SYNCHROTRON RADIATION TECHNIQUES FOR THE CHARACTERIZATION OF NB_3SN SUPERCONDUCTORS

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Abstract

The high flux of high energy x-rays that can be provided through state-of-the-art high energy synchrotron beam lines has enabled a variety of new experiments with the highly absorbing Nb_3Sn superconductors. We report different experiments with Nb_3Sn strands that have been conducted at the ID15 High Energy Scattering beam line of the European Synchrotron Radiation Facility (ESRF). Synchrotron x-ray diffraction has been used in order to monitor phase transformations during *in-situ* reaction heat treatments prior to Nb_3Sn formation, and to monitor Nb_3Sn growth. Fast synchrotron micro-tomography was applied to study void growth during the reaction heat treatment of Internal Tin strands. The elastic strain in the different phases of fully reacted Nb_3Sn composite conductors can be measured by high resolution x-ray diffraction during *in-situ* tensile tests.

INTRODUCTION

Non-destructive methods like high energy diffraction or micro-tomography can complement the destructive microscopic techniques that are commonly used for the materials characterisation of Nb_3Sn superconductors.

Due to the low x-ray energy of laboratory x-ray sources, diffraction measurements with such sources are done in reflection geometry and are limited to a penetration depth of some tens of µm. Therefore, such experiments can not be used for non-destructive studies of Nb_3Sn strands. In contrast, neutrons or high energy x-rays from synchrotron sources allow *in-situ* diffraction measurements in transmission geometry with the highly absorbing Nb_3Sn strands. The x-ray transmission as a function of x-ray energy that has been calculated for a Ta alloyed Internal Tin (IT) strand with different diameters is shown in Figure 1. For the 1.25 mm-diameter strand, x-ray energies above 70 keV are needed for measurements under optimum conditions.

Here we describe three synchrotron experiments for the materials characterisation of Nb_3Sn strands, notably powder diffraction for phase analysis during *in-situ* reaction heat treatment (HT), fast synchrotron micro-tomography with µm spatial resolution for monitoring void growth during *in-situ* HT, and high resolution diffraction for measuring the 3-D strain state in the different phases of the composite superconductors under *in-situ* tensile loading.

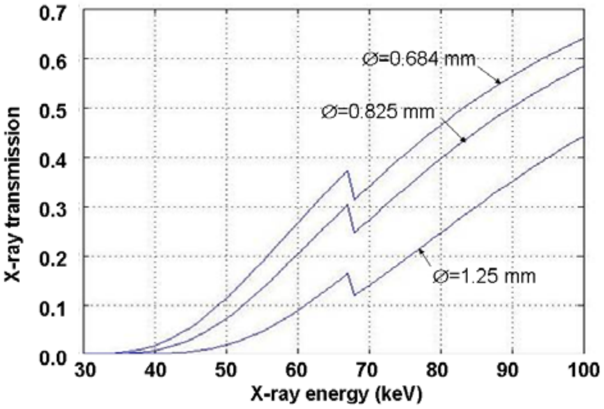

Figure 1: X-ray transmission as a function of x-ray energy for an Internal Tin Nb_3Sn strand with different diameter as a function of x-ray energy. The x-ray transmission for obtaining optimum signal-to-noise ratio in tomography is about 20 %.

SYNCHROTRON X-RAY DIFFRACTION FOR PHASE ANALYSIS DURING IN-SITU REACTION HT

The phase transformations that occur during the reaction HT of Nb_3Sn superconductors prior to the superconducting A15 phase formation can degrade the microstructural and microchemical Nb_3Sn homogeneity and, thus, have a detrimental influence on the critical properties of the fully reacted superconductor. The phases that are formed strongly depend on the strand design.

Powder diffraction measurements in transmission geometry can be used for *in-situ* phase analysis in Nb_3Sn strands. A high flux of high energy x-rays is needed in order to acquire diffractograms with sufficient signal-to-noise-ratio that allows to monitor the growth of weakly diffracting phases formed during the reaction HT, as for instance the orthorhombic $NbSn_2$ and Nb_6Sn_5 [1].

Synchrotron x-ray diffraction measurements were carried out at the ID15B high energy beamline of the ESRF in transmission geometry, using a 87.000 keV monochromatic x-ray beam. Debye Scherrer diffraction pattern were acquired in transmission geometry using a 2-

D detector. A diffraction pattern acquired for a Nb_3Sn PIT strand with a MAR345 image plate detector is shown in Figure 2 before and after radial integration.

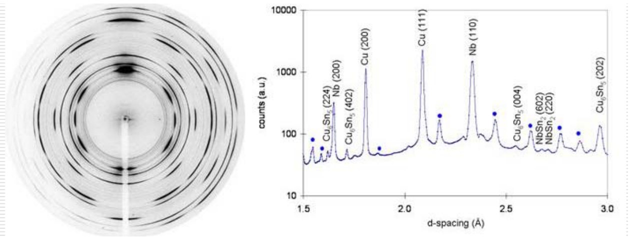

Figure 2: Debye Scherrer diffraction pattern acquired with a MAR345 image plate detector. In the right plot the same pattern is shown after radial integration. The diffractogram of the PIT strand has been acquired at RT subsequent to 490 °C HT. The diffraction peaks of a Cu-Nb-Sn phase are indicated by full dots.

For *in-situ* heating a dedicated furnace built at ID15 that enables an accurate sample temperature control during the diffraction experiments has been added to the experiment. In Figure 3(a) we show a sequence of 85 diffractograms that has been obtained with a PIT strand during *in-situ* reaction HT with a ramp rate of 60 °C h^{-1} and subsequent 4 hours isothermal heating at 675 °C. A semi-quantitative description of the phase transformations is shown in Figure 3(b).

NB_3SN PHASE GROWTH MONITORED BY SYNCHROTRON X-RAY DIFFRACTION

The Nb_3Sn growth kinetics during isothermal HT can be monitored by measuring prominent Nb_3Sn diffraction peak areas acquired during *in-situ* reaction HT. In Figure 4 the Nb_3Sn growth in the PIT B215 strand during 660 °C HT is shown. During the first 20 h diffractograms have been acquired *in-situ* (empty symbols). Longer heat treatments have been realised *ex-situ* (full symbols).

SYNCHROTRON MICRO-TOMOGRAPHY FOR MONITORING THE FORMATION OF VOIDS DURING IN-SITU REACTION HEAT TREATMENT

Void formation in Nb_3Sn strands degrades the microstructural strand homogeneity and causes localized stress concentrations and therefore can degrade the electromechanical properties of the superconductor.

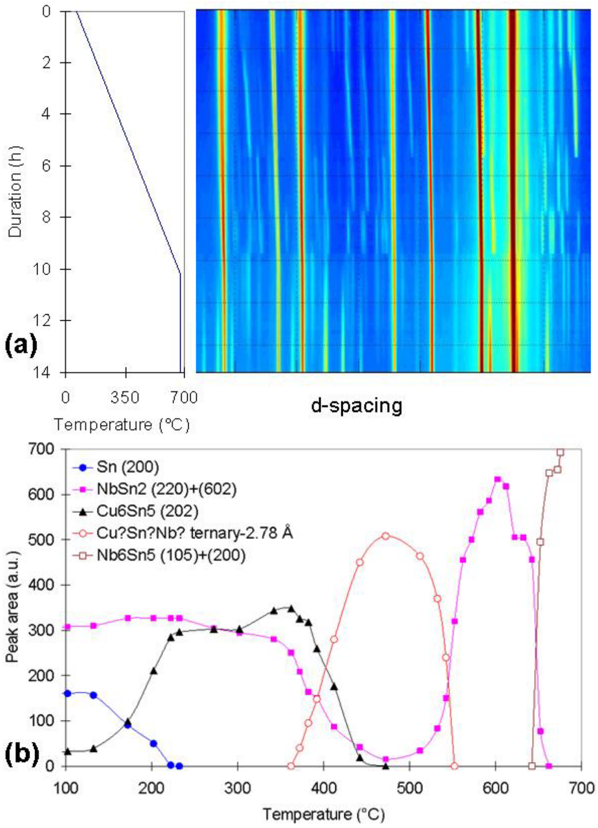

Figure 3: Variation of the diffraction patterns of a Nb_3Sn PIT strand during *in-situ* HT with a ramp rate of 60 °C h^{-1} and 4 hours isothermal 675 °C HT (a). The evolution of the diffraction peak areas of all Sn containing phases, apart from α-bronze, is shown in (b).

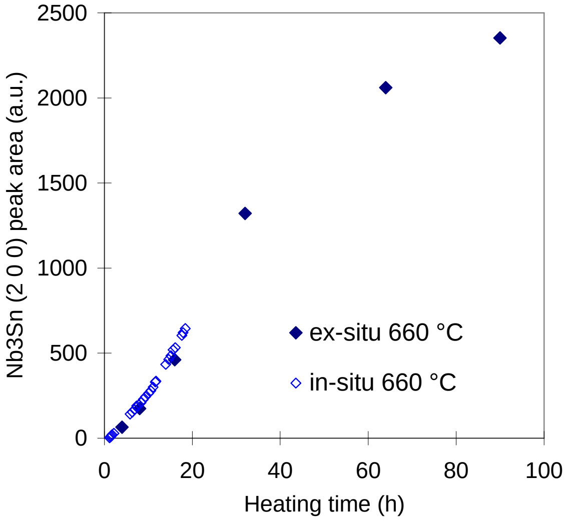

Figure 4: Nb_3Sn growth during isothermal 660 °C HT of PIT B215 monitored by Nb_3Sn (200) diffraction peak area measurements.

A quantitative description of void volume, shape and distribution within Nb_3Sn strands can be obtained using synchrotron micro-tomography. Sample preparation artefacts can be excluded by non-destructive tomography measurements.

The distribution of the voids that are formed during the reaction HT of an IT strand can be seen in the transverse and longitudinal strand cross sections shown in Figure 5.

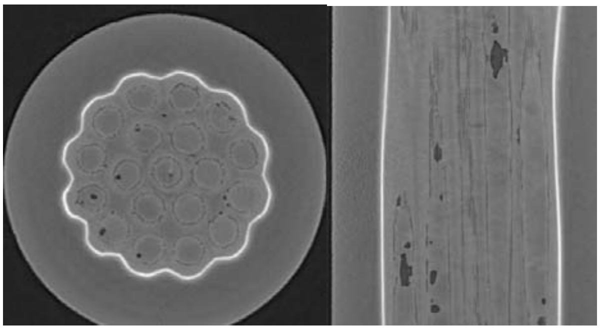

Figure 5: Transverse (left) and longitudinal strand cross section of an IT Nb_3Sn strand after *ex-situ* 580 °C HT.

The resolution of synchrotron micro-tomography measurements at ID15A is sufficient for the detection of the interfilament voids in IT strands that are formed as a consequence of differences in diffusion coefficients of Sn and Cu and Cu in Sn. The size of these voids shown in Figure 6 is about 1 µm.

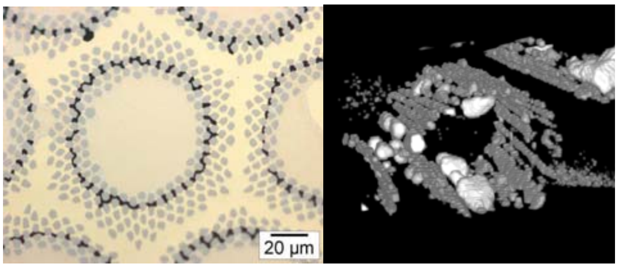

Figure 6: Optical micrograph of a strand cross section (left) and 3-D tomogram of the interfilament voids in one diffusion center of an IT Nb_3Sn strand after *ex-situ* 580 °C HT.

Due to the short acquisition time of less than one minute per tomogram, tomography experiments during *in-situ* HT are possible at the ID15A beamline of ESRF [2].

A sequence of 114 tomograms has been acquired during an *in-situ* HT of the IT strand shown in Figures 5 and 6. In Figure 7 a 3-D view of the voids formed in the Sn source regions of the IT strand at different temperatures is shown. The tomograms have been acquired during *in-situ* HT with a ramp rate of 60 °C h^{-1} with three additional isothermal holding steps for 2 h at 200 °C, 5 h at 340 °C and 2 h at 540 °C.

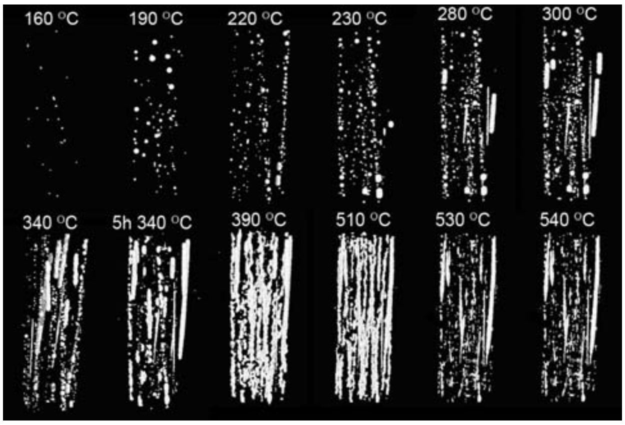

Figure 7: Voids inside an IT Nb_3Sn strand at different temperatures during *in-situ* reaction HT [3].

After the acquisition of each tomogram the strand sample has been moved in a monochromatic 88.005 keV x-ray beam for a diffraction measurement. The combination of micro-tomography and diffraction in one experiment allows to distinguish between different void growth mechanisms. The comparison of the void volume and Cu_3Sn volume as a function of the HT temperature (see Figure 8) shows that part of the voids in IT strands are formed because of density changes upon the formation of intermetallic phases [3].

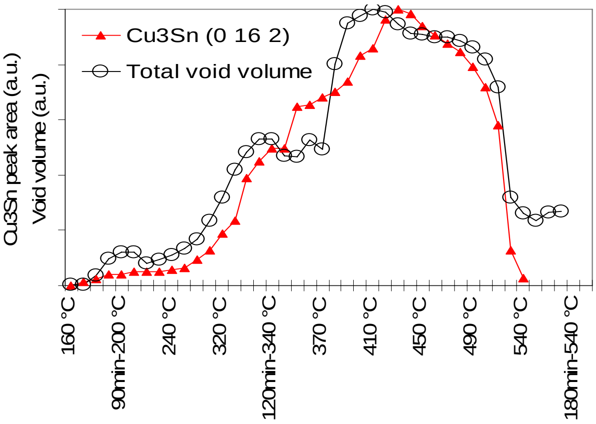

Figure 8: Comparison between the void volume and the Cu_3Sn volume formed during the reaction HT of an IT Nb_3Sn strand.

HIGH RESOLUTION SYNCHROTRON DIFFRACTION FOR MEASURING THE 3-D STRAIN STATE WITHIN NB$_3$SN COMPOSITE CONDUCTORS DURING IN-SITU TENSILE LOADING

High resolution diffraction measurements are a widely used tool for measuring the residual stress in a variety of different materials. In the case of Nb_3Sn composite wires, high resolution diffraction measurements can be applied for instance to measure the internal stresses caused by the mismatch of thermal expansion coefficients during the cool down from the processing temperature to the operating temperature and to measure the strain state in the different strand phases as a function of an externally applied wire stress, e.g. during a tensile test. As an example, the stress distribution in bronze route design Nb_3Sn strands at room temperature and at 4.2 K has been measured by neutron diffraction [4]. Because of the relatively low neutron flux it was necessary to use bundles of parallel aligned strands in order to increase the sample volume in order to acquire diffractograms with sufficient signal-to-noise-ratio within reasonable durations.

When the elastic strain of the different wire phases needs to be measured under a well defined uniaxial tensile stress, diffraction measurements must be carried out with a single strand configuration. At the POLDI strain scanning experiment at the SINQ neutron source of the Paul Scherrer Institut (PSI) it has become possible to study single multifilament composite wires under well defined macroscopic stress [5] [6]. However, acquisition times for acquiring Nb_3Sn neutron diffractograms with sufficient statistics are still in the order of hours and only the strongest Nb_3Sn reflection could be measured.

Due the high x-ray flux that can be provided at ID15, which exceeds the neutron flux of the most powerful neutron sources by many orders of magnitude, synchrotron experiments can be much faster and only a small sample volume is needed. At ID15B using a Trixell Pixium 4700 detector diffractograms with excellent signal-to-noise-ratio can be acquired in 10 seconds. This allows to perform diffraction measurements during *in-situ* tensile tests with standardised strain rates.

The relatively small scattering angles of the high energy x-rays used make it possible to add a dedicated tensile rig for stress-strain measurements of superconducting strands within a glass LHe cryostat (provided by University of Geneva) to the ID15B beamline and still record diffractograms with a large d-spacing interval [7].

Figure 9 shows preliminary results of the elastic strain in the different phases of a fully reacted PIT strand as a function of the composite strain measured with an extensometer. The resolution of the diffraction measurements that can be achieved is better than 0.01 %.

CONCLUSION

The continuously increasing x-ray flux provided through high energy synchrotron beam lines has enabled several new experiments with Nb_3Sn composite conductors. The main advantage of high energy synchrotron radiation experiments as compared to destructive microscopic techniques is that measurements can be performed *in-situ*. A better understanding of the superconductor processing could be obtained through the combination of different synchrotron techniques in one experiment, i.e. diffraction and micro-tomography during *in-situ* HT. High resolution synchrotron diffraction measurements can provide new information about the strain state within composite conductors under mechanical loading.

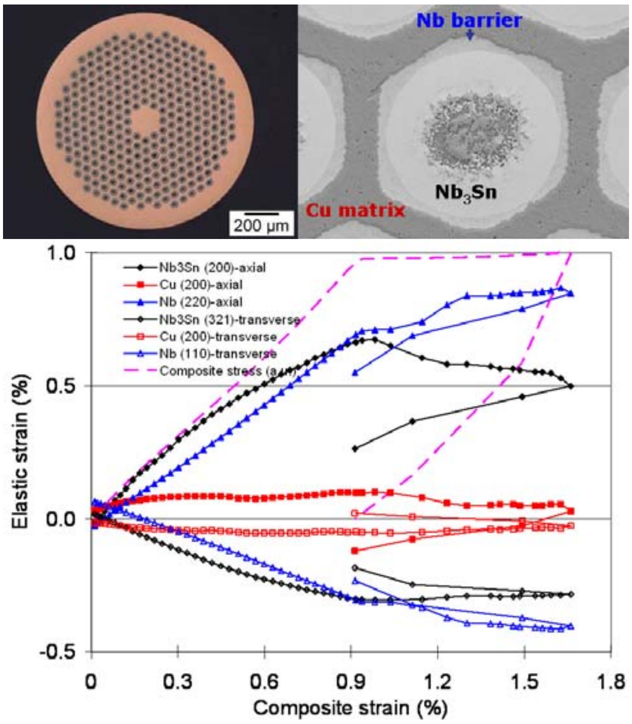

Figure 9: Elastic strain determined from selected reflections of the different phases of a fully reacted PIT strand, as a function of the composite strain, measured at 4.2 K with an extensometer. The nearly stress free lattice parameter used for the calculation of the elastic strain is an estimate.

REFERENCES

[1] M. Di Michiel, C. Scheuerlein, "*Phase transformations during the reaction heat treatment of powder-in-tube Nb_3Sn superconductors*", Supercond. Sci. Technol. 20, (2007) L55-L58.

[2] M. Di Michiel, J.M. Merino, D. Fernandez-Carreiras, T. Buslaps, V. Honkimäki, P. Falus, T. Martins, O. Svensson, "*Fast microtomography using high energy synchrotron tomography*", Rev. Sci. Instrum. 76, 043702, (2005)

[3] C. Scheuerlein, M. Di Michiel, A. Haibel, "*On the formation of voids in Nb3Sn superconductors*", Appl. Phys. Lett., 90, 132510, (2007)

[4] W. Goldacker, R. Flükiger, IEEE Transactions on Magnetics 21(2), 807-810 (1985)

[5] L. Thilly, P.O. Renault, V. Vidal, F. Lecouturier, S. Van Petegem, U. Stuhr, H. Van Swygenhoven, "*Plasticity of multiscale nanofilamentary Cu/Nb composite wires during in situ neutron diffraction: Codeformation and size effect*", Appl. Phys. Lett. 88, 91906, (2006)

[6] C. Scheuerlein, U. Stuhr, L. Thilly, "*In-situ neutron diffraction under tensile loading of powder-in-tube Cu/Nb3Sn composite wires: effect of reaction heat treatment on texture, internal stress state and load transfer*", Appl. Phys. Lett. 91(4), 042503, (2007)

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[7] F. Buta, C. Scheuerlein, M. Di Michiel, B. Seeber, R. Flükiger, “*High resolution X-ray diffraction experiments to investigate the electromechanical behavior of* Nb_3Sn *superconducting wires*”, poster presentation at the SPS Annual Meeting, Geneva, 2008