Formation of Nano-Domains by Microscopic Thermal Non-Equilibrium Generated in GHz High Frequency Microwave Field

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ABSTRACT
Recently, a new method to form nano-structures consisting of small magnetic domains of 5~20 nanometers with random orientations has been developed when some ferromagnetic materials are exposed to the magnetic field at microwave frequencies.

INTRODUCTION
Following to the astonishing reports of microwave sintering of powder metals by R.Roy and D.Agrawal¹, extraordinary behaviors on microwave in the E & H field² were reviled by the authors. The cooperation with NIFS Japan and Penn State University found the microscopically thermal non-equilibrium at E and H field node. These systematic investigation namely, (1) the in-situ observations during heating at the optical microscope level (10~100μm), (2) the ex-situ scanning electron microscopic (SEM) observation at the level of 1~10 μm, and finally (3) the transmission electron microscopy (TEM) analysis to nano meters level. The series of experiments have revealed the formation of nano-domains spreading uniformly throughout the material, under the microscopically thermally non-equilibrium state.

EXPERIMENTS
The microwave H and E fields selectively radiated the testing material (sample) that are set either in the H field or E field maximum in the TE103 mode in 2.45 GHz rectangular cavity. The complex system of two-dimensional visible light spectroscopy and optical microscope made the state-of-art measurement tool to investigate non-thermal equilibrium states under microwaves as illustrated in fig.1.

![Diagram of experimental setup](image)

Fig.1 Single mode cavity and in-situ observation system for micron scale hot spots
**Creation of Hot Sops by Microwave Exposures** (3)(4)

The testing materials pressed to a small cylindrical pellet with 6 mm diameter and 3 mm height. The infrared pyrometer detects average temperatures, and the waveguide directional coupler monitors the forward, reflected and net powers. Figure-2 shows the temporal evolutions of hot spots in the sample consisting of hematite Fe₂O₃ and magnetite Fe₃O₄ in the H-field maximum. Figure 3 shows the drifting of the hot spots. Within seconds of microwave application, as illustrated in fig.2, many “local hot cores” in the order of 100μm have generated huge temperature gradients of 200~400 degrees Celsius, hence a few thousands of degrees Celsius per mm were maintained for a significant period of time. The hematite particles did not get heated directly by microwave. The visible light spectroscopy observed only the black body emission. It suggests that the high temperature plasma is not generated to cause the heating of the material in the process. Fitting the intensity ratios at different wavelengths in 700~740nm to the black body emissions profiles, localized temperatures are estimated in two-dimensional images overlapping to the video images. The brightest (white area in the video images ) is 1300 degree Celsius and the dark area was almost 900 degree Celsius. The infrared pyrometer indicated average temperature of 1100 degree Celsius in the 2 mm detecting area. These hot spots appeared only in H-field node. In the E-field node, the strong visible light emission appeared over the surface of the sample with separated atomic line spectrums. The intensities of the emissions were by 2~3 orders of magnitude higher than that of black body.

![Fig.2 Observation of Hot Spots by Selective heating to Magnetite (stronger microwave absorbing material) and Hematite (weaker Microwave absorbing) Left picture is visible light video image and The right is isothermal contours estimated by the Planck Equation using the visible light spectroscopy. Small hot spots created in the magnetite powder.](image)

**Creation of Nano-domains**

The ultimate structures at nano-scales were observed by TEM forming nano-glasses consisting of small magnetic domains of 5~20 nanometers with random orientation of the axis (fig.3).

The fine particles of Fe₃O₄ with sizes of 0.2-0.5 μm heated in the selected microwave H or E field were sliced by 60-70 nm thick with a microtome ( LEICA ULTRACUT UCT) by dispersing in epoxy-resin plates. The high resolution transmission electron microscope (TEM; a JEOL JEM-3200) observed the images of the original Fe₃O₄ powder and those excited in the selected E field and H field. Since the sliced original single crystal has flat and homogeneous surfaces, one can see the well ordered lattice patterns over the whole crystal. The sample heated in the H field
node (maximum E field) shows the uneven structure of the sample. The sample heated in the maximum H field exhibits the presence of nano-crystalline domains of which lattice directions are random. The domain sizes of this sample are in the range of 5-15 nm (approximate average size is 10 nm). The randomness of the lattice orientation indicates that the particle-particle magnetic interaction is negligible in the solidification process. Each magnetic domain can be highly excited by application of the oscillating magnetic field and cohesive rotation of the domains can be induced synchronously with the oscillating field.

![Image](image_url)

Fig.3 Continuous Nano-structure created by microwave irradiation to Fe₃O₄ Powder

**CONCLUDING REMARKS**

It is very interesting that the temperature increase in the sample is accelerated at a temperature close to the Curie point and never raises above 1100 degree Calculus (but it is below the melting point of 1580 degree Calculus). Thus this high-temperature state never liquefies the sample but maintains the temperature by the continuously applied magnetic field. The applied field energy could be spent for the rotational of the magnetic domains with negligible inter-domain interaction at the temperatures higher than the Curie point. The sudden removal of the field at 1200 degree Calculus is expected to cause the rapid deceleration of the domain rotations that causes the binding of the domains. However, the temperature of 1200 degree Calculus K is so higher than the Curie
temperature that one cannot expect any strong force for the lattice alignment of the nano-domains. On the other hand, the heating in the maximum E field did not show any Curie point effect and the temperature increases monotonically up to 1200 degree Calculus. We could not expect the rotational excitation of magnetic domains so that the cooling the sample should produce larger micro-crystals as observed in the SEM and TEM images.

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