
**In situ studies of light metals with synchrotron radiation and neutrons**

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In situ studies of light metals with synchrotron radiation and neutrons


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Abstract. High-energy X-rays and neutrons offer the large penetration depths that are often required for the determination of bulk properties in engineering material research. In addition, new sources provide very high intensities on the sample, which can be used not only for high spatial resolution using very small beams, but also for high time resolution in combination with a fast detector. This opens up possibilities for a wide range of specific engineering in situ experiments. Typical examples that are already widely used are heating or tensile testing in the beam. However, there are also more challenging experiments in the field of light metals, like e.g. friction stir welding, dilatometry, solidification, or cutting. Selected examples are presented.

Introduction

Neutron scattering is widely used in materials research involving diffraction, small-angle scattering, or tomography [1]. The corresponding techniques for investigation of texture, residual stress, precipitates, and other elements of the microstructure have been developed over decades and are still under development today. Available sample environments include, e.g., furnaces and load frames for in situ testing. Neutron experiments sometimes suffer from the low brilliance of the sources, and the design of sophisticated instruments for increasing the flux on the sample is a challenge. However, neutrons have unique properties (scattering power, magnetic moment, penetration depth, energy regime) making them indispensable not only for material research. The new European Spallation Source (ESS) will greatly improve the situation and enable flux-hungry experiments that were not possible before the advent of such sources [2].

X-ray scattering and micro-tomography using synchrotron radiation has evolved over the last decade as an increasingly utilized technique for characterizing the internal structure of materials. This development is based on the higher brilliance that synchrotrons offer, compared to laboratory X-ray sources, leading to an increased resolution in reciprocal and real space. Synchrotron sources can offer a wide energy spectrum, thus high energies (50–150 keV) can be used for achieving large penetration depth in light metals. The high brilliance of the source can be used for high spatial or time resolution, enabling experiments that are not possible with laboratory sources or neutrons. In general, both probes, neutrons and X-rays, can be regarded as complementary [3].

The Helmholtz-Zentrum Geesthacht utilizes both neutron and X-ray experiments for engineering material research [4]. New beamlines were built at the third generation synchrotron source PETRA III at DESY with an instrumentation that has a focus on in situ experiments [5]. The principal layout of a high-energy synchrotron experiment is sketched in Fig. 1. The scattering angles are small at high X-ray energies so that complete Debye-Scherrer rings can be recorded with an area detector [6]. When the area detector is placed several meters away from the sample, also small-angle scattering can be recorded. The beam size can vary between 0.01 and 1 mm; sub-micron beams can be produced by focussing X-ray lenses. Closed sample environments with vacuum or shielding gases are built with beam windows.
In situ experiments require special sample environments applying specific environmental conditions to the sample. These cover all sorts of heating and cooling, application of mechanical forces or different fields or media. The challenge is to combine a complex and bulky sample environment with the need to feed the neutron or X-ray beam through it. Sometimes neutrons are advantageous for this purpose because they enable large scattering angles, sometimes high-energy X-rays are advantageous because of their small scattering angles. Another requirement is fast detectors for kinetic studies.

One example for successful in situ experiments at the HZG beamlines at DESY is a transportable machine for in situ friction stir welding experiments with a state-of-the-art welding head. Another example is an in situ cutting experiment, where the formation of cutting chips was studied [7]. In situ experiments with a dilatometer will be introduced in the following. A special challenge for in situ tomography experiments is the need for a precise rotation of the sample for 180°, which must not be hindered by the sample environment. Moreover, this rotation needed for one tomogram has to be fast enough to follow the sample kinetics. This requires high flux, a fast detector, and stable mechanics. The second example shows investigations where also in situ tomography and diffraction experiments dealing with the solidification of Mg alloys are planned. Finally, the investigation of isothermal precipitation kinetics in Al alloys using in situ small-angle neutron scattering is introduced.

The transformation mechanism of ternary phases in Nb-rich γ-TiAl alloys

Intermetallic γ-TiAl based alloys are a class of novel, lightweight structural materials with attractive mechanical properties for advanced high-temperature applications. Conventional titanium aluminides are two-phase alloys consisting of tetragonal γ-TiAl and small amounts of hexagonal α2-Ti3Al. Due to their low density, their high yield and creep strength up to 800 °C, and their good oxidation resistance they are used to replace heavier Ni-based superalloys in industrial and in aviation gas turbines as well as in automobile engines [8]. However, their low ductility impedes broad industrial application up to now. Recently, intermetallic γ-TiAl based alloys with additional amounts of the ternary bcc β-Ti(Al) phase attracted increasing attention due to their improved workability at elevated temperatures [9, 10, 11, 12]. Depending on alloy composition and heat treatment, the ductile high-temperature β phase can transform to several ordered phases at lower temperatures. However, actually available phase diagrams of these multiphase alloys are quite uncertain and the precipitation kinetics of some metastable phases is far from understood.

Thus, in situ high-energy X-ray diffraction experiments were performed at the HZG beamline HARWI II at Deutsches Elektronen-Synchrotron (DESY) [13]. At a photon energy of 100 keV (\(\lambda = 0.12398 \ \text{Å}\)) the resulting diffraction rings were recorded on a Mar555 flat panel detector each 120 s with an exposure time of 20 s. The highest possible frame rate with that detector is 0.37 s\(^{-1}\), but also significantly faster detectors exist.
A commercial dilatometer DIL 805A/D (Bähr-Thermoanalyse GmbH) with modifications for working in the synchrotron beam was used for heating and quenching of samples with a diameter of 4 mm [5]. The samples were heated up to 1100 °C and subsequently quenched to 700 °C with quenching rates of 10, 1000 and 100 K·min⁻¹.

Depending on the quenching rate, reversible transformations of the cubic B2-ordered β₀ phase to different ω related phases are observed in Ti-45Al-10Nb (Fig. 2). At low quenching rates the hexagonal B8₂-ordered ω₀ phase is formed while at high quenching rates the metastable intermediate trigonal ω'' phase can be preserved. The results indicate that the complete transformation β₀ → ω₀ consists of the steps β₀ → ω'' and ω'' → ω₀, which are both diffusion controlled. The transformation step β₀ → ω'' only needs atomic rearrangements over relative short distances which can occur significantly faster than the diffusion processes necessary for the ω'' → ω₀ transformation.

With the modified commercial dilatometer in the synchrotron beam, elongation and phase content can be measured simultaneously during precise and fast heat treatments. Moreover, installations of a deformation as well as a DSC unit are options to expand the measurement possibilities. This makes the dilatometer a unique sample environment for in situ materials research experiments.

**Microstructure analysis of Mg alloys by micro-tomography**

Mg-alloys represent a suitable alternative for lightweight parts in the automotive and aeronautic industries. Therefore, the study of Mg-alloy solidification is an important issue for the optimization of manufacturing processes. The solidification of metals is a complex process that starts with the nucleation of a solid embryo in an under-cooled region of a liquid melt, proceeds with the growth of a primary solid phase, and ends with the precipitation and growth of secondary phases [14, 15]. Most of the experimental evidence to explain these mechanisms relies on static or ex situ studies of as-cast specimens. However, some further questions on how a microstructure reaches its final state and how such a process can be controlled on behalf of the final properties of the material can only be addressed through in situ characterization techniques.

With 3rd generation Synchrotron light sources (e.g. PETRA III at DESY) it is possible to perform in situ X-ray diffraction, radiography, and tomography experiments to study the physical phenomena associated with the solidification of metals. Up to present, only a few X-ray synchrotron tomography studies of as-cast and extruded microstructures of commercial Mg-alloys have been performed [16, 17] in order to investigate the size, morphology and distribution of primary and secondary phases after processing. As an example, tomography experiments at room temperature...
have allowed to compare the microstructures of as-cast and extruded alloys, and investigate the amount, size, and distribution of intermetallic phases in the interdendritic regions. Fig. 3 shows the presence of micro-porosity can be identified. A resolution of 1.5 µm was achieved for cylindrical precipitates. The precipitation kinetics at aging temperature of about 0.1 cm3 can be probed. Thus, SANS is ideally combined with transmission electron microscopy (TEM) and atom probe (3DAP), for which the field of view is very small.

**Precipitation kinetics in AA60xx Al alloys**

Small-angle neutron scattering (SANS) is a classical method for the study of precipitation kinetics in alloys [21]. With standard SANS instruments, precipitate size distributions in the range of about 1–100 nm can be determined. When the chemical compositions of matrix and precipitates are known, also precipitate volume fractions can be determined. The advantage of SANS is the sensitivity for small changes in the size distribution because a relatively large sample volume of about 0.1 cm3 can be probed. Thus, SANS is ideally combined with transmission electron microscopy (TEM) and atom probe (3DAP), for which the field of view is very small.

Age-hardenable Al alloys play an important role in aircraft construction. Existing alloys are modified and new alloys are developed to improve, e.g., strength and fatigue properties according to increasing demands. Such alloys are strengthened by second-phase precipitates with sizes of a few nanometres. Thus, small-angle scattering is ideally suited for the characterization of these precipitates. The precipitation kinetics at aging temperature can be studied *in situ* with a furnace in
the beam. When X-rays from a synchrotron source are used for such studies, very high time resolution can be achieved because of the high intensity. However, some alloys do not have a significant contrast between matrix and precipitates and the small-angle X-ray scattering (SAXS) signal is negligible. Nevertheless, such alloys can still be studied with SANS because the scattering contrast for neutrons is completely different.

This is also the case for the Al alloy AA6056. The precipitates contain Mg and Si so that no SAXS signal was observed. SANS, however, produced a strong signal. SANS measurements had been carried out at the instrument SANS-2 of the Helmholtz-Zentrum Geesthacht. The mean neutron wavelength was 0.58 nm, the detector distance was 1 m. A furnace with ceramic heating elements in close contact with the sample was used for the experiment [22]. The aging temperature of 190 °C was reached within 13 s so that early stages of the isothermal reaction could be observed. The data acquisition time in this stage was 30 s, giving rather weak statistics. Thus, the integrated intensity $Q$ was calculated, which is proportional to the precipitate volume fraction and the scattering contrast. When the chemical composition of the precipitates does not change, e.g. in the case of classical nucleation and growth, and the covered range of scattering vectors is large enough, $Q$ is a measure for the precipitate volume fraction.

Two stages could be distinguished in the precipitation reaction at 190 °C: in the beginning, $Q$ grows with $t^{0.8}$, while after about 2 hours $Q$ grows with $t^{0.2}$ (Fig. 5). The mean precipitate radius grows to about 2.5 nm in the observed time range. The alloy AA6013 with a slight variation in the composition shows a very similar behaviour. The further evaluation of the SANS data includes the determination of precipitate sizes as a function of isothermal aging time; the results will be published elsewhere.

Such data can only be produced by small-angle scattering techniques. The statistics and time resolution of the presented SANS measurements can still be significantly improved by using neutron sources stronger than a medium-flux reactor, such as FRM II [23] or ESS [2]. The results can help to improve the understanding of details of the precipitation reaction, either by direct evaluation or by comparison with model predictions. In this way, they can also contribute to the improvement of mechanical properties of such materials and the development of alloy modifications.

**Outlook**

The unprecedented brilliance of third-generation synchrotron sources like PETRA III and the unprecedented flux of spallation neutron sources like ESS enable new *in situ* experiments with extremely high spatial and time resolution. This opens up new possibilities for investigations of microstructural elements and technical processes in the field of engineering materials research.
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References


[23] http://www.frm2.tu-muenchen.de