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Microwave-induced temperature fields in cylindrical samples of graphite powder – Experimental and modeling studies

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ABSTRACT

Microwave heating of graphite powder is the key process of a number of promising applications and emerging technologies ranging from kiln crucibles to synthesis of graphene. However, this process is typically studied in terms of structural parameters of the resulting products, whereas the prime electromagnetic and associated temperature characteristics remain insufficiently studied. In this paper, we investigate microwave heating of graphite powder in a batch (non-resonant) and waveguide (resonant) reactors using multiphysics (electromagnetic–thermal) modeling and experimentation. Temperature-dependent material parameters of graphite power are determined from measurements and appropriate physical models. Non-uniform temperature fields and their trend for relatively quick homogenization are shown to be conditioned by high values of the loss factor and thermal conductivity. These parameters are also found to be responsible for the effect of post-microwave heating. It is demonstrated that processing of graphite power in a resonant reactor may be convenient for controlling the heating rate and the level of temperature uniformity.

1. Introduction

As a form of pure carbon, graphite appears to be a well-studied material, but recently, its behavior under microwave irradiation has received special consideration in different technical areas. The growing interest in this topic seems to be linked with the fact that microwave heating of graphite can be found in the core of a number of emerging methods of efficient production of graphene and other carbon-based materials.

The extraordinary electrical, mechanical, optical, and thermal properties of graphene have shown that this material has the potential to revolutionize many applications, including solar cells, lithium-ion batteries, water desalination, nanocomposites, and medical diagnostics/sensors, to name a few [1,2]. A bottleneck for widespread adoption of graphene-based technologies is efficient mass production of graphene sheets, as the known methods are still technically demanding, expensive, and not always successful in producing graphene of desirable quality [1–3]. Chemistry currently plays an increasingly important role in the development of practical techniques of graphene production [3–5], and there are a number of chemical methods for generating graphene from graphite and its derivatives, including techniques based on exfoliation of graphite oxide [4,5], which appear to be scalable and afford the possibility of high-volume production. Since the exfoliation process is energy consuming and must be carried out at high temperatures, there are reports (e.g., [6–18]) about using microwaves as an alternative, greener and more efficient, energy source in producing graphene.

On the other hand, known as a strong absorber of microwaves, graphite has been also studied as a material that can be transformed by microwave heating, giving rise to new materials with tailored properties [19,20]. Other works were focused on graphite as a material for a crucible used in kilns for hybrid microwave heating [21], a substance assisting in microwave pyrolysis [22,23] and in microwave carbothermal reactions [24].

The quoted papers show many examples of quite peculiar and hardly predictable behavior of graphite-based samples (including production of sparks or electric arc formation [20,25,26]) under microwave irradiation, but do not outline a clear picture of microwave heating of graphite as a multiphysics phenomenon. One of the reasons for that is the deficiency of systematic studies of interaction of the material with microwaves in closed cavities. In most papers dealing with microwave heating of graphite powder, the analysis was focused on structural parameters of the resulting

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products rather than on the prime electromagnetic and associated temperature characteristics.

In particular, to the best of the authors' knowledge, there are no studies focused on the process of formation and evolution of microwave-induced temperature fields in graphite powder. For instance, in [21], temperature of a sample heated in a microwave oven was recorded (using a thermocouple) at a single (non-specified) point, but no underlying electromagnetic characteristics of the microwave system were considered. In [24,25], a sample of graphite powder was heated in a dominant-mode waveguide applicator in positions corresponding to maxima of the electric and magnetic fields of an empty waveguide without studying what actually happens with the fields in the presence of the heated material. In all these papers, intrinsic non-uniformity of microwave heating was not addressed, as the considered graphite samples were chosen to be very small and uniformity of heat release there was assumed to take place. Results of computational studies of microwave heating processes involving graphite samples [23,27] cannot be taken with confidence since the models employed in these works used an inadequate assumption that graphite's loss factor is negligible – in fact, graphite is known to be a strong microwave absorber (see, e.g., [20,21,28]).

Overall, the lack of studies of the basics of heating of graphite by microwaves results in lack of clarity about the related physical phenomena and available options for controlling the characteristics of microwave-assisted processes involving graphite and its derivatives. Yet, understanding the features of formation of temperature fields in graphite samples may be helpful in explaining particular effects, developing applied technologies employing controllable microwave heating of graphite-based materials, and designing enlarged microwave systems for up-scaled processes.

In this paper, we report the first results in the original study of microwave-induced temperature fields in cylindrical samples of graphite powder. Temperature fields are considered in two distinct (from a microwave engineering viewpoint) systems – in a waveguide/traveling-wave reactor and in a small non-resonant batch reactors, fed by a solid-state generator allowing for rigorously controlling frequency and magnitude of the source. Temperature is measured with the use of a fiber optic sensor (in different points inside the sample) and an IR camera (on the surface of the sample). The transient heating processes in both systems are analyzed with the use of a multiphysics model computing 3D temperature fields from 3D patterns of dissipated electromagnetic power; the computation is done in the framework of an iterative procedure solving the coupled electromagnetic and heat transfer problems. Whenever possible, the model uses temperature-dependent material parameters of graphite powder that are either determined experimentally, or calculated from appropriate physical models. Computations and experiments show high heating rates and inhomogeneous distributions of dissipated microwave power that tends to even up relatively quickly. An effect of post-microwave heating is demonstrated and can be explained by the non-uniformity of microwave heating and high thermal conductivity of graphite powder. Controlling the heating rate and temperature distribution is shown to be possible in a traveling-wave system where the sliding shorting wall controls a regime of field propagation.

2. Experimental system

Control of microwave heating is one of the major challenges of microwave power engineering, and a search for some correlation between the temperature field and electromagnetic characteristics of the system is a crucial part of development of any efficient microwave-assisted application. Such a search, however, may result in a somewhat indefinite output due to the typical radiation properties of a magnetron, a core device of all conventional sources of microwave energy, emitting signals of a rather chaotic magnitude in a certain frequency band; examples of measured magnetron spectra can be found, e.g., in [29–31]. In order to overcome this issue, in this study, we worked with the MiniFlow 200SS (SAIREM SAS, Neyron, France), a microwave system that employs a solid-state generator providing precise control over the frequency of excitation (from 2.43 to 2.47 GHz with a step size of 0.1 MHz) and the level of microwave power (from 0 to 200 W with a step size of 1 W).

The experiments were carried out in both reactors of the MiniFlow set. The traveling-wave reactor (Fig. 1(a) and (d)) is built on a bended WR430 waveguide section with an enlarged cylindrical space for the sample to be processed in the system. This space is bounded by a cylindrical Teflon tube (internal diameter 60 mm, height 94 mm) and accessible (for measurements and visual observation) through two (vertical and horizontal) metal tubes serving, due to their lengths and diameters, as cutoff circular waveguides preventing leakage from the cavity. The reactor is excited through a coax-to-waveguide transition. On the opposite end, the terminal section contains a sliding shorting wall controlling wave propagation in the system.

The batch reactor (Fig. 1(b)) consists of a small cylindrical cavity (internal diameter 63 mm, height 46 mm) containing a concentrically positioned cylindrical Teflon cup (internal diameter 23 mm, height 67 mm) intended for holding the sample in a cylindrical vessel which is put into the system through a cylindrical hole in the cavity's upper lid. Similarly, due to the large thickness of the lid (53 mm), this hole serves as a cutoff waveguide. The cavity is fed via a coaxial cable whose internal conductor is connected with a metal T-shaped structure inside the cavity.

In the experiments, we worked with synthetic graphite powder (Sigma–Aldrich Co., LLC) whose particles had diameter < 20 μm in a Pyrex vial (internal diameter 20 mm, height 75 mm). Internal temperature of the sample was measured by a fiber optic sensor (Neoptix, Inc.) held, except its tip, inside a glass capillary (ID 2 mm) and placed at different heights h in the cylindrical sample in the vial (as shown in Fig. 1(c)). Temperature on the cylindrical surface of the sample was also recorded through the side hole (diameter 15 mm) of the waveguide reactor (Fig. 4 (a) and (d)) with the use of the FLIR i7 infrared camera (FLIR Systems, Inc.) capable of monitoring the temperature field against the associated scale which is interpreted as the available minimum and maximum temperature within the pictured field.

3. Computational technique

Computer simulation of electromagnetic and thermal processes in both MiniFlow reactors was carried out in accordance with the computational scheme for an iterative solution of the electromagnetic–thermal two-way coupled problem [32–34]. Aiming to reach a high level of adequacy in representing microwave heating scenarios, this advanced technique accounts for temperature-dependent electromagnetic and thermal material parameters of the heated material and upgrades their values in all relevant grid cells after each heating time step. Internal structure of the reactors was precisely reproduced in parameterized 3D models developed for the full-wave 3D conformal finite-difference time-domain (FDTD) simulator QuickWave ver. 2013 [35]. The internal layouts of the reactor's components in the models are shown in Fig. 1(d) and (e). The computational algorithm was implemented using the core electromagnetic solver of the QuickWave package and its dedicated procedure for solving the heat transfer problems – the QuickWave Basic Heating Module.