Chemical Engineering and Processing 76 (2014) 1–5

Contents lists available at ScienceDirect

Chemical Engineering and Processing: Process Intensification

journal homepage: www.elsevier.com/locate/cep

Quasi-stable temperature of the steady state of microwave heated hematite

K. Kashimura[a,*], N. Sabelstrom[b], K. Imazeki[b], K. Takeda[b], M. Hayashi[b], T. Mitani[a], N. Shinohara[a], K. Nagata[c]

[a] Kyoto University, Gokasyo, Uji, Kyoto 611-0011, Japan
[b] Tokyo Institute of Technology, Ookayama 2-12-1, Meguro, Tokyo 152-8552, Japan
[c] Tokyo University of the Arts, 12-8 Ueno Kouen, Taito-ku, Tokyo 110-8714, Japan

ARTICLE INFO

Article history:
Received 27 June 2013
Received in revised form 29 November 2013
Accepted 2 December 2013
Available online 11 December 2013

Keywords:
Metal oxides
2.45 GHz
Microwave processing
Hematite
Stability of processing

ABSTRACT

Microwave heated materials often reach a quasi-stable temperature resulting in thermal runaway. To control steady state in microwave processing, it is important to predict the quasi-stable temperature of the steady state. We demonstrated that the microwave heating behavior of hematite varies significantly with its initial temperature. In microwave heating, hematite samples could not be heated from room temperature, whereas hematite samples preheated to 410 °C or higher was heated to a temperature of 1020 °C. The microwave heating behavior can be accurately predicted by considering the steady-state energy balance.

1. Introduction

Microwave heating has been attracting great interest as a new means of supplying energy to processes. Microwave heating can produce metal sintered compacts that differ from conventional ones due to the generation of non-equilibrium temperatures on the microscale [1–6]. Ishizaki and Nagata [2] constructed a high-power (12 kW) continuous 2.45-GHz microwave furnace and Peng et al. [3] constructed a 915-MHz, 225-kW microwave furnace to scale up steel production. The above mentioned studies all used microwave energies, resulting in rapid chemical synthesis. Microwave heating of powder compacts has been studied for several decades with a view to apply such a technique to industrial processes. Cheng et al. [7] compared the heating rates of metals, ceramics, and metal–ceramic composites in different microwave fields. They found that the magnetic component cannot be ignored when considering the energy loss in metals subjected to a microwave field. Ma et al. [8] systematically investigated the absorption and heating characteristics and the microstructural evolution of porous copper powder metal compacts irradiated with 2.45-GHz microwaves. They observed the heating characteristics of copper particles with various radii. The above studies [7–9] demonstrate that the Mie theory for a single particle can account for the high heating rates generated by the microwave magnetic field in microwave sintering experiments.

In contrast, few studies have used stability theory to predict the steady state temperature of microwave heating despite the importance of steady state temperature prediction for industrial processing. To realize efficient microwave processing, the temperature of high temperature in the steady state (i.e., the thermal runaway) should be predicted (in this paper, this temperature is defined as the quasi-stable temperature). However, two problems need to be overcome to make this possible. The first is the scarcity of data for the absorption properties as a function of temperature. Most measurements of microwave absorption properties have been performed at temperatures near room temperature. The absorption properties of materials like ceramics change drastically at high temperatures [10,11], which greatly affects the steady-state temperature. The absorption properties of a material need to be known to accurately predict steady-state temperatures. The second problem is the difficultly in constructing a high-power microwave applicator. It is in the microwave heating behavior of a poor microwave absorber that a quasi-steady temperature is likely to be observed. However, it is difficult to heat poor absorbers using purely microwaves.

In the present study, we experimentally demonstrate that the microwave heating behavior of high-power microwave applicators can be predicted by considering the steady-state energy

* Corresponding author. Tel.: +81 774 3601.
E-mail address: kashimura@rish.kyoto-u.ac.jp (K. Kashimura).

0255-2701/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.cep.2013.12.001

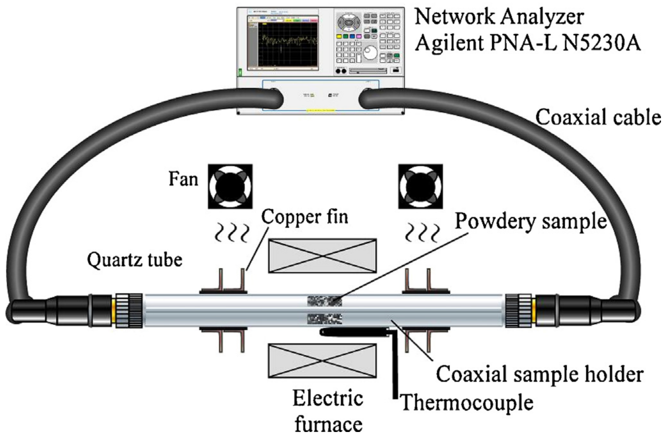

Fig. 1. Setup used to measure the relative permittivity. The sample holder is made of SUS316L stainless steel and is 300 mm long. An R-type thermocouple was attached to the sample holder using Pt wire. The apparent temperature (*Tapp*) varies almost linearly with the true temperature (*Ttrue*) according to $Tapp = Ttrue \times 1.0313 + 43.237$. This equation was used to correct the temperature for all data.

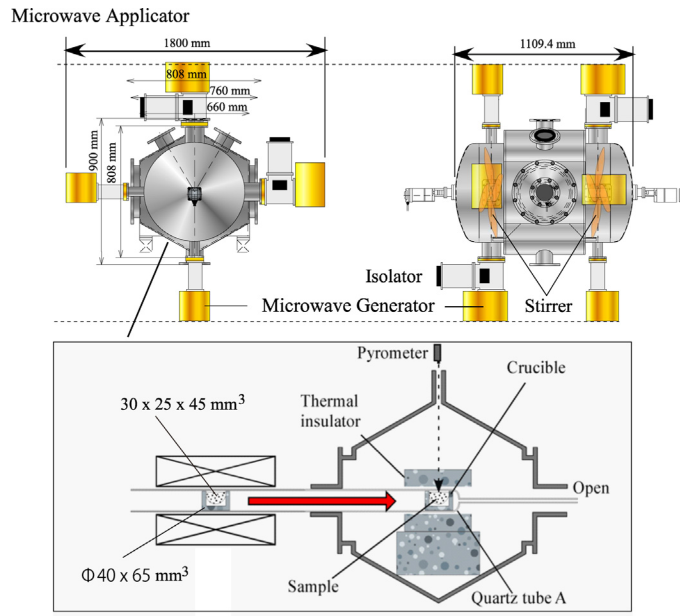

Fig. 2. Schematic diagram of the microwave applicator. The magnetrons generate 2.45 GHz microwaves with a total output power of 12 kW (1.5 × 8 kW). The applicator chamber in the shape of a hexagonal cylinder was closed by two half spheres; this shape promotes focusing of the microwaves onto the sample. In this experiment, 3 kW microwaves were applied to the sample. The samples, which were in a reaction room, were separated from the heating system using quartz tubes (quartz A, I.D.: 46 mm) and the temperature of the chamber was maintained by thermal insulation around the tubes.

balance. We first investigate the temperature dependence of the microwave absorption properties of hematite at frequencies in the range 1–13.5 GHz (hematite is considered to be a poor microwave absorber; we selected hematite to observe the quasi-steady-state temperature). Using a system whose reaction system is separate from the heating system, we found that the heating behavior of hematite varies significantly with its initial temperature.

2. Experimental procedure

2.1. Absorption properties of hematite

This study used hematite powder consisting of 0.5-μm-diameter α-Fe_2O_3. The real and imaginary parts of the relative permittivity (ϵ'_r and ϵ''_r) were measured over the temperature range 25–1000 °C by the coaxial transmission line method using a network analyzer (Agilent Technologies, N5230A), coaxial cables, and an APC7 coaxial sample holder over a microwave frequency range of 1–13.5 GHz (see Fig. 1).Using an electric resistance furnace, the temperature was increased at a rate of 10 °C/min during measurements. The transmission and reflection (*S* parameters) of the irradiated microwaves were measured about every 50 °C during the heating cycle. The network analyzer had an output power of ca. 1 mW. Each measurement was performed over a frequency range of 1–13.5 GHz and took about 2 s. The complex permittivity was calculated from the *S* parameters using the algorithm of the NIST precision method [12].

The compressions were applied to both sides of the powder and the compacts were fixed to a volume of $\phi 3.04 \times 7\ mm^3$ in this measurement. Because hematite particles have a diameter of 0.5 μm and a weight of 0.875 g, this device made compacts with a constant relative density of 1.75 g/cm^3.

In this experiment, the sample was heated using an electric resistance furnace. The heat was generated in an isolated furnace, meaning there was some latency in terms of the heat reaching the center of the coaxial chamber. The temperature was measured by a thermocouple attached to the outer wall of the holder instead of a thermocouple embedded in the sample (the heating rate was 10 °C/min). The measured temperatures were hence indicated temperatures, so calibration was necessary to obtain the true temperatures. To calibrate the outer thermocouple, the apparent and true temperatures were measured simultaneously during a heating cycle using an additional thermocouple embedded in the sample.

2.2. Heating system

Fig. 2 shows a schematic diagram of the microwave applicator used. This applicator has a 2.45-GHz magnetron oscillator (×8), an isolator, WRJ-2 waveguides, stirrers, and a furnace body. The magnetrons generate 2.45 GHz microwaves at a total output power of 12 kW (1.5 × 8 kW). The body consists of a hexagonal cylinder and a half sphere (×2); these shapes focus microwaves onto the sample. In this system, the reaction system is separate from the heating system. The sample was preheated by an electric furnace to a sufficiently high temperature and microwaves (3 kW) were focused onto the sample, as shown in Fig. 2. In these experiments, carbon and magnetite powders were also employed as samples for comparison with hematite.

3. Results and discussion

3.1. Absorption properties

Before discussing the quasi-stable temperature of the steady state during microwave heating, we must understand the absorption properties. Fig. 3(a)–(e) shows plots of the real and imaginary parts of the relative permittivity (ϵ'_r and ϵ''_r) of α-Fe_2O_3 powders (particle size: 0.5 μm; relative density: 1.75 g/cm^3) as a function of temperature for frequencies of 1, 2.45, 5, 7.5, 10, and 13.5 GHz, respectively. At all frequencies, ϵ''_r was observed to increase with increasing temperature at temperatures of 800 °C or lower. Since ceramics are highly conductive in this temperature, the permittivity of the α-Fe_2O_3 powder is $\epsilon''_r \approx (\sigma/\omega) \times i$ (where σ is the static electrical conductivity and *i* was the current from the electrical gradient). The increase in ϵ''_r must be due to an increase in the electrical conductivity, considering that there were no chemical reactions in this experiment. Some researchers reported anomalous behaviors of chemical reactions even though the phase diagram indicates that hematite is stable at high oxygen partial pressures. We did consider