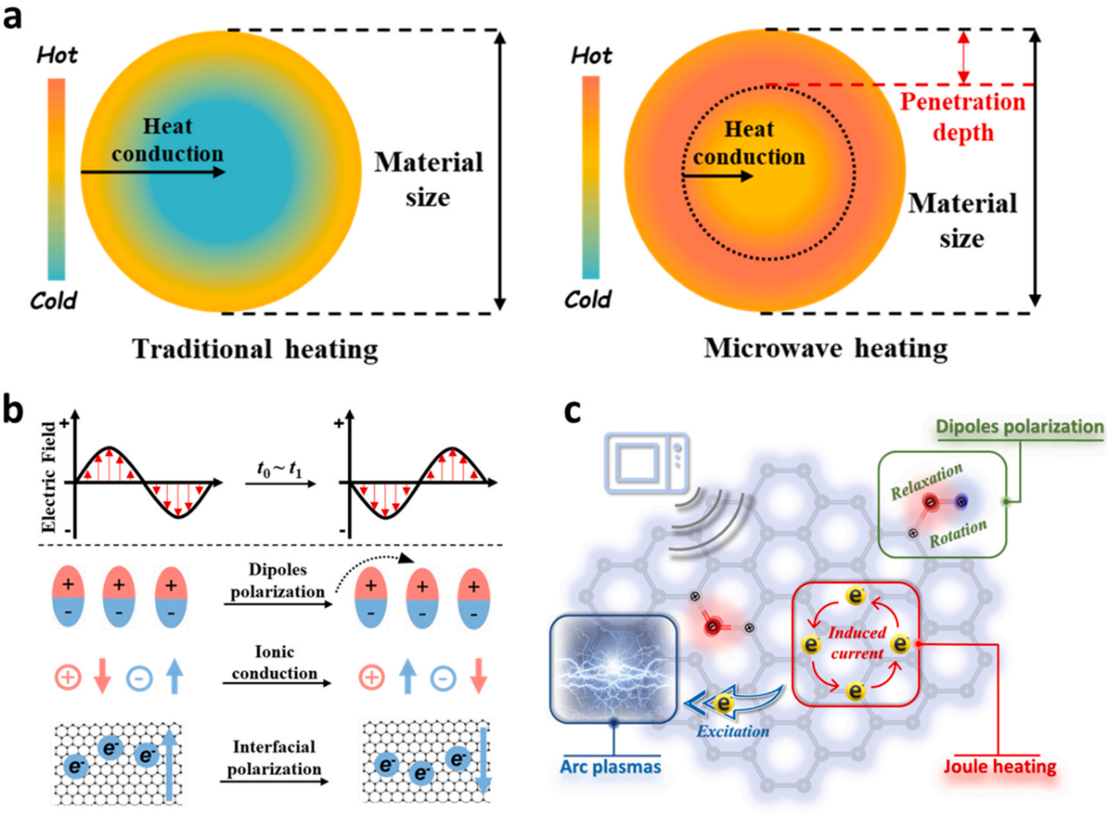

Fig. 2. (a) The comparison of microwave heating and traditional heating; (b) The summarized interaction mechanisms of electric field component and absorber. (c) Typical/intimate interaction mechanisms between a modern carbon and external alternating electromagnetic field.

absorption [54]. To be specific, the penetrated and reflected microwave are useless for materials engineering, to make matters worse, these unfavorable waves have the underlying negative effects on the internal circuits or magnetron component of microwave reactors. That is to say, the absorbed microwave energy is the only available source for materials processing, accompanied by the different thermal effects. In this part, the interaction mechanism and derived effects between microwave irradiation and carbon absorbers, the factors affecting the MA properties of carbon materials, together with related key parameters to evaluate the interaction effects are introduced to deepen the understanding for carbon-enabled microwave chemistry.

2.1. Interaction mechanism of carbon and microwave

Typically, the microwave reactions can be operated in the liquid and solid phases depending on the states of microwave absorbers. For the microwave-driven/assisted liquid reactions, the interaction mechanism among microwave and liquid medium involves two main processes: dipoles polarization and ionic conduction. As for the dipole polarization, the electric field component of microwave causes the polar molecules to rotate and try to align along the same orientation at a certain frequency. The accompanied friction and collision by the increased molecular rotation and moving, ultimately result in in-situ generated microwave heating. In the case of ionic conduction, the ions in liquid medium will move through the solution driven by electric field force. Also, due to the constant-fluctuation characteristic of ions along with electric field, the friction and collision of ions with surrounding molecules similarly cause the significant increase of the local temperature [55,56]. When solid media are adopted, at the surfaces or grain/phase boundaries of dielectric materials, the interfacial polarization can also be aroused due to the delocalization of bound charges from the equilibrium position [9]. The electrons periodically move and collide with the atoms in polarized regions, thus arousing the *Joule* current and triggering the temperature increment. In view of the above-mentioned mechanisms, the interaction of electric field component and absorber is summarized in Fig. 2b, which have great significance in initiating the reactions. In addition, the microwave reactions referring to solid phase absorber can also be triggered by magnetic field component of microwave, and the magnetic loss of certain solid ferromagnetic media, such as Fe, Co, Ni, and their derived compounds, plays the key role in improving the reaction temperature [57].

In the case of carbon absorbers, the dielectric loss is dominated, with a low magnetic loss due to the weak magnetic property when exposed to microwave fields. The intimate interaction between carbon materials and external alternating electromagnetic field usually involves diverse processes: the rotation/relaxation of dipoles from the defects and residual chemical bonds/functional groups as well as the high-frequency motion/excitation of conjugated electrons on carbon surface (Fig. 2c). Generally speaking, these processes will lead the fast heating effect on macro-scale as well as super-heating effect on nano-/micro-scale. The fast heating is the most common phenomenon, while the latter one usually in forms of hotspots or intensive light emission is hardly detectable under the microwave treatment [46,58,59].

Specifically, the fast heating is partly powered by the *Joule* heating as mentioned above. Furthermore, the dipole polarization has been found to transform the energy into heat, and thus boosting the fast temperature increasement. As for the light emission, due to strong interaction between the nearly free-moving electrons and electric field, the kinetic energy of these electrons increases and enables them to jump out of conjugate area on carbon surface quickly, resulting in the ionization of the gaseous species with an obvious light emission in the limited time and space. This phenomenon is perceived as arc plasmas or discharge. And an intensive generation of such ionized species/plasmas may hold great potential for the involved microwave reactions due to the characteristics of microscale dimensions and a unique high-energy-density.

Hotspots, as the isolated regions with excessive temperature, can occur in a tiny space due to the thermal instability derived from the nonlinear dependence of the electromagnetic properties of the whole material on temperature [60,61]. In other words, the dielectric properties in certain regions vary with the localized concentration of dipole/π-electrons on carbon surface, and result in different ability for microwave energy absorption. In addition, when microwave irradiate on carbon materials with the cavity structure, the surface can suffer from the microwave irradiation with higher intensity due to the multiple internal scattering modes. The absorption regions with the high energy corresponds to a fast heating rate, thus resulting in the formation of hotspots.

2.2. *Factors affecting the MA properties of carbon materials*

In view of the fact that the *Joule* heating caused by π-electrons is responsible for the thermocatalysis and conversion reactions, the carbon materials with sp^2-dominated hybridization states are widely used to initiate microwave reactions. In general, the energy conversion efficiency of microwave is also mainly influenced by the structure of carbon changing from nano-scale, micro-scale, to macro-scale. There is no doubt that the size of the π–π conjugated structure is related to the electrical conductivity of carbon materials and the formation of *Joule* current within the conjugated regions (Fig. 3a). But it is not always favorable. For the carbon materials with highly-ordered π–π conjugated structure (such as graphite), most of the incident microwave is reflected instead of being absorbed. As for the absorbed part, it can only dissipate on the surface or near-surface of carbon materials. On account of this, some advantages in terms of fast and uniform heating will no longer be operative in these cases.

Furthermore, the surface properties in terms of defects and functional groups of carbon materials also have the significant effects on its MA properties, further determining the materials engineering/manufacturing efficiency to some extent. In detail, the abundantly clustered defects and residual bonds on the carbon surface can increase the thermal energy caused by dipole polarization (Fig. 3b). Particularly, the n-type doping (such as nitrogen [62,63] and sulfur [64]) in carbon materials can provide extra electrons to the conjugated systems and create local dipoles, leading to the enhancement of *Joule* heating and dipole polarization for carbon materials. However, it was presented by our groups that the excessive defects or residual groups on the surface of graphene restricted the formation of *Joule* currents, thus leading to the remarkably decreased microwave absorption as well as the correspondingly low heating rate [65]. The quantitative relation of the dielectric loss and quality of rGO including size, defects, and residual bonds still need to be systematically studied.

The porous structure and wrinkled surface microstructure of carbon materials also have positive effects on the MA property of counterpart (Fig. 3c, d) [66–70]. The multi-reflection/scattering modes within the large pore and wrinkled surface are helpful for enhancing the attenuation of incident microwave. Furthermore, the curly and ultra-thin structure of the porous carbon materials provide the abundantly conductive paths for electron transfer, which boost the time-varying microwave-induced *Joule* currents and the generation of thermal energy in the resistive network. As such, it should also be carefully considered and finely operated when evaluating sp^2-dominated carbon materials as heating media in order to enhance the energy conversion efficiency (Fig. 3e).

2.3. *Key parameters to evaluate the interaction effects*

The key parameters of microwave additives or reactors should be carefully evaluated to guarantee the effective reactions. Before the microwave irradiation, the dielectric loss tangents (tan (δ)) of materials, which indicate the ability of the materials to be polarized at a given frequency and temperature, can be calculated by equation as follows:

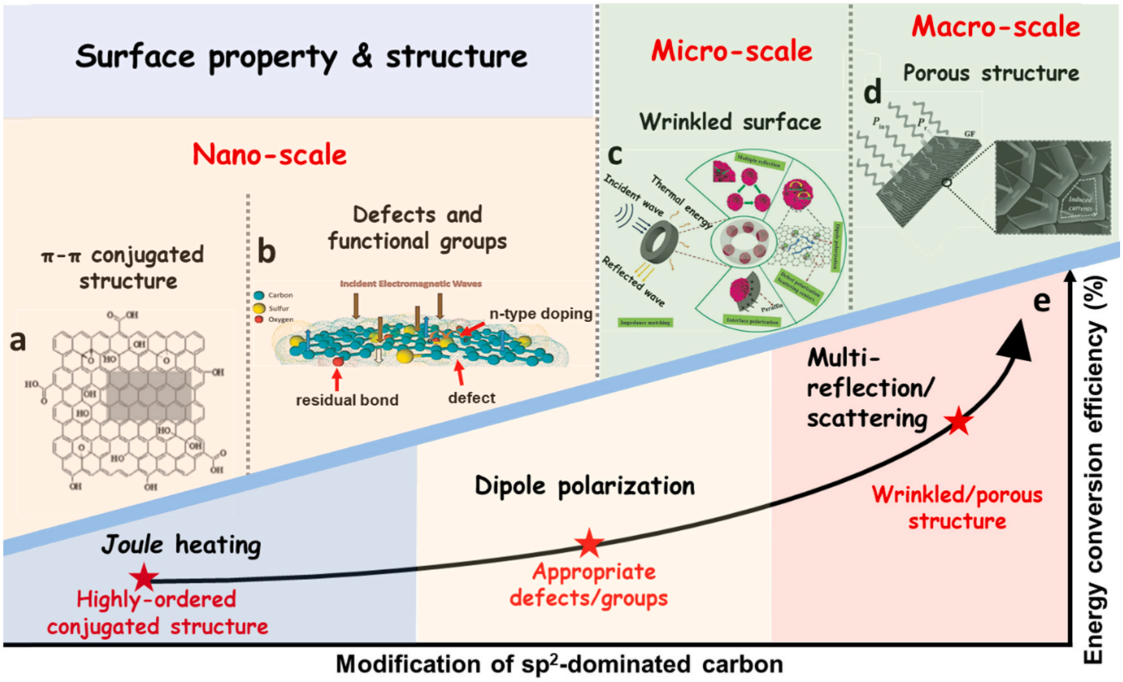

Fig. 3. Relationship between the structure of carbon changing from nano-scale, micro-scale, to macro-scale and energy conversion efficiency of microwave. (a) Schematic illustration of the rGO, where the shaded gray corresponds to conjugated region. Reproduced with permission [65]. Copyright 2012, Elsevier. (b) Schematic illustration for graphene with defects, residual bond and n-type doping. Reproduced with permission [64]. Copyright 2015, Royal Society of Chemistry. (c) Schematic illustration for the possible electromagnetic wave absorption mechanism of the wrinkled carbon microsphere. Reproduced with permission [66]. Copyright 2020, Elsevier. (d) Schematic illustration for the MA mechanism of the 3D free-standing graphene foam. Reproduced with permission [71]. Copyright 2015, Elsevier. (e) The energy conversion efficiency of sp^2-dominated carbon as a function of modification degree.

$$\tan(\delta) = \frac{\varepsilon''}{\varepsilon'} \tag{1}$$

Here, ε' is the dielectric constant, and represents the ability of the material to store electrical energy. ε'' is the relative loss factor of materials, and relate to the ability of the material to dissipate electrical energy.

On this basis, the average absorbed power (P) of materials can be calculated once the electric field intensity (E) and the angular frequency (ω) of microwave are confirmed following the equation [38]:

$$P = \omega\varepsilon_0\varepsilon''E^2 = \omega\varepsilon_0\varepsilon'\tan(\delta)E^2 \tag{2}$$

Here, ε_0 is the permittivity of free space ($8.854 * 10^{-12}$ F m^{-1}).

Moreover, penetration depth, that is determined by an internal point where the magnitude of microwave decreases to 1/e of the initial value on surface, has great importance for the ability to be heated by microwave irradiation [10]. Given that the microwave is composed of electric and magnetic fields and the carbon absorbers deliver the high dielectric loss and low magnetic loss, the corresponding penetration depth can be expressed as follows [72]:

$$D_p = \frac{\sqrt{2}C}{\omega\left\{\varepsilon'\left(\sqrt{1+tan^2(\delta)}-1\right)\right\}^{1/2}} \tag{3}$$

Here, D_p is the penetration depth and C refers to the speed of light.

Moreover, the penetration depth of microwave varies with the material types. Generally, polar solvent has a centimeter-scale penetration depth, e. g. water has a penetration depth of 1.4 cm under 2.45 GHz frequency of microwave and room temperature, so they can be heated effectively [11]. As for electron-rich solid systems like carbon materials, the corresponding penetration depth is only at the micrometer-scale, far lower than that of polar solvent, e.g. graphite has a penetration depth of 38 μm under same conditions [73]. Thus, the penetration depth becomes a critical parameter referring to the carbon-initiated microwave reactions. In particular, as shown in Fig. 2a, when the penetration depth is much smaller than the material size, the homogeneous heating is not achievable because the incident microwave is only absorbed at the surface or near-surface, while the heating of the inside part relies on the heat conduction as traditional one does. It is noteworthy that the well-matching between the size of carbon absorbers and the corresponding penetration depths should be taken into account to achieve the efficient and uniform heating in practical applications.

Furthermore, during the microwave heating process involved in carbon materials, both the extremely high temperature and heating rate can be obtained. As we know, the temperature as an important parameter determines the yield and properties of products during the thermochemical reactions, and the accurate measurement and control are crucial for the corresponding preparation. However, in terms of the frequently-used measuring tools, metallic thermocouples are not always suitable to measure the reaction temperature in microwave reactions since they can be heated by in-situ generated *Joule* currents on the surface of thermocouple, which is absorbers-independent. The infrared pyrometer or fiber sensors remains the principal tool in probing sample temperature in microwave equipment. Nevertheless, the measurement about the temperature information on nano-scale, especially in terms of the arc plasmas and local hotspots in carbon materials, are hardly realized. The optical emission spectroscopy has been conducted to monitor the system involved in arc plasmas directly, where the relative concentration of free radical can be acquired by collecting the optical signal in plasmas. Last but not least, it has been found that a feedback temperature control using microwave input power and/or irradiation time as manipulating variables is necessary for the precise temperature control in the reactors.

3. Carbon materials as microwave additives

It is indisputable that the structure and surface chemical properties of carbon materials have a significant influence on the MA properties, thus determining the conversion efficiency from microwave to thermal energy as well as the heating rate and attainable temperature. Consequently, diverse heating or super-heating effects can be produced when used different carbon materials as microwave additives. In this section, the recent research progress in carbon-participated microwave reactions is summarized and discussed. The corresponding three items including heating media, hotspots/arc plasmas generators and precursors are presented by utilizing the different carbon additives in microwave reactions. Concisely, traditional heating or super-heating referring to hotspots and arc plasmas can be produced when the carbon materials serve as microwave absorbers. Also, the carbon-based precursors are widely used and converted into the value-added products by microwave irradiation-driven structure reconfiguration.

3.1. *Heating media*

Owing to the extraordinary microwave heating effects and high thermal conductivity, the carbon materials (such as graphene [74], porous carbon [50,75,76], CNTs [77], and carbon black [78–80]) are widely used as heating media to promote the thermocatalysis and conversion reactions of surrounding/surface/inner species.

It was reported that the reduced graphene oxide (rGO) in the carbon world is the thinnest and most lightweight materials with highly efficient microwave attenuation property [81]. Accordingly, rGO is the widely-used heating medium in producing advanced materials [82–86]. For instance, a small amount of rGO can effectively absorb microwave for carbonizing cellulose nanofibers (CNFs). The introduced sodium before microwave irradiation can be maintained after the ultra-fast carbonization process for the CNFs, and offered favorable spaces for the insertion/extraction of sodium ions [74]. We recently found that randomly stacked graphene can also be used to construct a built-in microreactor, where the microreactor can enhance the yield of MOF-derived cobalt nanocatalysts up to 48.7 wt% and dramatic reduction of pyrolysis time (just 60 s) with low energy consumption (0.37% of traditional carbonization method) (Fig. 4a–c). The electrical conductivity and surface properties of graphene enabled the absorption of microwave irradiation, which was followed by the fast conduction of heat to the buried precursors [87]. It is easily adopted that the microwave heating in micro-/macro-scales is the result of energy accumulation by electrons and dipoles, sometimes, the graphene with fast heating effects might suffer from a self-quenching process by consuming the substrate functional groups in nano-scale and the corresponding effects were demonstrated by an impressive work (Fig. 4d, e) [88].

It had been verified that configuring rGO into a macroscopic and three-dimensional (3D) free-standing graphene foam can lead to the rapid absorption of massive incident microwave [71,89]. A typical work for applying the 3D carbon material as a heating medium was presented from Hu and the collaborators, as shown in Fig. 4f–h [47]. The nice work proposed a rapid, in-situ heating method to prepare the well-dispersed metal oxide nanoparticles with controllable size on a 3D carbonized wood host. The C-wood with the good thermal conductivity and moderate electrical conductivity contributed to the local *Joule* heating and guaranteed the rapid 3D heating of the overall materials.

Moreover, most of carbon materials can simultaneously serve as the substrates and heating media involved in microwave system due to the adjustable dimensions, indicative of the unique superiorities of microwave chemistry for the configuration of carbon-supported materials [90,91]. However, as for the coating/loading layer on carbon substrate, the thickness and dielectric property should be emphasized to avoid the non-uniform heating along the thickness direction and inefficient microwave absorption of carbon core. Yu et al. [92] prepared a nano-scale coating tied on 3D porous sponge-like CNTs in a mixed solution of polydimethylsiloxane (PDMS) and hexane. The CNTs served as the heating media/substrate, while the mixture exhibited a microwave-transparent characteristic. Impressively, the corresponding

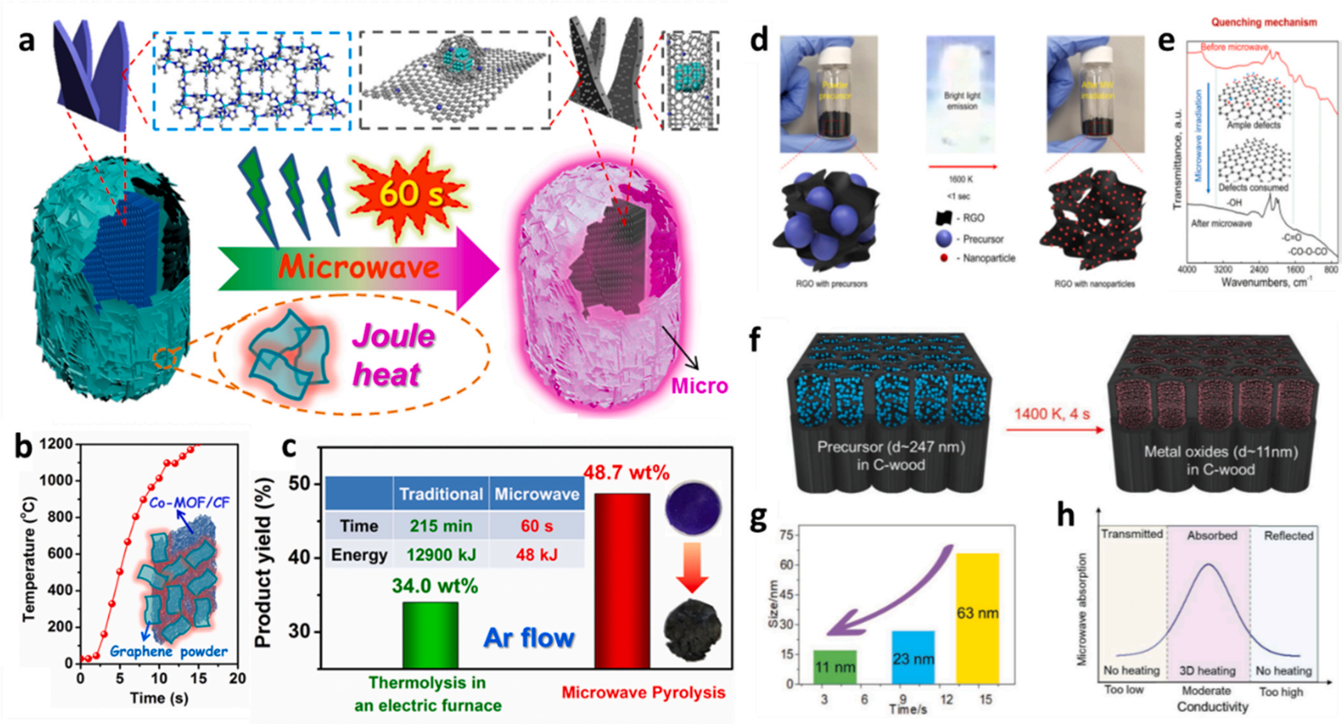

Fig. 4. (a) Schematic diagram for the preparation process from the Co-MOF to the Co-MOF-derived electrocatalyst using stacked graphene to construct a built-in microreactor. (b) Heating rate of graphene powder under microwave irradiation. (c) Comparison of electrocatalyst yield, total consuming time, and energy consumption by microwave and traditional carbonization method. Reproduced with permission [87]. Copyright 2020, Royal Society of Chemistry. (d) Digital photographs and schematics of the rGO loaded with precursor before and after irradiation with microwave, respectively. (e) FTIR and schematics of rGO before and after microwave treatment, indicating the consumption of functional groups during the microwave synthesis. Reproduced with permission [88]. Copyright 2019, Cell Press. (f) Schematic of the fabrication of metal oxide nanoparticles within the 3D C-wood substrate and corresponding digital photos. (g) Average size of nanoparticles with microwave time. (h) The MA performance of C-wood as a function of electrical conductivity. Reproduced with permission [47]. Copyright 2019, Wiley-VCH.

COMSOL simulations confirmed that the outer surface temperature of CNTs is the highest under microwave irradiation. And as the distance away from the CNTs surface increased, the temperature delivered a sharp decrease. So, the curing process of the PDMS should occur on the CNTs surface, and the coating layer thickness can be tuned by changing the concentration of the PDMS or irradiation time.

As mentioned above, the energy of microwave can be transformed into the *Joule* heat by the collision of π-electrons and carbon atoms, accompanied by a remarkable temperature increase of carbon absorbers. After that, the surrounding species will be heated by thermal conduction and radiation. The thermal conduction of materials is a result of collisions among atoms or molecules, while the thermal radiation is based on the infrared ray energy exchange from heating media to surrounding species. It was also found that the electrical energy supplied to π-electrons system on carbon materials is partly used to heat carbon materials themselves, and the rest of the energy can be dissipated directly into the surrounding species by the coupling of electrons on π-conjugated carbon system and the surface polaritons of nearby material. This effect had been confirmed by using the in-situ electron thermal microscopy to detect the remote heating behaviors of CNTs driven by direct current toward a silicon nitride substrate. Interestingly, at least 84% of the supplied electrical energy is dissipated directly into the substrate, rather than in the CNTs themselves [93,94]. It is conceivable that the alternating current driven by microwave on carbon surface bears some analogous effects. Even though it has not been applied to explain the processes referring to complex heat-transmission as yet, this effect might also be one of the potential possibilities for the realization of efficient microwave heating.

3.2. *Hotspots/arc plasmas generators*

Hotspots generated in carbon materials usually can be considered as “heat power source” in micro/nano-scale to drive the microwave-driven/assisted chemical reactions and have the great influences on this process. As mentioned above, the presence of hotspots is hardly detected based on current temperature measurement tools due to the limited resolution in time and space scales. Impressively, Yi et al. [95] proved the possible presence of hotspots by analyzing the recorded adsorption breakthrough curves of toluene vapor on activated carbon at microwave and traditional heating conditions. Generally, the temperature of the hotspots usually led to the ablation/shrinkage of the carbon materials channels, which is obviously troublesome and undesirable in most applications. Nevertheless, it was found that the formed hotspots can also be further used to prepare the advanced materials with unique and desirable features. Mai et al. [96] prepared the microporous soft carbon nanosheets (SC-NSs) via a microwave-induced exfoliation strategy, as shown in Fig. 5a and c–e. The conventional soft carbon compound (SC-MR) is obtained by pyrolysis of 3,4,9,10-perylene tetracarboxylic dianhydride (PTCDA). After the microwave irradiation, the skin-layer of SC-MR was oxidized and removed due to the formation of hotspots on surface. In detail, because the absorbed microwave irradiation on the surface is much higher than that of other regions of materials, the high heating rate will accelerate the oxidization process of surface layer to facilitate its removal. After the following exfoliation and pore-creating processes, the target nanosheets can be used as an advanced anode for sodium ion storage, as demonstrated in Fig. 5b.

Typically, the plasmas generated on carbon surface are composed of ball lightning and arc plasmas, as shown in Fig. 6a and b. The ball lightning plasmas are abundant at the beginning of microwave

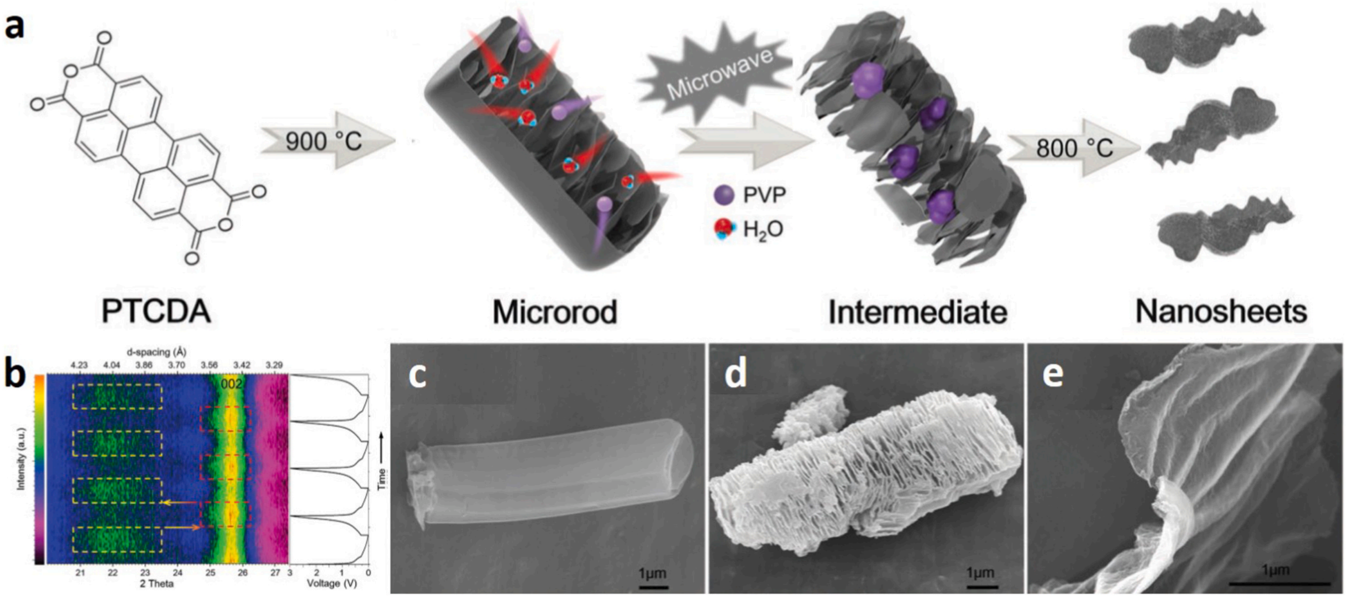

Fig. 5. (a) Schematic illustrations for the preparation of the SC-NSs materials. (b) The in-situ XRD patterns of SC-NSs for sodium ion storage. (c–e) Scanning Electron Microscopy images: the microrod (c), intermediate (d), and nanosheets (e).
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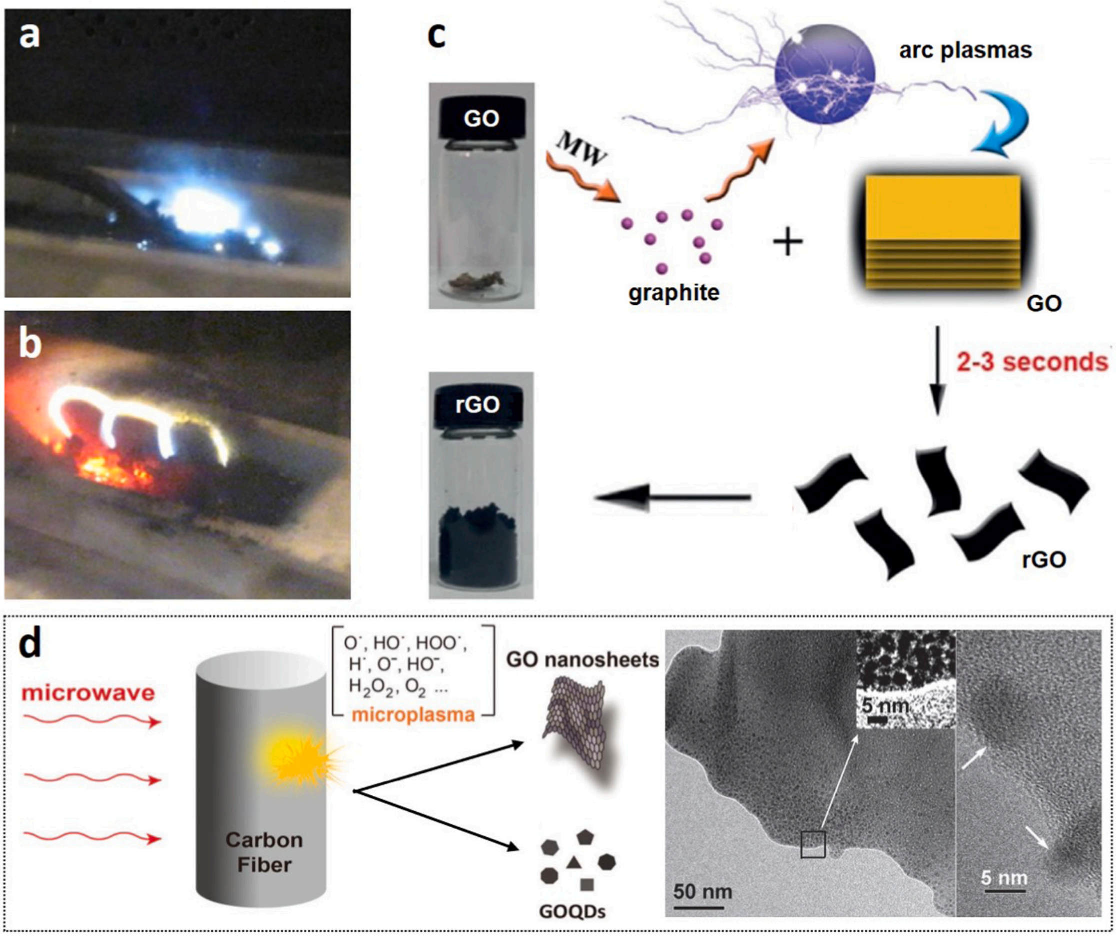

Fig. 6. The digital photographs of (a) ball lightning plasmas and (b) arc plasmas. Reproduced with permission [46]. Copyright 2011, Elsevier. (c) The exfoliation process of GO. Reproduced with permission [104]. Copyright 2017, Wiley-VCH. (d) The exfoliation mechanisms for the carbon fibers in water, and TEM images for GO nanosheets and GO quantum dots. Reproduced with permission [109]. Copyright 2018, Wiley-VCH.

treatment when the temperature of the carbon bed is still relatively low and this process maintains only a fraction of a second. However, these ball lightning plasmas has not been used for materials preparation, and even well explained by the widely-accepted mechanism. The arc plasmas, as common thermal equilibrium plasmas maintained at high temperatures, rely on the concentrated/focused electromagnetic energy and are derived from the ionization of the surrounding molecules in small volumes. The high-energy-density features need to be highly concerned and utilized, which can trigger an extraordinary space-confined reaction and will be of help for designing and modulating the microstructure and surface chemistry of materials [46,97–99].

It was reported that the arc discharge between graphite/coal rod electrodes under certain atmospheric conditions can result in a great diversity of effects, such as doping, transformation, intercalation, exfoliation, and reduction, especially in producing the advanced/value-added carbon materials [100–102]. This is also the case for arc discharge produced in the microwave system, where the various atmosphere, such as Ar, CH_4/N_2, CH_4/CO_2 and even air, can realize the above-mentioned effects [103–107]. For instance, the arc discharge of carbon in argon atmosphere can produce nitrogen doped carbon rods with the help of pyrrole. Carbon nano-filaments can be synthesized through microwave-driven decomposition on an activated carbon or char, where the excited electrons on carbon absorbers led to the activation of the CH_4 molecule and played a decisive role in the transformation process. Xu et al. [104] reported that a small amount of graphite can act as the catalyst to greatly promote the microwave-assisted exfoliation and reduction of graphene oxide (GO) in air. The free-moved π-electrons in flake graphite can efficiently absorb microwave and convert the nearby gas molecules into plasmas with excited states. Subsequently, the plasmas with local high-energy characteristics exfoliated and reduced GO into graphene with high specific surface area/lattice crystallinity, large C/O ratio, as well as improved electrical conductivity in a few seconds, as shown in Fig. 6c. The prepared graphene could be used as the anode material for boosting lithium and sodium storage, respectively.

The gas molecules near carbon surface are indispensable for the generation of arc plasmas. In the solid or liquid phase systems, the in-situ generated gas molecules can be excited. In this case, the arc plasmas in synergy with liquid/solid phase can also be used for advanced materials synthesis [73,108,109]. For instance, our group proposed a novel field-induced energy accumulation strategy to prepare the stage-1 dominated $FeCl_3$-graphite intercalation compounds in minutes. The ionized metal chloride with high energy density can be produced by the microwave-induced Joule heat and electron excitation, thus strengthening the intercalation reaction kinetics [108]. Yuan et al. [109] directly prepared monolayer GO quantum dots by irradiating carbon fibers with microwave in water, as shown in Fig. 6d. The carbon fibers absorbed microwave and intense arc plasmas are performed in the local regions near the surfaces along with obvious light emission and crackling sounds. The intense plasmas were generated by the excitation of surrounding vaporized H_2O molecule, and involved many highly reactive species such as O·, HO·, H·, O^-, HO^-, and O_2, which possessed high oxidation capability. Subsequently, carbon fibers were exfoliated and oxidized to produce the GO nanosheets from 100 to 200 nm in size and the GO quantum dots of about 5 nm in size as shown in Transmission Electron Microscope (TEM) images.

Typically, the hotspots and arc plasmas generated form carbon absorbers demonstrate the super heating effects with the localized high-energy characteristics driven by external microwave energy in a confined space. However, it is rare to emphasize such super heating effects when the carbon materials are exposed to microwave fields, which is “a dark box” and will hinder the sufficient reveal of corresponding reactions to a great degree. In future, the super heating effects as discussed in this part should be highlighted and arouse the attention of researchers if possible, which helps to deepen the comprehension for carbon-initiated microwave reactions, decouple the possible formation mechanism during nanomaterial manufacturing, and further expand the application of microwave technique.

3.3. Precursors

Converting the carbon-based precursors such as conjugated polymers, coal, and biomass into value-added products represents definite economic benefits and practical significance [5,11,110,111]. In this process, the fracture and re-formation of covalent bond is inevitable and usually requires harsh reaction conditions. Microwave-driven structural transformation has been developed and applied extensively due to the versatile control of significant reaction parameters and effects.

The self-configuration ability of carbon-based precursors can be associated to the accumulated energy that is determined by the MA property as well as the microwave irradiation time and power [112–115]. Moreover, for a certain carbon material, the optimal MA property can be obtained at a certain frequency band, which means that the incident frequency is also of great significance for microwave treatment. However, rare work devotes to reveal the preparation and conversion process of advanced materials by microwave in different frequencies. Thus, the examples we discussed here are on the basis of the microwave irradiation at 2.45 GHz frequency that is usually used in commercial microwave equipment. Li et al. [112] synthesized the oxygen-decorated graphene by irradiating carbon cloth in air for 6 s under 700 W. The inter-connected carbon nanocage can be obtained by irradiating Ketjen black with microwave for 15 min under 700 W, which seems to be a significant breakthrough for rapid, large-scale production of cathode materials in aluminum battery field, as shown in Fig. 7a and b [116]. Although the transformation mechanism from Ketjen black to carbon nanocage remains less concerned, it is believed that microwave energy absorbed by Ketjen black precursor might determine the corresponding transformation process. Moreover, the ratio of C=O to C–O on mesoporous carbon surface was optimized by changing the irradiation time under a fixed microwave power (100 W), which led to a high-performance electrocatalyst for electrochemical production of hydrogen peroxide (Fig. 7c and d) [117]. Inspiringly, the microwave heating exhibited the potential for modulating the surface chemical properties and micro/nano-scaled structure of carbon-based precursors, where the performance of products can be regulated to meet the actual requirements in energy storage and conversion fields [118]. Most of carbon-based precursors can arouse the self-configuration process by the absorbed energy, while others with inferior MA properties cannot achieve this process even under long irradiation time and high power. Thus, some approaches to improving the MA properties and/or constructing high-energy environment are essential to increase the energy accumulation and achieve the structure reconfiguration of carbon-based precursors.

For the modulation of MA property, most of precursors such as GO and biomass usually require pre-heat treatment or pre-reduction to improve the dielectric loss. For instance, by radiating ethylenediamine-reduced precursor with microwave, our groups prepared the three-dimensional and ultra-light graphene aerogels with excellent elasticity. Remarkably, the restored conjugation of sp^2 regions and the enhanced π–π interaction in the cross-linking sites caused by microwave irradiation can help to increase the elastic stiffness [119]. Irradiating the slightly reduced GO with microwave leads to the extremely high energy release, and gives rise to ultra-high heating rate and temperature. And then the fast removal of residual groups and reordering of the carbon atoms on basal plane within seconds are achieved. Voiry et al. [14] reported a simple, rapid method to reduce rGO into graphene with high values of $> 1000\ cm^2\ V^{-1}\ s^{-1}$ for electron mobility using 1–2 s microwave irradiation, as shown in Fig. 8a–c. It was found that the Raman spectrum for microwave-reduced rGO (MW-rGO) was similar to that of graphene obtained from CVD method. When MW-rGO was adopted as the catalyst support for oxygen evolution reactions, the onset potential decreased to < 200 mV and the catalytic activity was dramatically

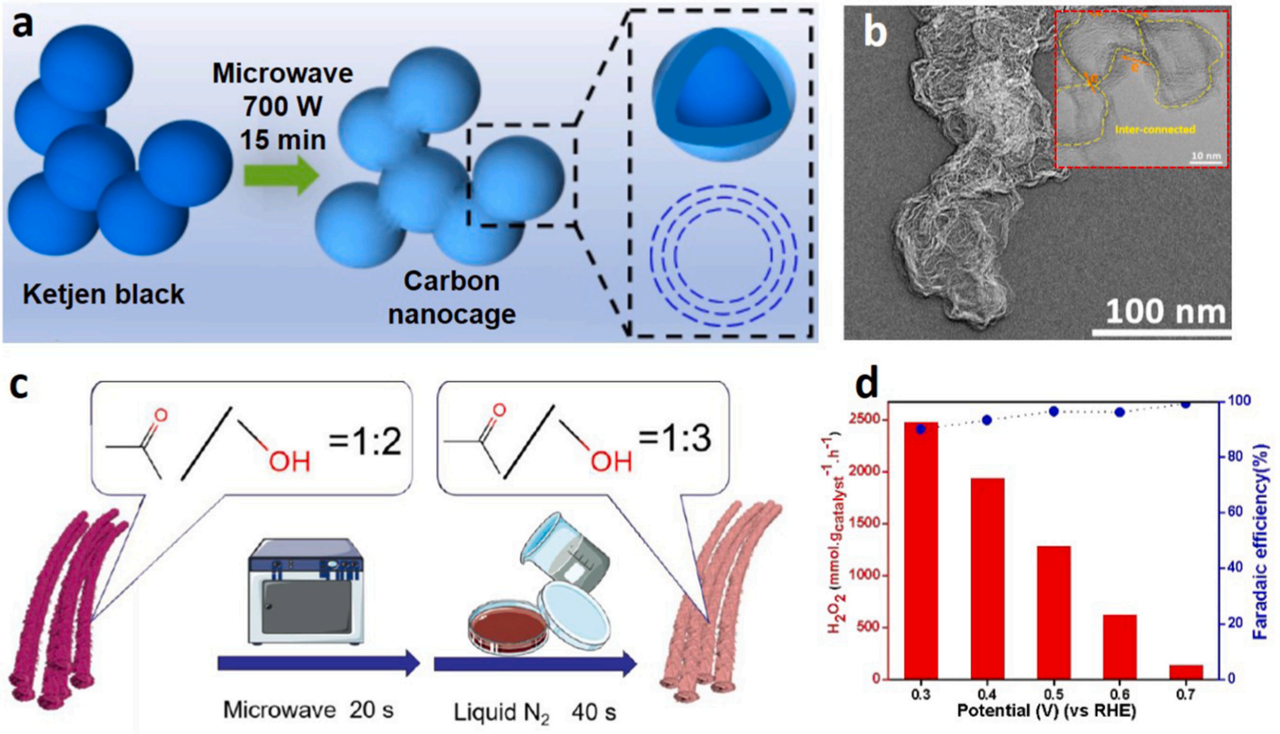

Fig. 7. (a) Schematic illustration for the structural transformation from Ketjen black to inter-connected carbon nanocage. (b) The TEM images of carbon nanocage (inset: the corresponding high resolution TEM image). Reproduced with permission [116]. Copyright 2020, Elsevier. (c) Schematic of the preparation for CMK3–20 s (d) Faradaic efficiency and H_2O_2 production capability for CMK3–20 s catalyst under different applied potentials. Reproduced with permission [117]. Copyright 2019, Royal Society of Chemistry.

enhanced. Obviously, the promising strategy featured the super advantages in tuning the micro/nano-scaled structure of desired materials. Ji et al. [120] prepared a microwave-treated hard carbon (MV-HC) by irradiating a low-temperature pre-annealed hard carbon (LT-HC) with rich defects, as demonstrated in Fig. 8d–f. It was decoupled that the ultra-fast characteristics of microwave treatment can not only make the defects of hard carbon be well kept, but also enhance the electronic and ionic conductivity. This resulted in the higher slope capacity of 196 mA h g^{-1} than that of the LT-HC (162 mA h g^{-1}) and conventional-heated hard carbon (HT-HC, 127 mA h g^{-1}).

Furthermore, it has been proved that positive modification of the carbon precursors by some special non-carbon additives with high dielectric loss can lead to the optimized MA properties [121]. Metal nanoparticles and derivatives are widely used as another type of microwave absorbers to produce porous graphene under a microwave heating process, which shows great potential in configuring the porous nanomaterials. Zhou et al. [48] employed the metal nanoparticles to promote the microwave combustion of carbon within tens of seconds, leading to the formation of the porous graphene with controllable pore size. Park et al. [49] also synthesized palladium-embedded 3D porous graphene with nanohole structures by multistep microwave reactions. They found that every nanohole were embedded with the Pd nanoparticles, which indicated that the Pd nanoparticles as microwave absorbers functioned to catalytically oxide the near carbon atoms and thus engineer the nanoholes on the graphene sheets. In addition, the microwave-driven inorganic salts (KOH, $ZnCl_2$) activation from biomass to activated carbon with super high specific surface area have aroused wide attentions in energy storage fields [122,123]. Impressively, the inorganic salts play dual roles of microwave absorber and activating agent in these processes. For instance, a porous carbon with large surface area (1229 m^2 g) and rich oxygen functional groups (C/O ratio of 1.66) was obtained through a convenient/rapid microwave-driven carbonization combining with KOH activation for pre-carbonized camellia oleifera shell. When used as the supercapacitor electrode, the porous carbon prepared by microwave achieved a higher capacitance of 251 F g^{-1} than that of the activated carbon (214 F g^{-1}) derived from traditional carbonization/KOH activation method in 6 M KOH electrolyte [50].

Moreover, microwave plasmas with high-energy environment can also be used to convert the carbon precursors into other target materials with different microstructures and physicochemical properties [124,125]. To be specific, microwave can transfer the energy into the internal energy of gas phase, and thus the plasmas, such as glow, corona, and arc discharge plasmas, generated and participated in the desired reactions as the high-energy media. The arc plasmas are derived from the electron excitation on absorber surface, which have been systematically discussed in the previous section. The generation of glow plasmas usually require an ultra-high power (usually range from 600 W to 6 kW) and low gas pressure. Generally, the advanced carbon materials can be modified and synthesized by using glow plasma-enhanced etching and chemical vapor deposition. More interestingly, these solid carbon precursors can also be converted into value-added products by glow plasma-induced solid-vapor-solid transformation process. For instance, the transformation of graphite into diamond was achieved when H_2 was used as gas source of plasma. The transitional hydrocarbon radicals was in-situ formed through etching the graphite with hydrogen plasma, and then deposited on the near Si wafers in the form of diamond crystal [23,52]. In the meantime, the corona discharge plasmas can be generated in highly inhomogeneous electric fields caused by irradiating an absorber with very large curvature of surface. As such, the high-quality "Snowing" graphene was synthesized by utilizing microwave-induced corona discharge on SiO_2/Si in methane atmosphere [16].

In addition, most of liquid solvents can also absorb microwave and promote the transformation of carbon-based precursors. Moreover, the corresponding physicochemical characteristics in terms of chemical composition, structure, size and morphology can be tuned with the help

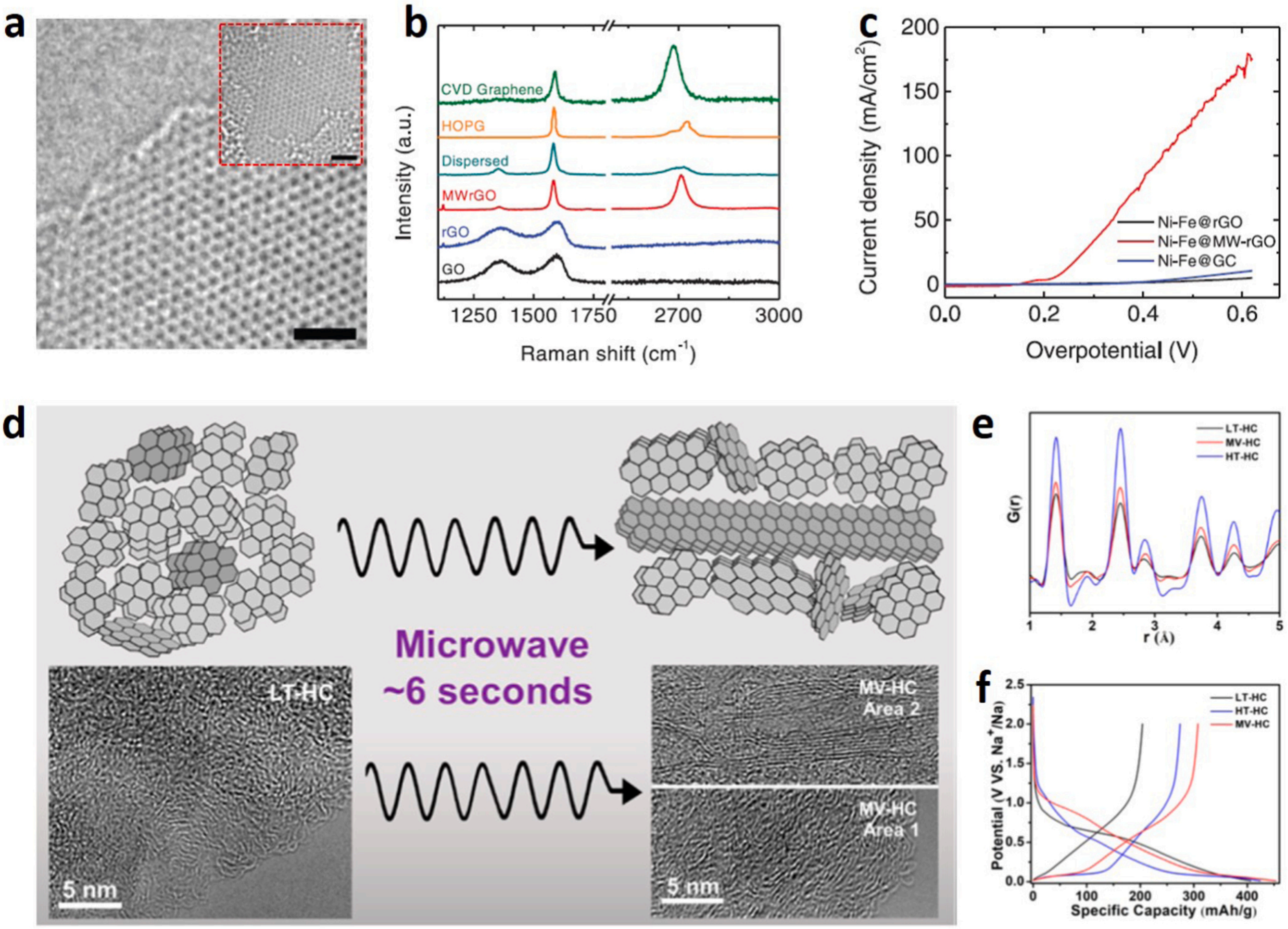

Fig. 8. (a) The high resolution TEM images of MW-rGO showing highly ordered structure, inset: single-layer rGO present the high density of defects. Scale bars, 1 nm. (b) Raman spectra of MW-rGO and other graphene-based samples. (c) Polarization curves of Ni-Fe layered double hydroxides deposited on MW-rGO compared with glassy carbon and rGO. Reproduced with permission [14]. Copyright 2016, American Association for the Advancement of Science. (d) Schematic illustrations of the microwave-assisted transformation of MV-HC and TEM images of LT-HC and MV-HC. (e) The neutron total scattering results of MV-HC, LT-HC, and HT-HC. (f) First-cycle galvanostatic curves of MV-HC, LT-HC, and HT-HC at 20 mA g^{-1}. Reproduced with permission [120]. Copyright 2018, American Chemical Society.

of diverse solvents (such as water, polyols, and ionic liquids) and their smart combinations [51,55,126,127]. Notably, as for an open system, the attained reaction temperature is enslaved to the boiling point of the used solvent. For example, the boiling points of ethylene glycol and glycel are 198 °C and 290 °C, respectively, which hinder the preparation of carbon materials when an inescapable high reaction temperature is involved. Moreover, microwave-driven/assisted hydrothermal/solvothermal methods have also been applied to the fast preparation of carbon materials in closed reaction systems at elevated pressures and temperatures [56].

4. Conclusions and outlook

In summary, this review proposed the state-of-the-art carbon-enabled microwave schemes regarding the engineering of advanced materials, from the mechanisms to the manufacturing. Owing to the key features of microwave heating (e.g. uniform, volumetric, fast and selective heating), it has been proven as one of the most effective pathways to manufacture and process the carbon materials than conventional ones. Reciprocally, due to the remarkable and adjustable physicochemical characteristics, the carbon materials are widely used as additives to promote the development of microwave-assisted/driven reaction and nanomaterial manufacturing in terms of micro- and macro-scale. In essence, the conversion processes from microwave energy to terminal thermal and/or light energy determine the reaction efficiency, where the carbon materials can play a “bridge” or “catalyst” role by accumulating the intrinsic energy.

The microwave effects involved in carbon materials benefit from the *Joule* current as well as dipole polarization driven by electric field components. And the MA properties can be tuned by changing the surface property and micro/nano structure of carbon materials. On account of this, driven by the microwave, the chemical reactions enabled by carbon additives show unique and indispensable advantages in materials engineering. The uniformly heating environment or localized high temperature micro-environment derived from hotspots/arc plasmas is easily formed when the carbon materials serve as microwave absorbers, which will be helpful for the corresponding reactions involved in the thermocatalysis and conversion processes. Most carbon-based precursors can also achieve the self-configuration due to the extraordinary MA properties under microwave irradiation. In addition, the approaches to improving the accumulated energy to achieve the structure reconfiguration of carbon-based precursors also have been reviewed, which hope to extend the applications of microwave processes involved in carbon additives.

Although the microwave heating continues to grow as a star technology, the fundamental research into the mechanism of microwave heating has not grown at the same rate and there are still plenty of technical bottlenecks that need to be tackled:

(1) It is well known that the dielectric loss of carbon materials has great significance in the overall energy conversion efficiency, nevertheless, it is difficult to establish a standard library for evaluating its loss capacity due to the diversity of carbon species and the difference of its surface properties.
(2) Also, in order to guarantee the uniform heating, sometimes the pre-mixing of carbon absorbers and reactants is indispensable. However, the recycle and structural changes of absorbers are often neglected, which may have adverse effects on the properties of products.
(3) In addition, the advantages of microwave processes involved in carbon additives are not sufficiently exploited yet. For instance, the localized superheating effects that are from the hotspots and arc plasmas with multiple capabilities to enable or intensify the specific chemical reactions, are less concerned in recent years.
(4) Furthermore, the involved molecular dynamic process in nanoscale is lack of model building and precise characterization as yet. For example, the microwave plasmas or fast heating process can be monitored by some operando techniques, such as optical emission spectroscopy as well as color ratio pyrometry, even so, the reactions information referring to limited space is hardly obtained.

Current studies referring to microwave-driven/assisted schemes have achieved the scale-up of products in the industrial level to some degree. However, there are still some issues remain to be finely responded:

1) For example, the microwave reactors in batch mode operation may feature low cost, however, small capacity and long processing time restrict its practical application. Thus, developing the microwave reactors in continuous mode operation for large batches of process at reasonable yields and reproducibility is highly desired.
2) In addition, a distinct drawback to microwave heating is the lack of heat uniformity for absorbers with microns to centimeter scale in size in industry. In fact, the heat uniformity for absorbers is dictated by the frequency of incident electromagnetic wave and penetration depth. By adopting variable frequency of microwave equipment, the penetration depth of absorbers will change, thus, the heating homogeneity can be improved and the quality of product will be enhanced to a large extent. These corresponding strategies will provide the guidance for carbon-enabled microwave reactions especially in industrial level.

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.

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