EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH
Laboratory for Particle Physics

Departmental Report

CERN/AT 2008-39

SYNCHROTRON RADIATION TECHNIQUES FOR THE CHARACTERIZATION OF Nb_3Sn SUPERCONDUCTORS

C. Scheuerlein[1], M. Di Michiel[2], F. Buta[3]

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 has been measured by high resolution x-ray diffraction during in-situ tensile tests.

1 CERN, Geneva, Switzerland.
2 European Synchrotron Radiation Facility, Grenoble, France
3 University of Geneva, Geneva, Switzerland

Presented at the Applied Superconductivity Conference (ASC 2008)
17-22 August 2008, Chicago, USA

CERN
Accelerator Technology Department
CH - 1211 Geneva 23
Switzerland

01 March 2009

Synchrotron radiation techniques for the characterization of Nb_3Sn superconductors

C. Scheuerlein, M. Di Michiel, F. Buta

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 has been measured by high resolution x-ray diffraction during *in-situ* tensile tests.**

Index Terms— **Diffraction, superconducting wires and filaments, tomography.**

I. INTRODUCTION

UNTIL recently the materials characterization of Nb_3Sn strands has been performed almost entirely by destructive methods, apart from studies performed with neutron techniques. Non destructive methods like high energy x-ray diffraction or micro-tomography can complement the destructive microscopic techniques that are commonly used to study 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 cannot 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. As compared to neutron sources, the flux obtainable by synchrotron sources can be many orders of magnitude higher. Therefore, synchrotron measurements can be very fast and they require only a small sample volume. In addition the relatively small scattering angles of high energetic x-rays make it easier to add auxiliary equipment (furnace, tensile rig, etc.) to the experiment in order to perform experiments *in-situ*. Different synchrotron techniques can be combined in one experiment (e.g. diffraction and micro-tomography during *in-situ* heat treatment (HT)).

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 Fig. 1. For the 1.25 mm-diameter strand x-ray energies above 70 keV are needed for measurements under optimum conditions.

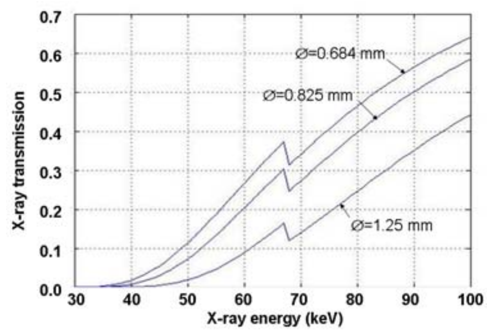

Fig. 1: X-ray transmission as a function of x-ray energy for an Internal Tin Nb_3Sn strand with different diameter. The x-ray transmission for obtaining optimum signal-to-noise ratio in tomography is about 20%.

Here we describe three synchrotron experiments for the materials characterization of Nb_3Sn strands, notably powder diffraction for phase analysis during *in-situ* reaction HT, synchrotron micro-tomography with µm spatial resolution for monitoring void growth during *in-situ* HT, and high resolution diffraction for measuring the strain state in the different phases of the composite superconductors under *in-situ* tensile loading. The potential of synchrotron micro-tomography for damage characterization in Nb_3Sn strands is assessed too.

II. 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 depend strongly on the strand composition.

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 European

Manuscript received 15 August 2008.

C. Scheuerlein is with CERN, Accelerator Technology Department, CH-1211 Geneva 23, Switzerland, phone: ++41 (0)22 767 8829, e-mail: Christian.Scheuerlein@cern.ch.

M. Di Michiel is with the European Synchrotron Radiation Facility, 6 rue Jules Horowitz, F-38000 Grenoble, France.

F. Buta is with the University of Geneva, Group of Applied Physics (GAP), CH-1211 Geneva 4, Switzerland.

Synchrotron Radiation Facility (ESRF) in transmission geometry, using a 87.0 keV monochromatic x-ray beam. Debye-Scherrer diffraction patterns were acquired in transmission geometry using a MAR345 image plate detector from Marresearch GmbH, Germany, with an area of 345 mm diameter. A diffraction pattern acquired for a Nb_3Sn PIT strand is shown in Fig. 2 (before radial integration).

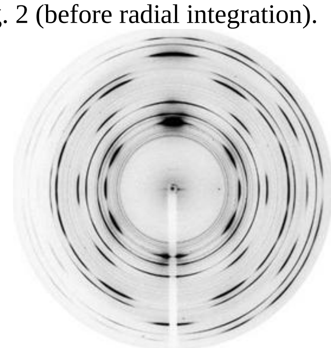

Fig. 2: Debye-Scherrer diffraction pattern of a reacted Nb_3Sn strand acquired in transmission geometry with a MAR 345 image plate detector.

For *in-situ* heating a dedicated x-ray transparent furnace that enables an accurate sample temperature control during the diffraction experiments has been added to the experiment. The color intensity plot in Fig. 3(a) shows a sequence of 85 radially integrated diffractograms. The diffractograms have been acquired during *in-situ* reaction HT of a PIT strand 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 Fig. 3(b).

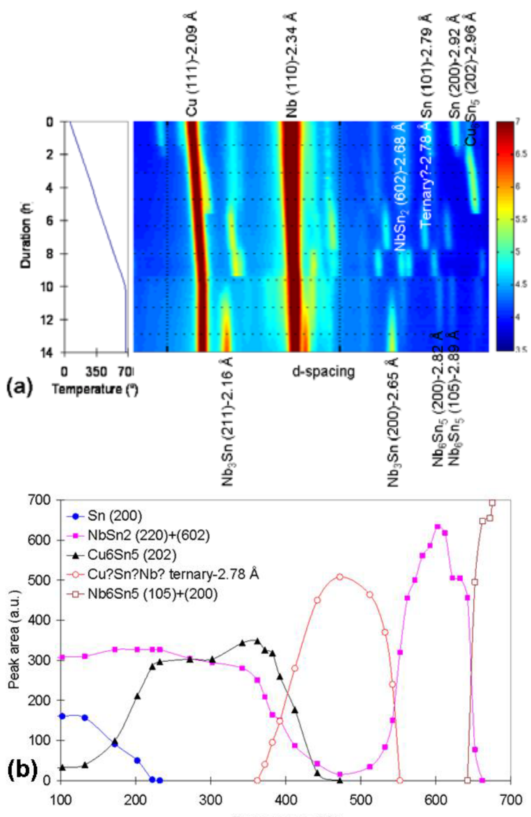

Fig. 3: (a) Variation of the diffraction patterns of a Nb_3Sn PIT strand during *in-situ* HT with a ramp rate of 60 °C h^{-1} and 4 hours isothermal 675 °C HT. (b) Evolution of the diffraction peak areas of the Sn containing phases detected in the PIT strand prior to Nb_3Sn formation as a function of the HT temperature. An un-identified phase, for which the evolution of the diffraction peak area (peak with d-spacing~2.78 Å) is also plotted, is presumably a ternary Cu-Nb-Sn phase.

III. Synchrotron micro-tomography for monitoring the formation of voids during in-situ reaction HT

Void formation in Nb_3Sn strands degrades the microstructural strand homogeneity and causes localized stress concentrations and therefore degrades the physical superconductor properties.

A quantitative description of void volume, shape and distribution within Nb_3Sn strands can be obtained using synchrotron micro-tomography. Sample preparation artifacts can be excluded by non-destructive tomography measurements. The distribution of the voids that are formed during the reaction HT of a Nb_3Sn strand fabricated by the Restacked Rod Process (RRP®) can be seen in the transverse strand cross section and in the 3-D view presented in Fig. 4.

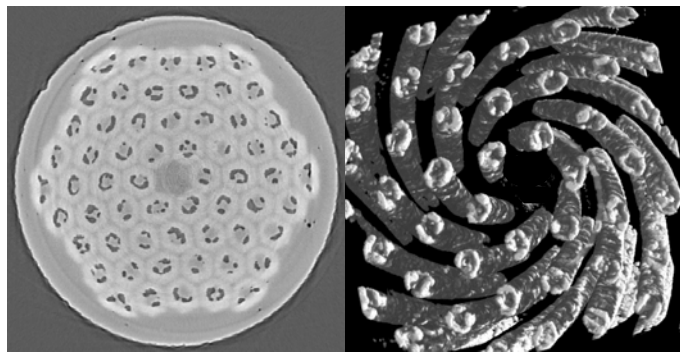

Fig. 4: Transverse cross section of an *ex-situ* processed Nb_3Sn RRP® strand obtained by synchrotron micro-tomography (left) and 3-D view of selected pores inside the strand (right). The total void volume corresponds with 7% of the entire strand volume.

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 Fig. 5 is about 1 μm.

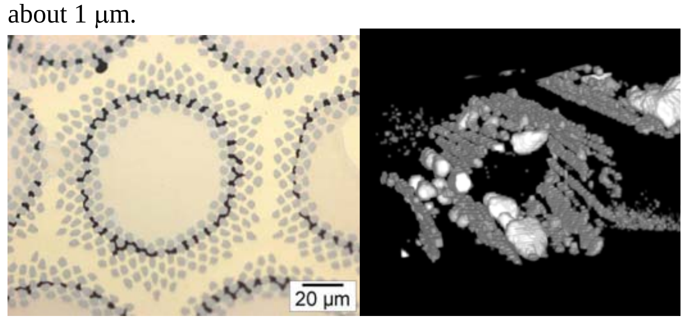

Fig. 5: Optical micrograph of a strand cross section (left) and 3-D tomogram of the interfilament voids in one diffusion center of a IT Nb_3Sn strand after ex-situ 580 °C HT. In the optical true color image the voids appear in black and in the tomogram voids are represented in white.

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 a HT of a Nb_3Sn IT strand. In Fig. 6 a 3-D view of the voids formed in the diffusion centers 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.

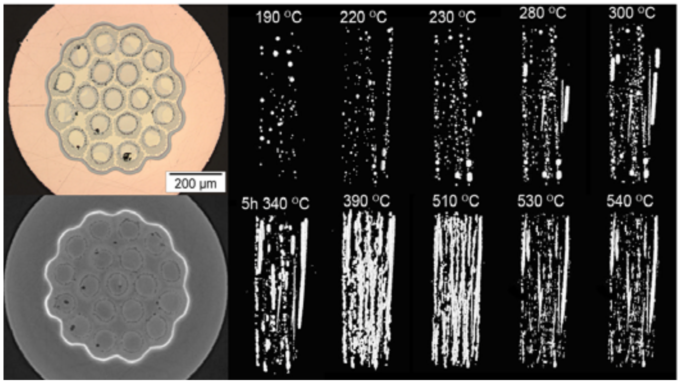

Fig. 6: Transverse cross sections of an *ex-situ* processed Nb_3Sn Internal Tin strand (left) and 3-D view of the pores inside the same strand acquired by synchrotron micro-tomography at different temperatures during *in-situ* HT (right).

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 Fig. 7) shows that part of the voids in IT strands are formed because of density changes upon the formation of intermetallic phases [3].

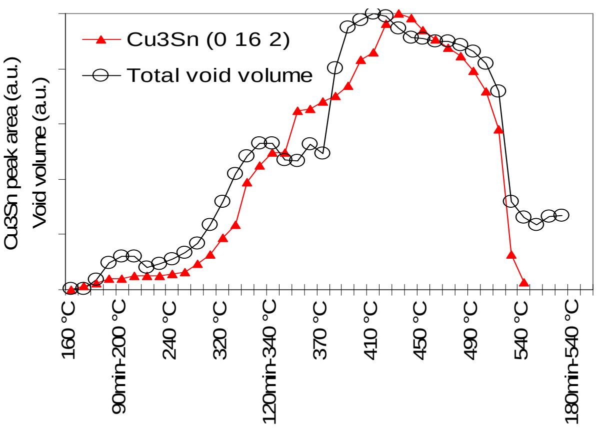

Fig. 7: Comparison between the void volume and the Cu_3Sn volume (in arbitrary units) formed during the reaction HT of an IT Nb_3Sn strand. The Cu_3Sn volume is estimated from the (1 16 2) peak area evolution.

IV. Synchrotron Micro-Tomography for Studying Crack Formation in Nb_3Sn

The reversible and irreversible strain induced degradation of superconducting Nb_3Sn strands is a major limitation for the application of Nb_3Sn cables in high field magnets for fusion devices or particle accelerators. The formation of cracks in Nb_3Sn under mechanical loading of the superconductor is assumed to be the main reason for the irreversible strain induced degradation of the critical current.

Synchrotron micro-tomography might be a tool for the damage characterization in Nb_3Sn strands and complement metallographic experiments. In the transverse cross section of a reacted PIT strand after transverse compression that is presented in Fig. 8 it can be seen that cracks can be detected in the Nb_3Sn part of some tubes. However, the presently achievable spatial resolution of the synchrotron micro-tomography is only sufficient to detect relatively large cracks and monitoring the onset of crack formation in Nb_3Sn strands remains a very challenging task. The spatial resolution of tomography experiments is related to the field of view. A somewhat better spatial resolution may thus be obtained using significantly thinner Nb_3Sn strands, or possibly extracted filaments.

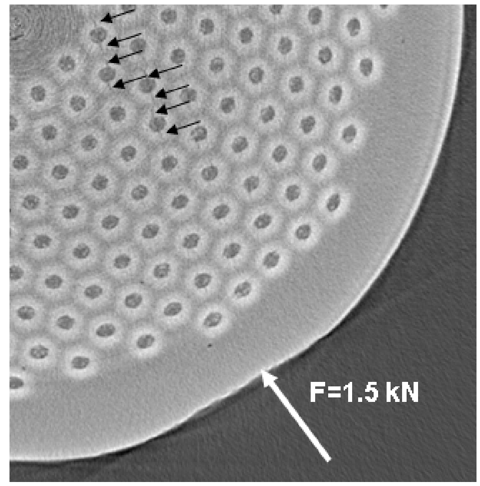

Fig. 8: Transverse cross section of reacted PIT B215 strand with a diameter of 1.25 mm after transverse compression at room temperature with a force of 1.5 kN that was applied over a strand length of 12 mm.

An advantage of tomography experiments over metallographic techniques is that the crack propagation through the sample can be observed (see Fig. 9).

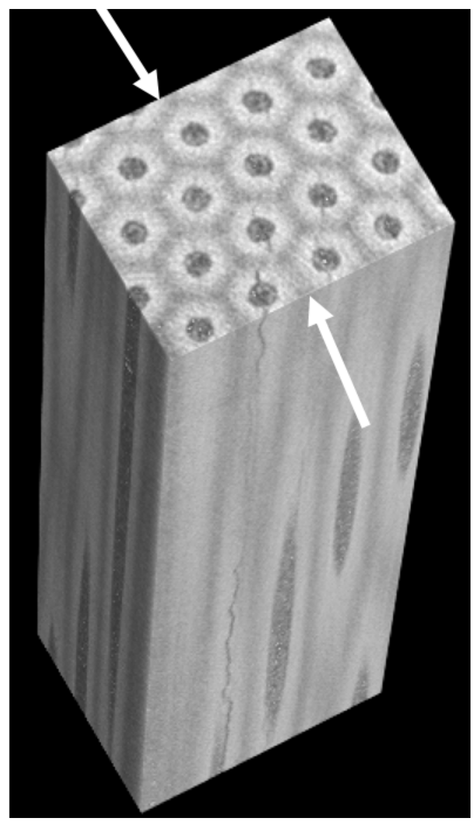

Fig. 9: Longitudinal cross section through a cracked tube of the reacted PIT B215 strand after transverse compression at room temperature with a force of 1.5 kN that was applied over a strand length of 12 mm. The arrows indicate the direction of the transverse forces to which the sample has been exposed. The crack propagation through the Nb_3Sn part of the tube is clearly visible. The crack formation in the PIT strand is strongly influenced by the hexagonal tube shape.

V. High resolution synchrotron diffraction for measuring the 3-D strain state within Nb_3Sn 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.

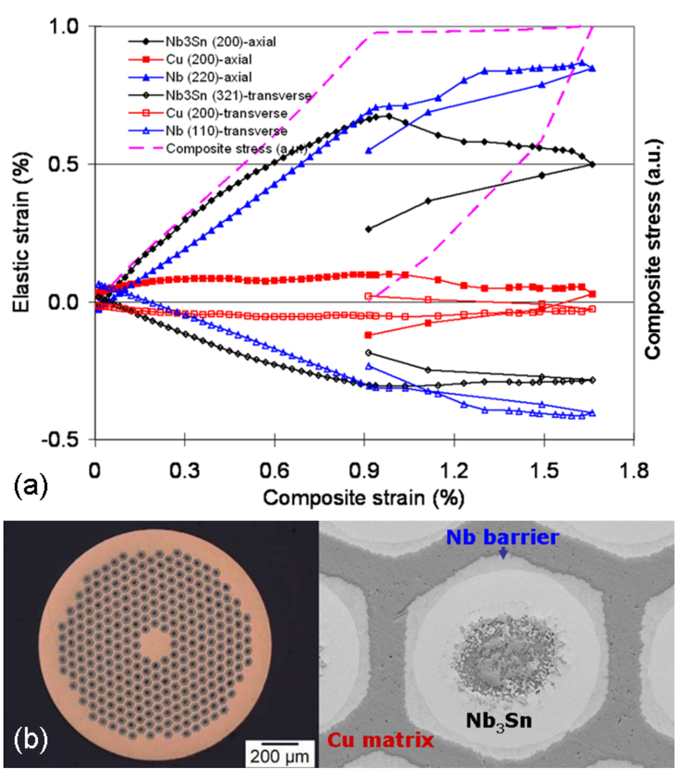

Fig. 10: (a) Elastic strain determined from selected reflections of the different phases of the reacted PIT strand shown in (b) as a function of the composite strain, measured at 4.2 K with an extensometer. The nearly stress free lattice parameter used for the calculation of the elastic strain is an estimate.

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 diffraction measurements during *in-situ* tensile tests with standardized 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 the University of Geneva) to the ID15B beamline and still record diffractograms with a large d-spacing interval [7].

Fig. 10 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%.

VI. 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 Nb_3Sn 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 superconductors under mechanical loading.

Acknowledgment

We are grateful to L. Thilly (University of Poitiers), H. Reichert (Max Planck Institut für Metallforschung, Stuttgart), R. Flükiger, B. Seeber (University of Geneva) and M. Jewell (National High Magnetic Field Laboratory, Tallahassee) for advise and suggestions.

We acknowledge the ESRF for beam time on ID15A and ID15B.

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, L55-L58, 2007.

[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 Nb_3Sn superconductors", Appl. Phys. Lett., 90, 132510, 2007.

[4] W. Goldacker, R. Flükiger, IEEE Transactions on Magnetics 21(2), pp. 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.

[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", private communication.