Microscopic and Spectroscopic Observations of Plasma Generation in the Microwave Heating of Powder Material

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(Received 6 May 2010 / Accepted 5 October 2010)

Structure formation during the plasma ignition was experimentally investigated under microwave irradiation of magnetite (Fe_3O_4) and graphite composite powder. It was found from microscopic images that there are stepwise structure formations: (i) a luminous layer, (ii) hotspots, (iii) flare like emissions, and (iv) a luminous body above the powder. The 1D-spatially resolved emission spectra show that the light of the luminous layer of (i) has CO bands, and the luminous body of (iv) has iron atomic spectra superimposed on the continuum spectrum. The structure formation indicates a process of plasma-material mixing through chemical reduction of the magnetite powder.

Keywords: structure formation, plasma-material interaction, microwave heating, emission spectrum, microscopic image

DOI: 10.1585/pfr.5.041

Microwave heating of material will attract more attention as a low-carbon heating technology, if electric power for microwave generation is provided by low-carbon energy sources such as nuclear, solar, and bio energies. The strong microwave irradiation of the material not only heats the material but also often induces the breakdown of gases. In controlling the breakdown or utilizing the breakdown plasma, knowledge of the plasma generation is crucially important.

In microwave processing oxide-powder materials are commonly used [1]. Their huge total surface area makes them reactive; thus, chemical interaction between the powder and plasma would be prominent in the plasma generation. The powder material intended for this study is a magnetite (Fe_3O_4)-graphite composite. This material is for the *microwave iron making* where the microwave is used for the energy required in the endothermic reduction reaction of magnetite, i.e., $Fe_3O_4 + 2C \rightarrow 3Fe + 2CO_2 - 0.32$ MJ/mol [2]. The plasma generation during the microwave iron making, however, is still only briefly explained by a local charging on the material that results in the arcing [3]. This article presents the microscopic images and emission spectra showing the stepwise structure formation in the plasma generation.

Figure 1 shows schematically the experimental setup. The shape of the composite powder material (specimen) is the cylindrical rod of 8-mm in diameter and 10-mm in length. Its weight was 0.619 g, and the weight ratio of the magnetite to graphite was $M_{Fe3O4} : M_C = 90 : 10$ which is equivalent to the molar ratio for the chemical formula as mentioned above. The specimen was located at the anti-node of the electric-field standing wave in the single mode cavity (TE_{103}, cw-2.45 GHz); thus, the field acting on the material is almost the ac electric field. The specimen is covered with a quartz tube filled with helium gas at atmospheric pressure. The observation was performed with the Microscopic Imaging System (type: MISM-08 made by Mutsumi Corporation) that consists of a microscope and a Czerny-Turner type imaging spectrometer, sharing the same incident axis [4]. The imagining spectrometer enables us to obtain visible spectra resolved 1D-spatially along the spectrometer-slit image located in the microscope field of view. As shown in Fig. 1 specimen temperatures were measured with IR pyrometers, and some gas analyses are performed.

Figure 2 shows the sequential microscopic images at the plasma ignition. The field of view includes the upper circular arc of the cylindrical specimen. The time $t = 0.0$ s is defined as the time when the thin glow layer emerged along the circular surface. The plasma ignition inciden-

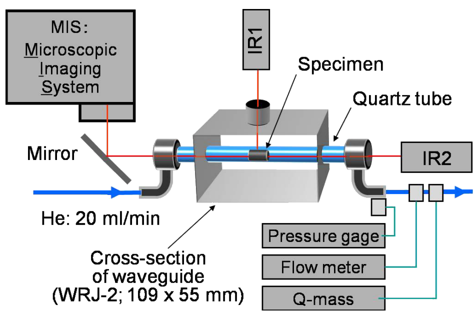

Fig. 1 Schematic diagram of the experimental setup.

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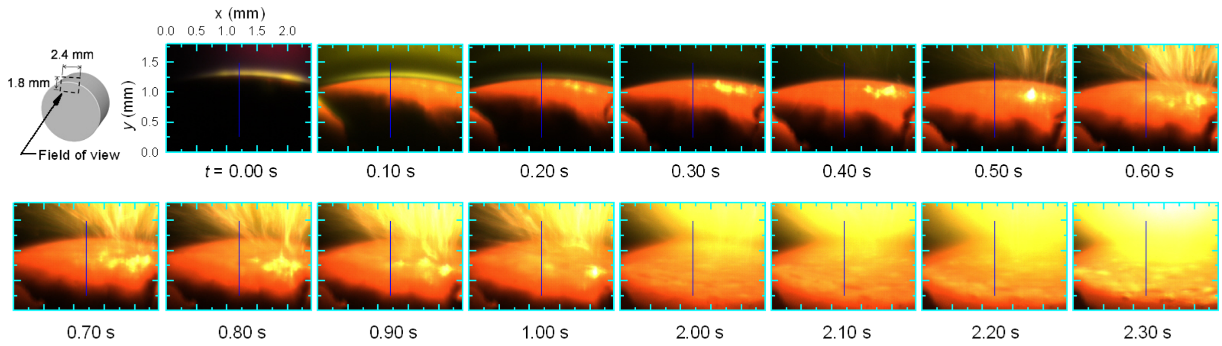

Fig. 2 Sequential microscopic images in the ignition of an atmospheric plasma on magnetite-graphite composite powder material. There can be four structural steps: (i) a luminous layer, (ii) hotspots, (iii) flare like emissions, and (iv) a luminous body above the material. Each vertical line indicates the spectrometer-slit image for the 1D-spatially resolved spectra shown in Fig. 3.

tally started at 195 s after the beginning of the microwave irradiation. The upper surface temperature was 512°C just before the plasma ignition.

As shown in Fig. 2 the structure formation has four steps: (i) a luminous layer, (ii) hotspots, (iii) flare like emissions, and (iv) a luminous body. The luminous layer of (i) fades away by $t = 0.3$ s, but hot spots of (ii) are gradually generated on the surface of the specimen from $t = 0.1$ s, and then flare like emissions of (iii) appear above the specimen from $t = 0.4$ s. Those hot spots as well as flare like emissions increase in number, and finally they grow up to be a luminous body of (iv) for $t \geq 0.9$ s. The boundary between the luminous body and the specimen is not clear, suggesting formation of a plasma-powder mixture. Note that the quadrupole mass analyzer for the exhaust gas showed that components of both CO and CO_2 arise after the ignition, implying progress of the chemical reduction.

The 1D-spatially resolved spectra corresponding to Fig. 2 are shown in Fig. 3. In the first half of the ignition (see the bottom image of Fig. 3 (a)) the thermal spectrum is prominent in the upper part of the material, indicating the temperature of 1480°C at $y = 1.2$ mm. On the other hand, several peaks are significant in $y = 1.26$-1.37 mm of which the region is coincident with that of the luminous layer as shown in Fig. 2. Some of those peaks, such as 439 nm, 451 nm, 519 nm, and 561 nm, with arrows shown in the upper frame of Fig. 3 (a) can be attributed to band spectra of CO ($B^1\Sigma$ - $A^1\Pi$), which are degraded to the shorter wavelength. Three sharp peaks at 468 nm, 472 nm, and 481 nm are assigned as atomic spectra of Zn I. The datasheet of the magnetite powder shows that the zinc makes up 0.03 wt% of it. In the last half of the ignition (see Fig. 3 (b)), new peaks with arrows at 438 nm, 527 nm, and 533 nm are superimposed on the continuum for the luminous body above the material. Those peaks can be assigned as the spectra of Fe I, suggesting inclusion of iron particles from the material.

Those stepwise structure formations can be characterized by the process of plasma-material mixing through the chemical reduction of powder magnetite. The spectra of CO and surface temperature over 1400°C imply that a layer of molten iron (with a few-wt% carbon) is produced on the powder material by the chemical reduction in the first half of the ignition. It is possible that the layer of the molten iron forms into a protrusion by its cohesion. The protrusion enhances the local electric field as well as the energy flux from the plasma, and thus hotspots arise. Unstable development of the hotspot leads its collapse, and then fragments of the protrusion flow into the plasma, as shown as flare like emissions. Finally, a luminous body containing iron particles is produced.

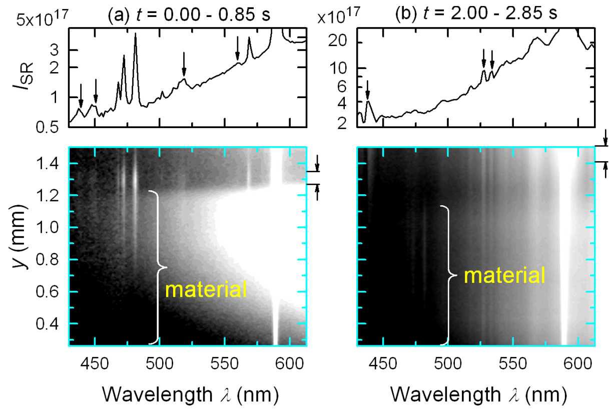

Fig. 3 Emission spectra obtained in different periods during the plasma ignition. The position y corresponds to that of Fig. 2. The unit of ISR is photons m^{-2} s^{-1} nm^{-1} sr^{-1}. The spectrum in each upper frame is the spatially averaged spectrum in the region indicated by the arrow at the right axis of the bottom frame. The strong peak at 590 nm is Na I.

This research was supported by a Grant-in-Aid for Scientific Research on Priority Areas 465 of MEXT.

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