Neutron and synchrotron X-ray measurements: unique tools in the non-destructive toolbox

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1. Introduction

Neutron and X-ray beams provide unique, powerful and non-destructive access into the heart of materials and components, over length scales ranging from the centimetre to the atom. The Institut Laue-Langevin ILL – Europe’s leading neutron source - and the ESRF – Europe’s leading synchrotron light source, combine 80 experimental stations providing neutron and X-rays beams with exceptional properties to overcome the limitations of standard laboratory characterisation techniques. Regarding challenges in the aeronautic and space sectors, we will detail some advantages obtained by synchrotron and neutron beams in stress mapping and computed tomography.

The ESRF’s high-energy synchrotron X-rays (SRX) can map efficiently the strain and stress at the surface and up to several hundreds of microns below to give depth profiling with high spatial resolution. Neutrons give access to the full stress tensor from 60µm below the surface up to the interior of the component (e.g. 30 cm in Al, 6 to 7 cm in Ti, Ni or steel). Both techniques are able to measure in real time and non-destructively the stress distribution during mechanical loading, or during heating/cooling processes. A current exciting development is the investigation of residual stress build-up during additive manufacturing. Note that a combination of neutron and X-rays datasets can be offered by the institutes.

With regards to computed tomography, synchrotron X-rays and neutrons provide complementary information. SRX provide remarkable insights into materials up to the nanoscale with a resolution as high as 30 nm. At the microscale, synchrotron X-rays enable vastly more accurate (e.g. with phase contrast imaging and lower beam hardening effect) and faster data collection than lab equipment – 2D radiography can be performed at MHz rate for monitoring rapid phenomena and 3D CT can be done at ~Hz speed. With these capabilities, it is possible to monitor dynamic processes in real time (e.g. crack propagation, exploding fuse or bed fusion in additive manufacturing). Neutron tomography makes use of the specific properties of thermal neutrons to penetrate deeply a large majority of materials (e.g. most metals) whilst being highly sensitive to other elements, notably hydrogen and lithium, enabling the imaging of water, polymers or explosive charges embedded in metallic structures, without metal induced image artefacts neither radiation damage.

This article is rather an overview of two synchrotron and neutron-based techniques – strain/stress mapping and tomography- in the field of aerospace, and engineering on a larger scope.
2. Strain and stress mapping using neutron and synchrotron X-rays diffraction

2.1. Introduction to diffraction technique for stress determination

The description of diffraction technique for the measurement of stress has been extensively described in various articles and books [1, 2]. We thus provide here a short overview.

![Figure 1](image1.png)

**Figure 1** This drawing illustrates the various stress level that are described within a material: intragranular, intergranular and macrostress. Neutron diffraction gives macroscopic stress field; SRX is well adapted to the determination of intra- and intergranular stresses. SRX can also provide strain values at the macroscale.

![Figure 2](image2.png)

**Figure 2** Evaluation of strain is based on the diffracted signal by crystalline material. As far macroscopic stress is concerned, diffractive techniques rely on the multiple diffraction signal given by a large number of grains. In case multiple significant phases are present, diffraction signal is collected from the various phases.

Strain can be expressed as the following:

\[
\varepsilon = \frac{d - d_0}{d_0}
\]

Where \( d \) is the average lattice spacing measured in a given region of the piece and \( d_0 \) is the reference lattice spacing, ideally from a stress-free coupon made of the same material (same batch) as the piece under investigation. When comparing post-treatment processes, the reference sample is often the piece itself as-built, i.e. before undergoing post-treatment or aging processes. SRX and N techniques are relative techniques and absolute values can be obtained after a careful definition of a reference [3].

Neutron diffraction set-up allow the measurement of additional independent strain components when principal directions are unknown. Additionally, ISO technical specification was published in 2012 as far neutron stress diffraction is concerned [4]. Geometries of data acquisition for each technique and main features are summarised in Figure 3.
Neutron stress determination is a non-destructive technique, applicable to mock-ups as well as realistic sized engineering components [5, 6]. It allows mapping of stress fields from the bulk of the work piece to its surface with adjustable lateral resolution, providing the full stress tensor for each measuring point. Neutron techniques are well suited for in-situ or in operando investigations, using even complex sample environment. Examples are heat treatment, tensile testing, fracture dynamics, casting or additive manufacturing [7-13].

SALSA, which stands for “Stress Analyzer for Large Scaled engineering Applications”, is a stress diffractometer dedicated to engineering sciences and industrial R&D [14] available at the ILL. SALSA provides a maximum of flexibility in terms of sample size and shape, allowing laterally resolved stress- and texture determination. The resolution is variable between 0.6 and 4 mm and allows stress mapping with penetration depths of 60 mm in steel, 70 mm in Titanium alloys, 40 mm in Nickel and 300 mm in Aluminium, to give some examples. The minimum sample thickness for stress determination is 1 mm. Uncertainty on stress values are in the range of 30-50 MPa.
A special technique allows the determination of stress profiles from the surface into the bulk with the first measuring point at 40 microns below the surface. This technique is applied to surface treatment investigations, comprising laser shock peening, induction hardening, spray coating etc. SALSA can host samples of up to 1.5 m length and a weight of 700 kg. The sample table is a hexapod, allowing tilt and translation in order to position the sample with a precision of 5 micrometres.

2.2. An example: investigation of Friction Stir Welds

As an example we present partial results of a test series with the company OHB-System regarding friction-stir welding (FSW) of aluminium [15]. OHB System\(^1\) is one of the leading space companies in Europe, delivering satellites and high-tech components for the space sector.

Friction stir welded (FSW) Aluminium plates of 6 mm in thickness were investigated using the set-up shown in Figure 5. Special interest lied on the stress field across thickness within nugget and heat affected zone (HAZ). The gauge volume was defined by three focussing collimators to a dimension of 0.6x0.6x2mm\(^3\). The sample is then position and scanned through the gauge volume with the help of the hexapod stage. For each position, the Bragg-diffraction peak Al(311) is recorded.

![Figure 5](image.png)

*Figure 5 Set-up for measurements at SALSA in two welded plates having different welding parameters. The scattering vector q indicates the investigated strain component: in this specific geometry it is the transversal one. The distance between beam optics and samples is about 400 mm, leaving room for sample movements.*

Figure 6 shows 2D maps and profiles of the three principal stress components: longitudinal (in welding direction), transversal (to the weld nugget) and normal (to the plate). The investigated area covers 16 mm in width from the centre of the weld into the HAZ and 3 mm in depth to the middle of the weld thickness. Origin is the surface central point of the weld. The normalized\(^2\) 2D stress distribution indicates the gradient between tensile (+) and compressive (-) regions and their evolution in depth of the weld. As expected, maximum tensile stresses appear near the HAZ, with a more compressive state within the weld nugget. Note that all stress levels are normalized by the longitudinal component. The line profiles show the stress evolution parallel to the surface at different depths. The maxima follow the weld seam inside the material and a clear trend to tensile stresses from the surface to the middle thickness of the plate is detected.

\(^1\) [https://www.ohb-system.de/](https://www.ohb-system.de/)  \(^2\) Absolute values not shown for confidentiality reasons
3. Specificities and complementary in X-rays and neutron imaging

X-rays radiography and tomography are now well established NDT techniques in industrial activities. Although these benchtop techniques cover most of the needs, imaging set-up at large scale instruments offer specific and advanced features.

Thanks to very high brightness of synchrotron sources, it is possible to achieve very high spatial resolutions (e.g. in the nanometer range), very fast scans and additional imaging modality beside the usual absorption based contrast [16]. High energies available at ESRF allow increased penetration in metals.

Thermal and cold neutrons available at neutron research facility like the Institut Laue-Langevin (ILL) are a nicely complementary probe to X-rays [17]. Neutrons are highly sensitive to hydrogen, lithium, and isotopes of a given element while having a high penetration in metals, including dense ones like steel, Ni, W or Pb.
3.1. Recent advances in neutron tomography

Recent developments in neutron imaging now allow 200 µm spatially resolved tomography to be acquired within less than 2 s [18]. Spatial resolution as low as 4 µm has been recently achieved at ILL [19].

Main application of neutron imaging is found in the investigation of distribution and transport of H-rich fluid (e.g. water, oil) in geomaterial, engineered porous media or plants. Neutron tomography has been also successfully utilised in engineering to optimize water management in fuel cells [20] or visualise lithium distribution in batteries [21].

![Figure 7](image)

Figure 7 A neutron radiography of a piece made of composite having an Al matrix: the glue appears with a high contrast because it contains hydrogen.

3.2. The multiple aspects of synchrotron X-rays imaging

Synchrotron X-rays are about 1000 times more intense than lab X-rays sources allowing very fast radiography or tomography. As tomographic data can be acquired within few hundred of milliseconds, it becomes possible to follow evolving phenomena [22-26]. Due to its intrinsic coherence synchrotron radiation allows phase contrast tomography. Finally, as beams of micron size can be defined and energy can be finely tuned, other imaging modalities are accessible like diffraction imaging. Insights into the above mentioned image acquisition are given in the next paragraphs.

Absorption microtomography

Microtomography is an imaging technique, based on the absorption of X-rays by materials. Sample changers have been developed to be able to analyse a series of samples under the same experimental conditions. Additionally, various sample environment device are available for users: a traction/compression device, a fatigue device, a cryostat, a cryostream, ovens of different characteristics, a hot traction device, a hygrometry controller, etc.

The exceptional photon flux density and brilliance at ESRF allowed acquisition of tomograms over very short periods of time providing unprecedented temporal resolution for 4D (time lapse) tomography whereby fast morphological changes were tracked in 3D over time. For the first time we have been able to image the nucleation and propagation of failure within working cells in real time [26]. Simultaneous thermal imaging provided detailed information on the temperature of the cell during failure which was enabled by in situ containment apparatus to protect beamline equipment from energy intensive failures. For example, it was found that an internal cylindrical support (Figure 8a) played an important role in improving the safety of a cell by providing structural support for the tightly wound electrode layer, and acting as a channel for gases to flow uninterrupted from the base of the cell to the vent during failure. High speed radiography at 1250 fps captured the rapid propagation of thermal runaway (Figure 8c) in both cells and highlighted the cause of clogging by identifying the shift of the bulk material towards the vent; clogging led to an uncontrolled ejection of the cell contents.

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2 This paragraph has been adapted from [http://www.esrf.eu/home/UsersAndScience/Publications/Highlights/highlights-2015/structure-of-materials/som03.html](http://www.esrf.eu/home/UsersAndScience/Publications/Highlights/highlights-2015/structure-of-materials/som03.html)
Figure 8 Commercial Li-ion batteries (18650 NMC cells) investigated at ESRF. (a) 3D reconstruction of a fresh cell with internal cylindrical support, (b) thermal image of a failing cell, (c) radiograph showing the propagation of thermal runaway and (d) post-mortem 3D reconstruction where the majority of the copper current collector within the cell has melted.

**Phase contrast microtomography**

The X-rays beams produced by a third-generation synchrotron have a high degree of spatial coherence. In addition to the contrast related to the absorption of the material, this property brings another type of contrast, the phase contrast. Phase contrast images can be obtained by simply varying the distance between the sample and the detector. This is the so-called "propagation" technique [27]. The advantage of this imaging technique is its increased sensitivity, particularly for lightweight polymeric materials or composite materials whose different components have very close linear attenuation coefficients (e. g. Al and Si, see Figure 9).

![Absorption and Phase contrast images](image)

Figure 9 Tomography of an Al / Al-Si system. A slice from the same data set where absorption (left) and phase (right) are shown.
**Diffraction Tomography**

Diffraction Contrast Tomography combines the concepts of diffraction imaging and synchrotron X-rays microtomography to access the crystalline 3D microstructure of polycrystalline materials. The analysis of the shape and position of the diffraction spots, recorded at the same time as the absorption images, makes it possible to trace back:
- to the three-dimensional shape of the grains
- to the crystallographic orientation of the grains
- the elastic stress tensor of the grains

![Figure 10 Reconstructed microstructure of a beta Ti sample containing 1000 grains. From [28].](image)

**4. Few other key tools available at neutron and synchrotron centres**

In brief, we here provide further examples of techniques that can be useful to non-destructive investigation in the aerospace sector.

In-situ X-rays or neutron diffraction provide valuable insights about on-going processes when it comes to alloy, composites or catalysts design and manufacturing [29-32].

Small angle scattering using either neutron (SANS) or X-rays (SAXS) can be considered when one needs to investigate the evolution of the microstructure (i.e. features in the range of 1nm-1µm) under varying environmental parameters. SANS and SAXS address in particular domains like silicon-polymer composites [33], corrosion [34], porosity in coatings [35].

**5. Conclusion**

Both synchrotron X-rays and neutrons available on large instruments open-up possibilities in investigating phenomena/process while they are taking place. Unique features can be summarized as follow:
- Non destructive (high penetration)
- In-situ and in-operando investigations
  - Manufacturing/operating processes
  - Devices in operation
- Statistical, time resolved measurements

ILL and ESRF have both dedicated personnel to address requests from industrialist that one can contact directly: industry@ill.eu and industry@esrf.eu.

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