Microwave energy absorption driven by dynamic structural and magnetization states in $\text{Fe}_{85}\text{B}_{15}$ metallic glass ribbons

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The kinetics of the microwave crystallization of $\text{Fe}_{85}\text{B}_{15}$ metallic glasses was investigated *in situ* using the time-resolved x-ray diffraction method. It is shown that the recorded thermal profile during the microwave exposure of the ribbons bears a close relationship with the dynamic magnetization state during the decomposition of the amorphous phase into nanocrystalline α -Fe and Fe_3B phases. © 2009 American Institute of Physics. [doi:10.1063/1.3257697]

The interest in the microwave processing of metallic materials steadily increased since the first report by Walkiewicz *et al.*¹ The state of the art in this field was recently reviewed.² The microwave heating of metals relies on Joule heating and magnetic induction effects.^{2,3} The mechanism of microwave absorption in metals at high temperatures is less understood. For the electromagnetic heating of particulate metals several models were already proposed.^{3–8} Their objective is to predict the thermal evolution of the specimen during microwave exposure as monitored by the surface temperature measured with optical pyrometers.

At macroscopic sizes (several millimeters), the size and shape distributions of the powder bead are neglected and microwave heating is treated following the classical electromagnetic theory. The experimental heating rates are retrieved with various degrees of success.^{3,6,8} In some cases, the natural oxide layers that assist in initiating absorption are also accounted for via an effective medium approximation.^{7,8} At intermediate length scales comparable with the particle size (typically in the micron range), microwave losses vary significantly with the field penetration depth δ as well as with the geometry and dimensions of the microwave applicator. The relative size of each particle with respect to the skin depth determines whether its heating is driven by an electric E-field or magnetic H-field component locally acting at the particle site.^{4,5} Finally, at the submicron and nanometer scales, neither structural transformations nor changes in the granularity of the microstructure are taken into account. Materials are either considered homogeneous or effective medium approximations are used to define appropriate macroscopic average values for the physical properties relevant to microwave absorption. In this context, gaining insight into the heterogeneous nature of materials and their evolution during microwave irradiation is certainly of interest. *In situ* time-resolved experiments in either mixed or separate E and H fields can readily follow the kinetics of phase transformations and the microstructure development,^{9–11} to support modeling of microwave heating for heterogeneous solids.

We report on the microwave crystallization of amorphous $\text{Fe}_{85}\text{B}_{15}$ ribbons examined *in situ* by the time-resolved x-ray diffraction (TRXD) method. Iron-boron alloys were

thoroughly investigated in the past as model materials for multicomponent Finemet alloys. The ribbons were generously provided by VAC (Hanau, Germany). The synchrotron experiments were performed at the MSX04SA beamline at the Swiss Light Source. Diffraction patterns were acquired each second within a wide scattering angle range using the new Mythen II detector. The experiments were done in transmission using monochromatic radiation ($\lambda = 0.056$ nm). The amorphous $\text{Fe}_{85}\text{B}_{15}$ ribbons (30 μm thick) were mounted in sapphire capillaries and placed in the microwave resonant cavity so as to experience a mixed E/H field. For heating we used a 2.45 GHz single-mode applicator.⁹ The temperature was detected using an optical fiber thermometer.

The pyrometer temperature profile recorded during heating at constant microwave power (240 W) is shown in Fig. 1. The initial tuning of the resonant cavity is interrupted at the onset of heating (A). Between time A (onset, $t = 90$ s) and G (switch off, $t = 316$ s) heating is due only to microwave absorption by the sample. The x-ray pattern after treatment (Fig. 2) indicates the presence of the body-centered cubic bcc α -Fe (cI2, S.G. 229, PDF 3–65–4899) and of the body-centered tetragonal bct- Fe_3B (tI32, S.G. 82, PDF 39–1316) phases. It was previously reported that upon nucleation the Fe_3B phase forms eutectic spherulites nearly equal in size in which aggregates of acicular Fe nanocrystals are embedded.^{12,13} The Fe_3B crystallites rapidly reach a stable

FIG. 1. Temperature profile recorded during the *in situ* microwave heating of amorphous $\text{Fe}_{85}\text{B}_{15}$ alloy ribbon.

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FIG. 2. High-resolution XRD pattern collected after the microwave irradiation of $\text{Fe}_{85}\text{B}_{15}$ ribbon, showing the formation of α -Fe and Fe_3B phases.

size around 50 nm, as estimated from the width of the (321) peak using the Scherrer formula (Fig. 3). The α -Fe growth kinetics during microwave heating was obtained from the line-broadening analysis of the (110) Bragg peak (Fig. 4). The α -Fe grains tend to stabilize around a final size of 46 nm. The grain-growth kinetics is well described by the retarding effect of a size-dependent pinning force,¹⁴

$$G(t) = [G_m^2 - (G_m^2 - G_0^2) \cdot \exp(-kt)]^{1/2}, \quad (1)$$

where G is the crystallite size, G_m is the final maximum value, G_0 is the extrapolated grain size at $t=0$ and k the rate constant. A least-squares fit to the data in Fig. 4 yields $G_0 = 24.1 \pm 0.9$ nm, $G_m = 46.2 \pm 0.6$ nm, and $k = (4.86 \pm 0.46) \times 10^{-2} \text{ s}^{-1}$. The microstructural coarsening is therefore limited by the fixed size of the Fe_3B spherulites.¹³

The magnetic properties of the alloy drastically change during crystallization, which affects the efficiency of microwave energy dissipation. The magnetic energy losses arising during microwave exposure are caused by a time lag of the

FIG. 3. TRXRD patterns of the amorphous $\text{Fe}_{85}\text{B}_{15}$ alloy showing the nanocrystallization sequence during microwave heating.

FIG. 4. Grain-growth kinetics of the bcc α -Fe phase during microwave heating.

magnetization relative to the external magnetic field. The magnetization changes are either rotational or due to domain wall motion. The input microwave energy is consumed in the excitation of spin waves¹⁵ and in spin reorientation across domain walls.¹⁶ The energy losses are proportional to the magnetic energy product (area of the hysteresis loops) so that the coercivity and saturation magnetization control the efficiency of magnetic microwave heating of the material.

A steep thermal peak (ABC) is first noticed at the onset of microwave heating (Fig. 1). Its origin lies in the abrupt change of the magnetoresistance upon the resonant coupling to the microwave field.¹⁷ It is also reminiscent of resistance bumps observed in current annealing experiments.¹⁸ During this initial temperature rise due to Ohmic heating and the ferromagnetic resonance of the newly formed phases, the magnetic field penetration depth is also greatly enhanced. The favorable evolution of the resistivity and permeability with increasing temperature leads to a much higher magnetic skin depth. Further heating (CD) is caused by magnetic hardening: the coercivity increases due to the formation of the α -Fe and Fe_3B crystals.¹⁹ As coarsening proceeds (DE), the grain sizes G become larger than the domain wall thickness d and the coercivity decreases at a slower rate, as predicted by the random anisotropy model.²⁰ Pinning of the domain walls is most efficient (maximum coercivity) when $G \approx d$. This condition marks the gradual transition from a single- to multiple-domain magnetization regime.²¹ Beyond this point the hysteresis losses decrease due to magnetic softening and the sample is slowly cooling (DE). At $t=220$ s, the Curie temperature (513 °C) (Ref. 22) of bcc- Fe_3B is reached and a sudden drop of the temperature is recorded (EF). At the Fe_3B para-to-ferromagnetic transition, both permeability and resistivity change and so the resonant frequency shifts away from the one of the cavity. The sample further cools at a very low rate (FG), since the growth of the α -Fe nanocrystals is nearly completed (Fig. 4).

To conclude, the microwave crystallization of amorphous $\text{Fe}_{85}\text{B}_{15}$ ribbons was studied *in situ* using the TRXRD method. The observation of microstructure development during microwave heating allowed for a straightforward interpretation of the pyrometer temperature signal in terms of the sample magnetization dynamics. The thermomagnetic evolution of the specimen controls the magnitude of microwave energy losses at elevated temperature in strict correlation with the alloy microstructure.

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