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In-situ neutron diffraction study of a heating treatment for Nb_3Sn ITER superconducting wires.

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Abstract. In-situ powder neutron diffraction studies have been performed during the heating treatment of a set of Nb_3Sn wires obtained by the internal tin process. The different phases appearing during the process have been evidenced. A strong reorientation of the deformed copper and niobium metals induces an orientation on the Nb_3Sn formed. Among the phases formed, the δ-CuSn phase is decomposed at 890K to form the γ-CuSn and Nb_3Sn phases. The kinetics of these phases is in agreement with a diffusion-growth mechanism.

1. Introduction

The heat treatment (HT) of internal tin process to form Nb_3Sn composites requires the mixing of Sn from the filament bundle core with the interfilamentary Cu in order for each Nb filament to be adequately surrounded by Sn prior to Nb_3Sn formation. Due to an extremely slow kinetics, the best mixing heat treatment is complex and varies strongly by composite geometry, composition and/or manufacturer [1]. In order to achieve the best A15 Nb_3Sn with the higher superconducting properties it is important to know the formation pathway between Sn, Cu and Nb compounds. Most of the studies were performed on samples reacted at various temperatures and times, then quenched to analyze the results [2-8]. In-situ powder neutron study is a powerful technique that allows recording diffraction patterns of all the volume of the sample under HT and atmosphere suitable for synthesis.

2. Experimental

Multifilament wires have been prepared by the internal tin method and are composed by 18 sub wires with tin reservoirs containing 2% of titanium. The outer layer is composed of tantalum and the inner core of copper. For the neutron experiment, 40 wires of 0.815mm have been cut at a 60 mm length; the ends of the wires have been crimped on 3 mm. The bundle of wires has been introduced parallel in a quartz tube of 12 mm of diameter, and during all the neutron experiment an argon atmosphere has been circulating in the tube, with a bubbling apparatus.

Neutron experiments have been performed for 44 hours with the following heating treatment:

RT (298 K) ----- 2h----→ 733 K -----20h----→ 933 K ----22h-----→ 933 K. This HT was chosen because neutron time is very expensive and given for a maximum of 4 or 5 days for a diffraction experiment. The suggested HT [1] being too long (275h) to be fully realized under neutron beam. In another experiment, we have prepared ex-situ samples, heated for 275 hours, with temperature dwells at 210°C (25h), 350°C (50h) and different times of exposure at high temperature (up to 725°C and 200 h). These results will be presented later and are in agreement with the observations of this experiment. Neutron diffraction patterns have been recorded every 5 minutes on the D1B diffractometer of the

ILL-Grenoble, at a wavelength of 2.524Å in a dedicated furnace. Refinements have been done by the sequential Rietveld technique using the Fullprof program.

3. Results

Neutron diffraction patterns recorded during heating and temperature dwell are reported in Fig. 1 and 2 as three-dimensional plots. The Sn, Cu et Nb(Ta) phases are the initial phases observed, on heating the first modification is the reorientation of the copper phase with a new preferred orientation along the {001} direction with a cubic texture. After melting of the Sn phase, the ε-CuSn phase is formed at 658 K, the second phase appearing at 738 K is the δ-CuSn phase. The ε-CuSn phase disappears at 854 K. After reaching a maximum, the δ-CuSn phase intensity starts to decrease and the γ-CuSn and Nb_3Sn phases appear at 858 K and start to grow.

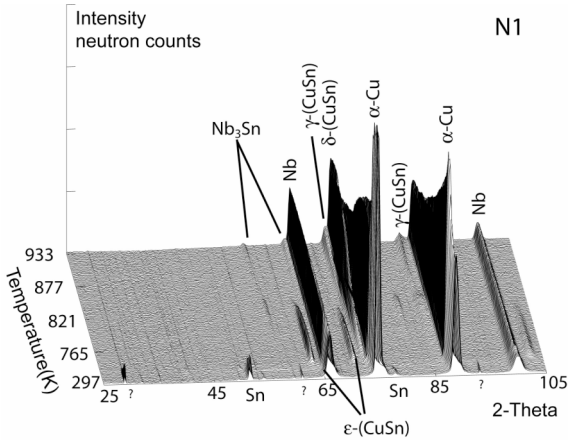

Figure 1: 3D plot of the neutron powder patterns during the heating ramp.

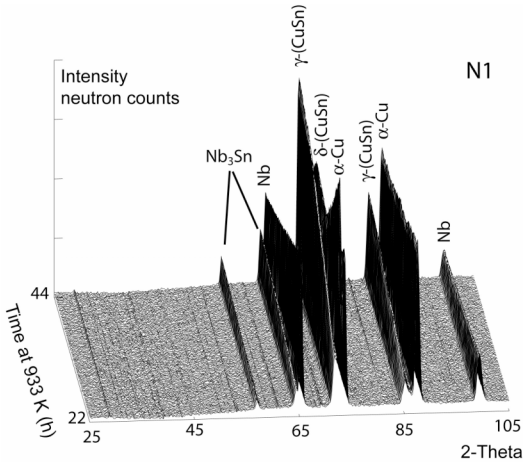

Figure 2: 3D plot of the neutron powder patterns during the annealing dwell.

We have refined the structure of the phases present with the following parameters. The Cu and α-Cu phases, space group (SG) Fm3m, a=3.618Å, preferred orientation [100]. The Nb and Ta phases, SG=Im3m, a=3.300Å, preferred orientation [100]. The γ-CuSn phase as Cu_3Sn, SG=Fm3m, a=3.669Å. The δ-CuSn phase as $Cu_{41}Sn_{11}$, SG=F-43m, a=18.148Å. The Nb_3Sn phase, SG=Pm3n, a=5.293Å, preferred orientation [100]. For each phase, only the scale factor (SF), the cell parameter a (all phases being cubic) and the preferred orientation (PO) have been fitted. Depending on the temperature, 2, 3, 4 or 5 phases have been used to refine the whole pattern. The fig. 3 is the Rietveld refinement result of the sample at the end of the annealing dwell (5 phases). Then cyclic refinements have been performed at each temperature and the results plotted in Figs. 4 to 8 for the different compounds as a function of time. The temperature profile as a function of time is also reported. In fig. 4, the a cell parameter of α-Cu follows the temperature profile, but the solution of Sn in the Cu lattice certainly increases the lattice parameter in the same time. The SF shows an evolution at point A = 353 K of the curve linked to the reorientation towards the cubic texture. The SF is very correlated with the preferred orientation and only the variation is signifiant. Then the SF presents an important decrease linked to the formation of the ε-CuSn and δ-CuSn phases. On the dwell temperature the SF remains constant. In Fig. 5, the a cell parameter of the Nb(Ta) is plotted versus time, the cell expansion follows the profile temperature. The SF increase up to the B point=858 K is correlated to Debye-Waller thermal variations, then the decrease of the SF is linked to the appearing of the Nb_3Sn phase. In Fig.6, the variation of the a and SF parameters present different features. At point C=738 K the phase appears, at point D=823 K, both parameters a and SF have a big change, not related to another phase, so it could be a phase transition due to the δ-CuSn phase. At point E=890 K both parameters have again a big change, in particular the

SF starts to decrease continuously, this change can be related to the formation of the γ-CuSn and Nb_3Sn phases. When the reaction is completed (full Nb_3Sn transformation), this phase no more exists. In Fig. 7 and 8 the γ-CuSn and Nb_3Sn show an increase of the SF starting at the same time (point F and I=858 K) with the same shape of the SF.

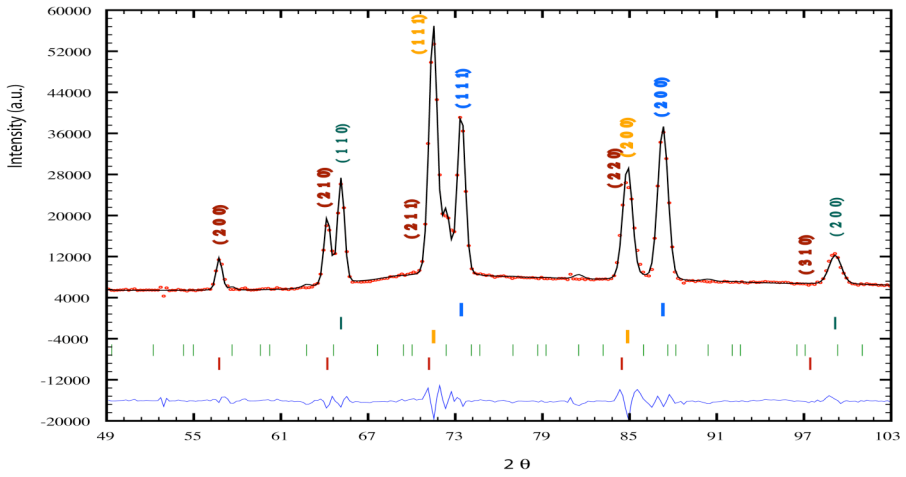

Figure 3: Rietveld refinement of the sample at the end of the annealing plateau. Ticks are the peaks of the different phases in the descending order: α-Cu, Nb, γ-CuSn, δ-CuSn et Nb_3Sn. The lower plot is the difference between experimental and calculated points.

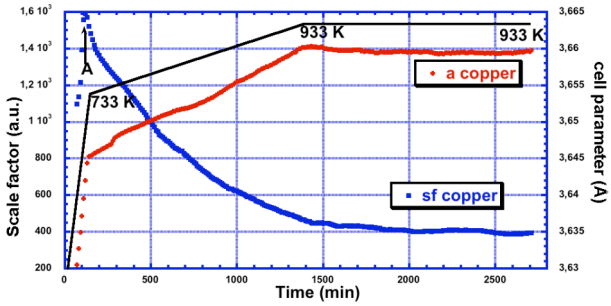

Figure 4: Calculated scale factor and a cell parameters of the Cu, α-Cu phases plotted as a function of the experimental time.

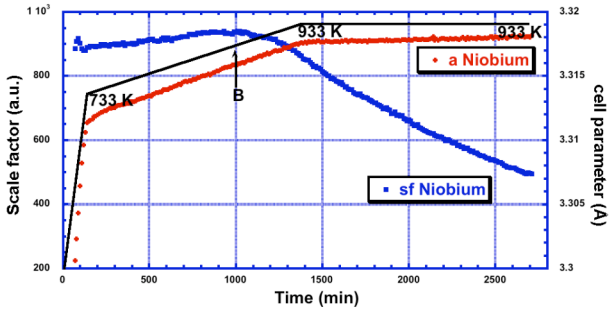

Figure 5: Calculated scale factor and a cell parameters of the Nb(Ta) phases plotted as a function of the experimental time.

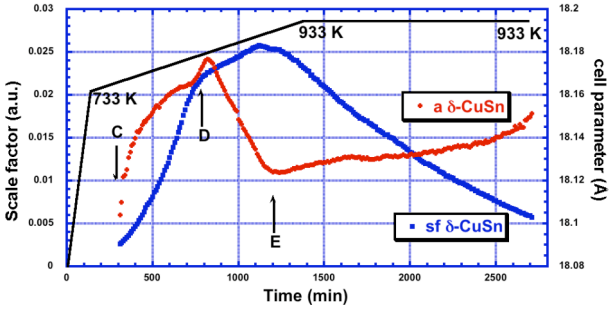

Figure 6: Calculated scale factor and a cell parameters of the δ-CuSn phase plotted as a function of the experimental time.

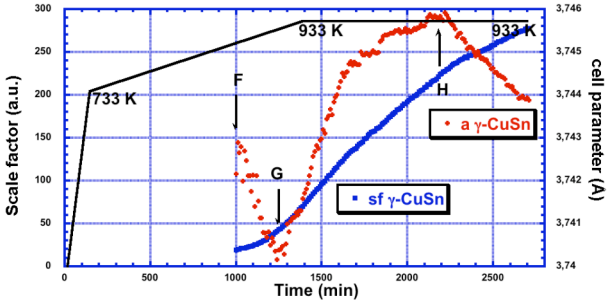

Figure 7: Calculated scale factor and a cell parameters of the γ-CuSn phase plotted as a function of the experimental time.

The a cell parameter of Nb_3Sn, after a small decrease, is stabilized at a value of 5.315Å at 933 K. The a cell parameter of the γ-CuSn phase has a different behavior, decreasing between points F and G

(858 K and 895 K), increasing between points G and H, then decreasing. It is difficult to interpret these parameter changes.

The variation of the SF of the Nb_3Sn is directly proportional to the quantity of phase formed in the bulk of the sample, so we have fitted this variation with time with a logarithmic function whose parameters are reported in Fig.8. This function describes well the variation in the constant temperature range and is in consistence with various models for phase formation by diffusion-growth [9,10].

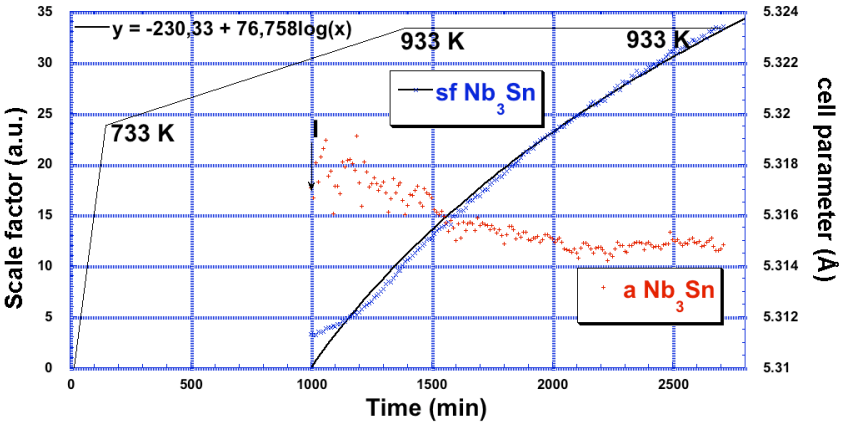

Figure 8: Calculated scale factor and a cell parameters of the Nb_3Sn phase plotted as a function of the experimental time. The line is the calculation of a logarithmic function whose parameters are reported.

4. Conclusion and discussion

We have evidenced by in-situ powder neutron diffraction the different phases formed during the internal tin process for Nb_3Sn synthesis. An important reorientation of copper is observed with a Nb_3Sn also orientated in the same direction. A clue point of transformation is the formation of the δ-CuSn phase that is formed then decomposed to form Nb_3Sn and γ-CuSn.

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