Study of the Evolution of Semi-Solid Alloys By Means of Phase-Sensitive Synchrotron Tomography

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Abstract. Recording the three-dimensional structure present in metallic alloys has become mandatory to help us understand the microscopic processes that determine the mechanical bulk properties of these materials. In the present report we apply phase-sensitive synchrotron tomography to investigate the microstructure of various commercial alloys used for semi-solid processing in three dimensions. Despite the lack of absorption contrast between the main elements of these alloys – aluminium, magnesium and silicon – we could resolve the individual phases present in the alloys with a resolution down to 1 to 2 μm. We detected coarse primary α-grains beside a fin-structured secondary constituent and find an excellent agreement with 2D sections obtained by light microscopy.

Introduction

Synchrotron tomography has become the spear-tip in the field of computed tomography (CT) ever since the construction of third generation synchrotron X-ray sources in the mid-1990s, constantly improving the speed and quality of three-dimensional data acquisition [1]. While manufacturers of laboratory CT instruments were working hard to catch up in terms of spatial resolution and acquisition time with the European (ESRF, DESY) and U.S. synchrotron facilities (APS, ALS) a new method of coherent imaging based on Fresnel propagation was proposed already a decade ago [2]. In analogy to coherent light and electron microscopy this imaging method applied well to biological and engineering materials that require strong enhancement of image contrast in order to visualize different material phases, micro-porosities and material interfaces that are hardly visible in absorption-based imaging. Fresnel-propagation-based imaging, first baptized with the misleading term phase-contrast imaging, was then extended to numerical retrieval of the phase-shift in the X-ray beam thus providing a good estimate of the three-dimensional distribution of the electron density in structural materials, leading to what is now known as holotomography [3].

The microstructures present in light engineering alloys quickly showed to be an interesting field of application both for propagation-based imaging and holotomography. Some of the constituting elements of such alloys (principally Al, Si and Mg) do not produce a sufficient image contrast when shadow images are recorded only showing the linear attenuation coefficient of the material projected along the X-ray beam. In particular

the three-dimensional analysis of micro-structured alloys as they are used for semi-solid casting require signal enhancement based on Fresnel propagation. However the limited availability of partially coherent synchrotron light sources and the relative complex procedure of numerical phase-retrieval have limited the exploration of semi-solid alloys with this technique on a larger scale.

We report our first findings from investigations of a variety of alloys which are processed in the semi-solid state. We shall frequently call these alloys "semi-solid" although we study them after full solidification. We use the experimental tomography stations at the **E**uropean **S**ynchrotron **R**adiation **F**acility (ESRF – ID19) and the **B**erlin **E**lectron **S**torage Ring Company for **S**ynchrotron Radiation (BESSY - BAM*line*) [4]. Fresnel-propagated micro tomography as well as holotomography are applied to magnesium-based engineering alloys (e.g. AZ91, AM60) and to aluminium alloys (e.g. A356) resulting in three-dimensional data sets of the metallic microstructure which are mandatory for understanding mechanical properties of the semi-solid state.

1. Methods

1.1 Fresnel propagation of hard X-rays

It is the very weak interaction of hard X-rays (wavelengths from 0.01 to 0.1 nm) that favour this type of radiation for the study of solid matter. In contrast to visible light the complex refractive index is almost unity for all kinds of materials. It is written as:

$$n = 1 - \delta + i\beta \tag{1}$$

with the imaginary component β due to the photoelectric effect and inelastic Compton scattering while the real decrement δ is due to elastic Rayleigh-Thomson scattering. For typical light metallic alloys δ is ~2-3 orders of magnitude larger that β which is of the order 10^{-9}. However δ is not directly accessible while β is recorded easily from contact images of the sample where the finite X-ray translucency of the material is measured in terms of the intensity I_0 following the Lambert-Beer law for the attenuation of an incoming X-ray beam of intensity I_{inc} transmitted along the z-axis and recorded in an image plane (x,y) immediately downstream of the sample (this position is marked by the subscript '0').

$$\frac{I_0(x, y)}{I_{inc}(x, y)} = \exp(-2B(x, y)) \quad \text{with} \quad B(x, y) = \frac{2\pi}{\lambda} \int_{sample} \beta(x, y, z) dz \tag{2}$$

$B(x,y)$ is the z-projection of the imaginary part of n, λ the X-ray wavelength. For homogeneous samples equation (2) is written in terms of the linear attenuation coefficient $\mu = 4\pi\beta/\lambda$. The information about δ is lost because we measure intensity which is the squared modulus of the complex wave $I_0(x, y) = |u_0(x, y)|^2$. However the transmitted wave carries information about both δ and β concealed in a complex transmission function $T(x,y)$:

$$T(x, y) := \frac{u_0(x, y)}{u_{inc}(x, y)} = \exp(i\phi(x, y) - B(x, y)) \tag{3}$$

with the wave's phase $\phi(x,y)$ corresponding to the line projection of the real part of n in analogy to $B(x,y)$ from equation (2). For the present application the knowledge of $B(x,y)$ is insufficient for distinguishing the different material constituents and we have to seek for additional information from $\phi(x,y)$. In addition to their high photon brilliance, modern synchrotron sources offer the unique opportunity to work with a partially coherent X-ray

beam. When we increase the distance between sample and image detector (cf. figure 1) interference patterns show up in the defocused shadow image of the sample. Phase-shift and absorption through the sample are both entangled in such images that are described by the theory of Fresnel propagation.
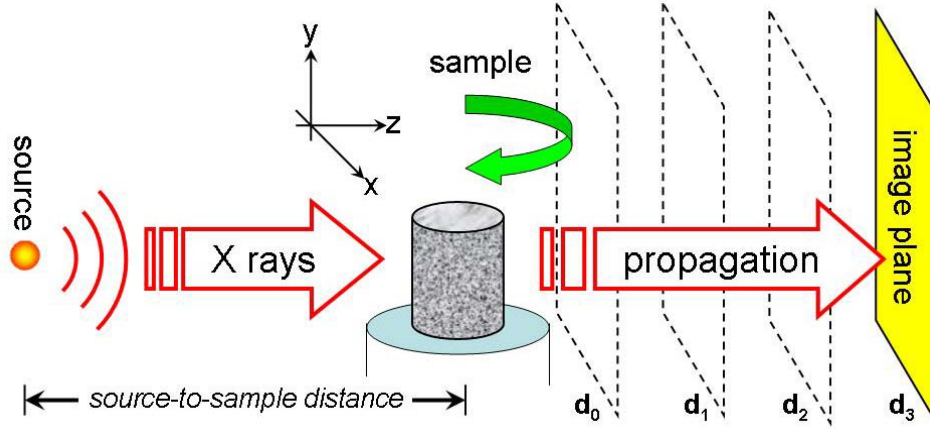

Figure 1. Schematic drawing of Fresnel-propagated micro tomography: X-rays that are transmitted by the sample propagate over a finite sample-to-detector distance allowing the diffracted and the non-diffracted part of the beam to interfere. The results are convoluted intensity images featuring Fresnel fringes that can be combined to numerically retrieve the wave's phase shift through the sample.

Retrieving the phase maps $\phi(x,y)$ from a series of Fresnel-propagated images each image recorded at a different defocusing distance downstream of the sample is the final challenge in our quest for a three-dimensional representation the microstructure. Without going into the details (for overview see [5]) we mention that different solutions to the ill-defined inverse problem of phase retrieval exist, using transfer of intensity equation and/or contrast transfer functions in order to linearize the propagation of intensity and then numerically reverse the process.

1.2 Micro tomography using synchrotron light

The latest achievements in the field of synchrotron tomography are presented in numerous publications [6]. We therefore restrict this text to the description of the particular set-ups that were used to obtain the present results. First experiments were performed at BESSY using the high energy BAM*line*. Recently more experiments have been carried out on the holotomography station at ESRF at beamline ID19. Both locations feature a precision motorized sample-stage and a high-resolution X-ray microscope that can be translated along the beam axis in order to realize Fresnel propagation. In contrast to micro tomography based on lab sources we used a quasi-monochromatic (provided by multilayer monochromators), highly brilliant and almost parallel X-ray beam allowing to image samples with features down to one micrometer in a short time frame and free of artefacts such as beam-hardening. Images at BESSY were recorded with an optical resolution of ~3 *μm* using 25 *keV* photons while the ESRF setup featured a resolution of ~1.5 *μm* using an energy of ~21 *keV*. Note that the term resolution is ambiguous and strongly depends on the definition one uses, e.g. the full-width at half maximum of the detector point-spread function (PSF). However in the general case the PSF is not of Gaussian shape and depends additionally on the photon energy used for each experiment.

2. Results

2.1 The engineering alloy A356

A common aluminium alloy used in thixocasting is A356, referring to the composition AlSi7Mg0.3, where the numbers refer to wt.% of Si and Mg. Samples of commercial A356 feedstock material for thixocasting were investigated both at the ESRF and BESSY. Figure 2 shows cross-sections obtained from absorption tomography (a), Fresnel propagation tomography (b) and holotomography (c) performed at the ESRF. For comparison we show a polished section of the same alloy observed with standard light microscopy (d). Note that the contrast is inversed because the greyscale of the phase map is supposed to represent the differences in the electron density between pure aluminium (dark particles) and (Al + Si) matrix (brighter background). The advantage of holotomography compared to Fresnel-propagated tomography becomes apparent when we attempt to separate the particles (foreground) from the matrix (background).
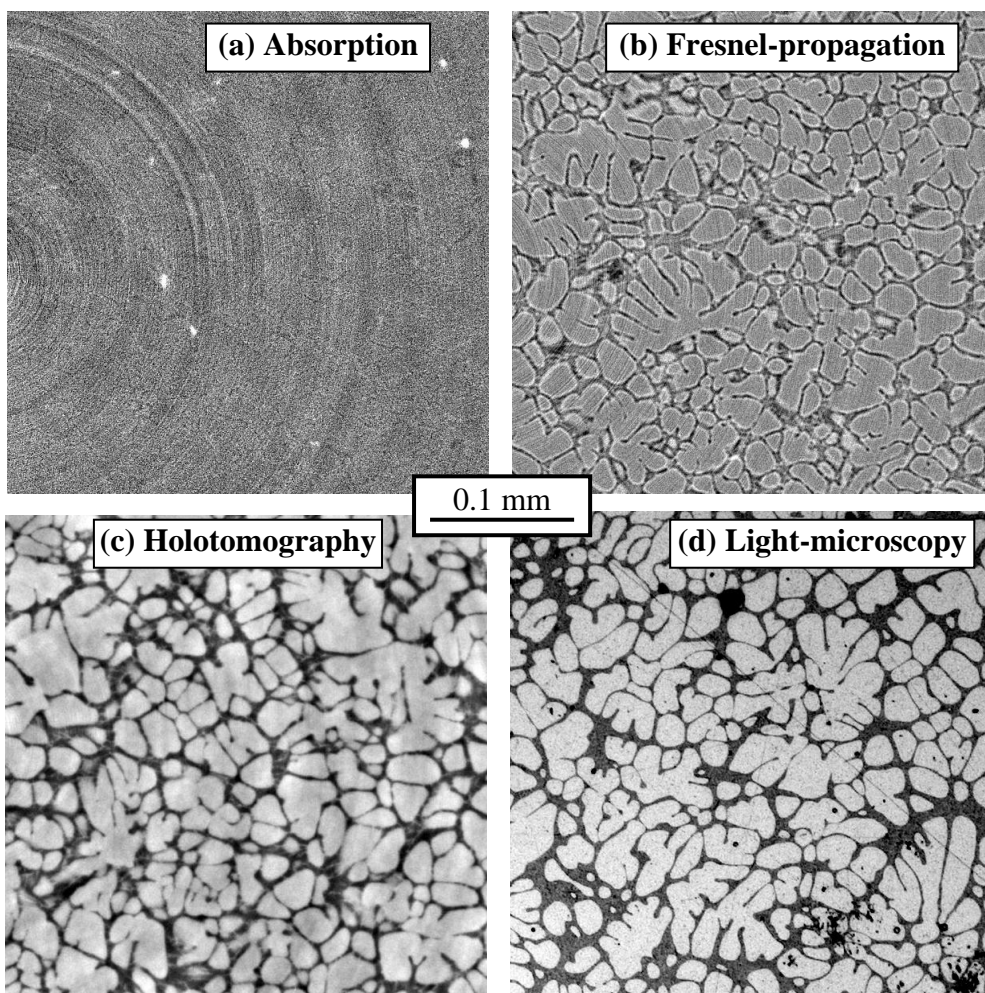

Figure 2. Four equally-scaled views of similar samples of A356 alloy: (a) A cross section of an absorption tomogram. (b) A Fresnel-propagated image of the same sample recorded at a defocusing distance of 0.1 m. (c) A retrieved phase-map using defocused images recorded with the detector placed 8 mm, 34 mm 70 mm and 100 mm downstream of the sample (numerical retrieval was performed at ESRF using an iterative algorithm). (d) For comparison with (c) we show a polished cross-section of the same alloy seen with light microscopy.

While for holotomography the simple application of a threshold is sufficient the Fresnel-propagated image features a strong contrast gradient at the interface between particles and

matrix. Therefore the application of a threshold in figure 2b would fail exactly at this particle-matrix interface. Using phase-retrieval we avoid a complex procedure of particle segmentation but the price we pay is the necessity to use multiple (3-4) propagation distances resulting in longer acquisition and data processing times.

2.2 Thixomolding and rheology of Mg-Al alloys

Magnesium-aluminium alloys such as AZ91 (MgAl9Zn) or AM60 (MgAl6Mn) are widely used in the fabrication of thin-walled light metallic housings (mobile phones, notebooks, etc.) and car components. At NMF GmbH Fürth Germany, AZ91 and AM60 are used for industrial thixomolding while at the same time rheological studies are performed using a high-temperature rheometer. With the latter we can perform shearing experiments under various conditions without the difficulties of changing these for the complex thixomolding process. We use Fresnel-propagated micro tomography in order to study the microstructure of samples extracted both from thixomolded components and from the rheometer. In the rheometer alloys are sheared at a constant rate and specific temperature at which the alloy is partially molten with the remaining solid fraction determined by temperature.
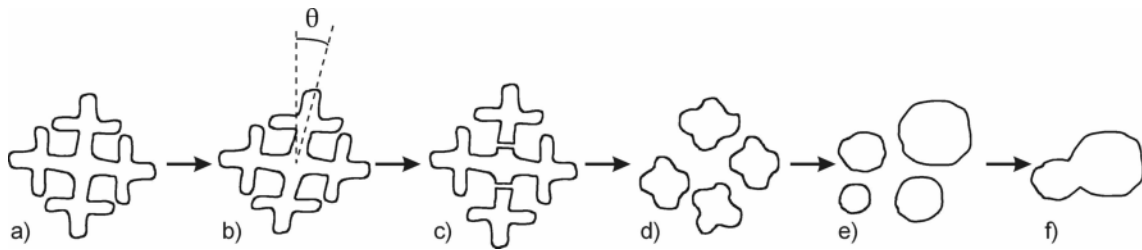

Figure 3. Model of structural changes in the semi-solid alloy that account for the decreasing viscosity when the slurry is sheared at a constant rate and temperature [7]: a. primary dendrite particles, b. shear stress causes first deformation, c. fracture of the primary dendrite arms, d.-f. coarsening and clustering of the solid fraction yielding finally large round particles that are embedded in a matrix.

After a given shearing time the semi-solid alloy is extracted from the rheometer and quenched in water. It is known that during shearing the primary dendrite structure is first deformed and fractured then rounded off due to coarsening and finally the smaller particles can form clusters yielding a primary solid phase of large round particles (cf. figure 3).
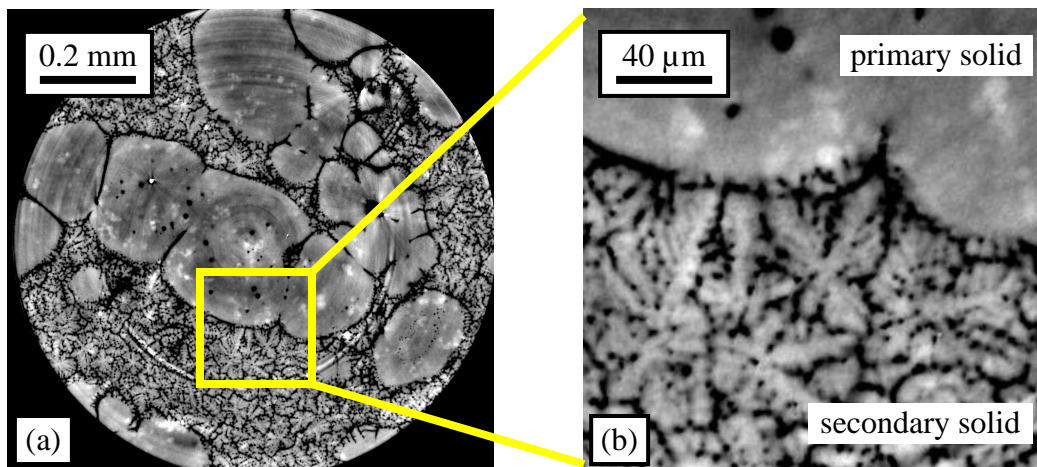

Figure 4. (a) Holotomography of AM60 alloy. Sample quenched after extraction from the rheometer. (b) Even the dendrite structure of the secondary solid magnesium phase is visible after magnification.

These structural changes are supposed to account for a drop of viscosity of the semi-solid slurry by at least one order of magnitude. The long term goal of the three-dimensional

structural analysis is to correlate evolution of microstructure with measured viscosity and determine the influence the experimental parameters (temperature, shear rate, shearing time, solid fraction) have on the final design of the material [8]. Again because of a lack of absorption contrast we apply Fresnel-propagated tomography and holotomography to AZ91 and AM60. Figure 4 features a cross-section of holotomography of AM60 alloy extracted from the rheometer. This time our investigation not only aimed the primary globular phase but in particular the fine-structured secondary solid phase. The mechanical behaviour of the thixomolded components (stiffness, creep resistance, etc.) is known to vary with the structure of this secondary phase and the "inter-phase" between large primary globular particles and secondary (for the case of fig. 4: dendrite-shaped) solid. Magnification of this interesting region (figure 4b) shows the method to apply well for the study of this level of detail. We can now investigate AM60 alloys with different microstructures of the secondary magnesium phase. Figure 5 shows two polished sections (a) from standard and (b) modified AM60 structure observed under the visible light microscope.
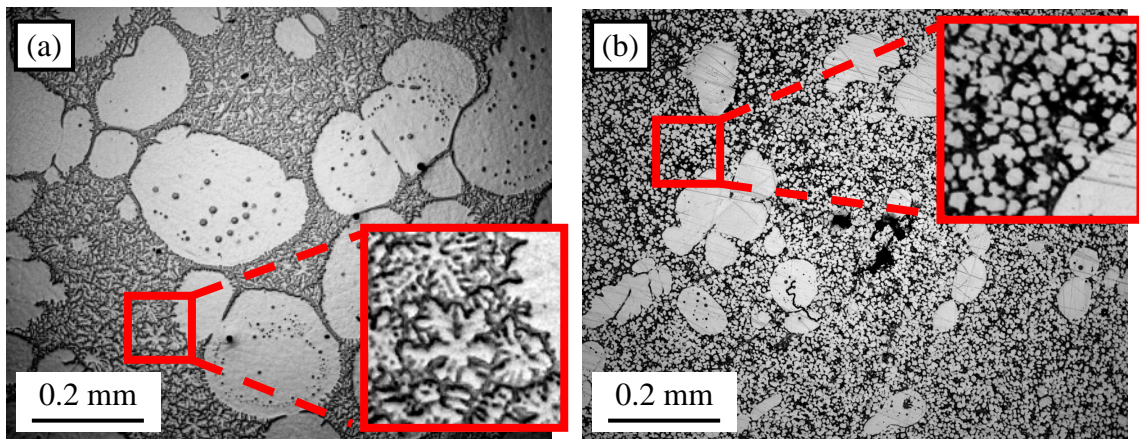

Figure 5. (a) Metallographic section of the same AM60 alloy as shown in figure 4. (b) Cross-section of a modified alloy cut from a thixomolded plate. Note the stark contrast of the secondary phases in (a) and (b).

Another important benefit of three dimensional imaging is illustrated in figure 6 where we chose to display the distribution of the primary globular phase of an AZ91 bulk alloy. Using a Fresnel-propagated tomogram recorded at BESSY (propagation distance: d ~ 370 *mm*) we can directly binarize the light α-magnesium phase and erase the fine structured secondary Mg-phase by application of a size filter to the labelled volume. Prior to the size filtering we used a structural opening based on the Euclidian distance-transform in order to remove the secondary particles connected to the primary phase. The resulting 3D image (figure 6b) reveals what could not be seen from cross-sections: almost all particles are connected and some are still showing a coarse dendritic form.

Image treatment and analysis of three-dimensional data is mostly performed using software from the Fraunhofer institute for industrial mathematics[1]. Tomography back-projection is done by the open-source program PyHST (ref. Mr. Mirone, ESRF software group). Phase-retrieval also employed ESRF software based on octave-code written by P. Cloetens et al.

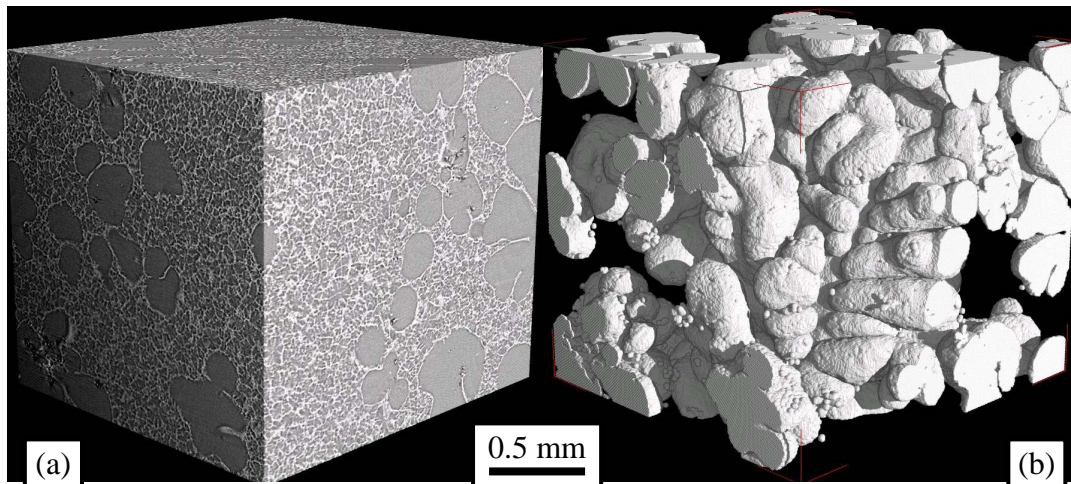[1] Software: Modular Algorithms for Volume Images (MAVI) ©Fraunhofer ITWM, Kaiserslautern Germany

Figure 6. (a) Fresnel-propagated raw data of a AZ91 alloy extracted from the experimental rheometer. The image was recorded at BESSY using a defocalisation distance of d = 370 mm. (b) The same data after binarization, structural opening (based on Euclidian distance transform), labelling and application of a size filter. Only the large particles of the primary solid phase remain while the secondary phase is erased.

3. Discussion

Our work shows an application of Fresnel-propagated micro tomography as well as holotomography to specimens of commercial aluminium-silicon and magnesium-aluminium alloys. Experimental data acquisition as well as phase retrieval algorithms are available and can be used for straight-forward pre-processing of large amounts of data. Using holotomography visible inspection as well as quantitative 3D analysis of the primary solid phase becomes an easy task because globular particles embedded in a matrix can be treated similar to well-studied foam-like structures.

However parameterization and characterization of the fine-structured secondary solid phase present in the magnesium-aluminium alloys has to be accompanied by a detailed study of the alloys mechanical bulk behaviour in order to find the optimum set of structural parameters (shape, size, connectivity, etc.). The wealth of information that can be created by the presented imaging methods will provide a new understanding of semi-solid alloys.

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