In-situ synchrotron x-ray study of MgB$_2$ formation when doped by SiC

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We have studied the evolution of the reaction $xMg + 2B + ySiC \rightarrow zMg_{1-p}(B_{1-q}C_q) + yMg_2Si$ in pressed powder pellet of 1, 2, 5 and 10 wt% SiC doping. We found a coincident formation of MgB$_2$ and Mg$_2$Si, whereas the crystalline part of the SiC nano particles is not reacting at all. Evidence for the incorporation of carbon into the MgB$_2$ phase was established from the decrease of the a-axis lattice parameter upon increasing SiC doping. A lower limit of the grain size of MgB$_2$ was estimated from the width of MgB$_2$(100) and MgB$_2$(002) reflection and the grain size estimate was found to decrease from $L_{100} = 795$ Å and $L_{002} = 337$ Å at 1 wt% SiC to $L_{100} = 227$ Å and $L_{002} = 60$ Å at 10 wt% SiC. Thus superconductivity might be suppressed at 10 wt% SiC doping due to the grain size approaching the coherence length [1].

The in-situ experiments were performed at the high energy beamline BW5 using $E = 77$ keV photons and a furnace holding a steel sample stick inside a quartz tube containing a flow of argon. The powder pellets with Ø = 2 mm and 1 mm thickness were wrapped in a 0.05 mm Fe foil to avoid evaporation of the Mg during the experiment. A lead ring was inserted in front of the MAR345 image plate detector in order to suppress the Fe(101) reflection by 3 orders of magnitude and thereby increase the possible exposure time [2]. Two dimensional diffraction patterns from the powder pellets were collected by exposing the image plate in 50 seconds during a heat treatment. The images were integrated into intensity versus d-spacing using the fit2d program and the sample-detector distance was obtained from the diffraction pattern of a piece of Al$_2$O$_3$ inserted in the furnace. Normalization by the transmitted direct beam and subtraction of a Fe foil background was done using MATLAB. The time evolution of the powder diffraction pattern is shown on figure 1, where the characteristic reflections of Mg and the crystalline part of the primarily amorphous boron is seen in the initial pattern, whereas the MgB$_2$, SiC and Mg$_2$Si is observed at the end of the heat treatment. Figure 2a shows the normalized integrated intensity of selected reflection during the heat treatment. It is seen that the formation of both MgB$_2$ and Mg$_2$Si starts well below the melting point of Mg at $T_{Mg} = 649$ °C, however the intensity of SiC is almost constant during the heat treatment since the peak at $t = 38$ minutes is an artifact of fitting of overlapping reflections. Thus it is concluded that only the amorphous part of the SiC is reacting and forms Mg$_2$Si. The remaining carbon from the SiC reacting to form Mg$_2$Si is incorporated into the MgB$_2$, which is seen by a decrease of the a-axis lattice spacing determined from the fitted peak position of the MgB$_2$(100) reflection. The $\Delta \alpha = -0.0055$ for the 10 wt% SiC doping as shown on figure 2b correspond to a carbon doping of $q = 0.06$ and no change of the c-axis lattice spacing is expected [3]. A broadening of the MgB$_2$(100) and MgB$_2$(002) reflections can be interpreted as an in-homogenous carbon doping or as a finite size effect due to a reduced grain size. By assuming the latter one can estimate a lower limit to the grain size from the Scherrer equation after subtracting the instrumental resolution determined from the diffraction pattern of Al$_2$O$_3$ used for calibration. This is shown on the right hand axis of figure 2b and a decrease of the grain size is seen by increasing SiC doping. Measurements of the critical current density $J_C$ of SiC doped MgB$_2$ shows that 5 wt% SiC doping is the optimal [4]. The decrease of $J_C$ at high doping levels might be caused by the grain size approaching the superconducting coherence length. Transmission electron microscopy examinations of the samples are planed to test this hypothesis.
Figure 1: Time evolution of the diffraction pattern of Mg + 2B + 10 wt% SiC powder pellet wrapped in Fe foil during a heat treatment where the sample is heated to $T = 700^\circ$ C as specified by the right hand figure.

Figure 2: a: Integrated intensity of selected diffraction peaks during a temperature history shown by the dashed line for the 10 wt% SiC doped sample. b: Relative change of the a-axis and c-axis unit vector as function of the SiC doping. The a-axis contraction of $\Delta a = -0.0055$ correspond to a carbon doping of $q = 0.06$. A lower estimate of the MgB$_2$ grain size as determined from the peak width of the MgB$_2$(100) and MgB$_2$(002) is plotted on the right hand axis and indicate that the MgB$_2$ grain size approached the coherence length of the superconductor at high doping levels.

References