Engineering Science at the I12 Beamline at Diamond Light Source

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Synchrotron-based engineering science covers a large field of applications. However, they are all connected in two ways. First, the very synchrotron techniques employed to study the various applications all work in the same way in that they determine structural parameters on the atomic and microscopic scale. Secondly, the portfolio of applications discussed here describes the complete life cycle of an engineering material, starting with processing of the base material—often from the melt—then the characterization of material properties, followed by the forming and joining into components, then component characterization during service, material aging, damage and failure and, finally, recycling or decommissioning. The structural problems which occur during the different stages in the life cycle of a material are complex, due to the advanced material technology of today’s devices. We have created alloys for special applications, compound materials with novel properties, sophisticated bulk and surface treatments, and new forming and joining techniques. We are also concerned with the effect of the material on the environment after it has ended its service.

Much of the structural characterization that contributes to understanding the ways materials behave under real-life conditions can be accomplished using X-ray imaging and diffraction techniques. It is particularly attractive to use hard X-rays with energies above 50 keV, as they offer various advantages. Foremost, penetration of several millimeters through most materials is possible, especially through aluminium alloys, steel, and Ni-based alloys. Even uranium can be imaged up to a thickness of 0.5 mm. Secondly, scattering angles are small, and scattered X-rays are confined within a narrow cone. Using standard area detectors, diffraction and scattering can be recorded at distances of several hundreds of millimeters downstream of the sample. This allows for ample space to install in-situ equipment around the sample. Finally, high-resolution X-ray transmission imaging employing attenuation and phase contrast is also possible at large distances behind the object. Refraction at high energies is very weak and consequently valuable edge-enhancement via in-line phase-contrast does not blur the image as quickly with increased camera distance as for lower X-ray energies.

If we combine these high-energy features with the option of a large beam of high intensity, we have the requisites for a facility that is capable of studying engineering materials and processes over a wide range of applications and with little restrictions in the sample environment. One such facility is beamline I12 at the Diamond Light Source, UK [1]. The beamline can operate in polychromatic (“white beam”) mode or monochromatic mode with a selectable energy between 53 keV and 150 keV. Techniques available to users are radiography, tomography, energy-dispersive diffraction, monochromatic 2D diffraction/scattering, and small-angle X-ray scattering (SAXS). Since commencing operations in November 2009, I12 has been visited by material science and engineering communities and has produced results that cover almost the complete life span of current and novel engineering materials, using a variety of in-situ equipment.

In what follows, we present a cross-section look at the basic results through the eyes of a team of the beamline scientists who facilitated these experiments.

Processing, melting, solidification

Solidification is the direct route from the liquid melt to the engineering component. During the process of cooling, a semi-solid multi-phase material is created, consisting of virgin grains permeated by a liquid fraction. The deformation mechanics of this mush is difficult to understand, as many effects occur concurrently, such as development solid networks between grains, rearranging and shear of individual grains, development of force chains, cracking of grains, opening of internal volumes, and the associated liquid motion [2, 3]. The dynamics of semi-solid materials during deformations, such as tensile [4], compression [5, 6], indentation [3, 7], or extrusion [8], could be studied using in-situ tomography (Figure 1).

But even casting without the application of external deformation is subject to thermo-mechanical processes such as coalescence between solid grains and reduced flow of the liquid phase. Coalescence starts when grains begin to touch each other but are still unable to take tensile loads. It ends at the rigidity temperature where the grains are sufficiently coalesced that the solid phase is able to transmit macroscopic tensile strains. During the ongoing cooling of the cast, this effect can lead to solidification cracking or so-called “hot tearing,” as the rigidity of the structure prevents further thermal contraction [9, 10].

Solidification defects also occur as a consequence of emerging microstructure as the melt undergoes various phase transitions while cooling down. Foremost, the formation of precipitations of a minority phase or contamination of higher brittleness than the matrix—the so-called intermetallic compounds—can decrease the lifetime of a component when they can act as nucleation centers for crack or void
formation. The question if intermetallics appear before voids or vice-versa can be studied using tomography. Some findings support the hypothesis that a pore forms inside a liquid channel that is already surrounded by intermetallics. These particles block the feeding of liquid, thus reducing permeability and accelerating pore growth along their surfaces [11].

The growth of dendritic patterns during alloy solidification is a well-known process. Dendrites initially form at morphological instabilities at the solid-liquid interface and then grow because of a combination of effects related to solute partitioning between the solid and liquid and the thermal undercooling of the system. One example where dendritic structures are obtained in a controlled manner is columnar Ni-based super alloys used for turbine blades which are manufactured by directional solidification [12]. Using time-resolved tomography, it was possible to reveal dynamics of dendrite growth in all three dimensions. The dendritic growth showed higher complexity than previously observed in radiographic studies, where growth is constrained to effectively two dimensions between thin plates (Figure 2).

Figure 1: Semi-solid indentation of globular microstructures: (a) schematic of the setup; (b–d) sequence of 2D longitudinal slices, with the indenter colored red, showing the onset and development of transgranular cracking of globular microstructures at 555°C during indentation with a speed of 2 µm/s (corresponding to the instances marked in f); (e) final room temperature scan; (f) load measurement (averaged); (g–j) corresponding 3D sectional views of segmented solid grains. The yellow dashed circles in b–e and the black circles in g and h highlight regions of interest where transgranular fracture of seemingly ductile grains is occurring. The blue circle indicates the region of apparent increase in liquid after indentation. Scale bar, 500 μm (from [3]).

Figure 2: (a to d) 3D morphology of single dendrite (SD) with side branches (red arrows) at t = 207, 261, 306, and 441 s, respectively (from [12]).
Material characterization

Characterization of material properties at I12 has been carried out exploiting powder diffraction and tomography. We highlight a few studies which exemplify the advantages of high-energy synchrotron radiation, both in diffraction and imaging.

Engineering alloys—for example, those based upon Al, Fe, Ni, and Ti—rely upon a phase transition to give optimized properties. For a special class of nickel-based alloys—the so-called super-alloys—one uses an ordering reaction to transform the matrix partially into precipitates made up of compounds of Ni with Al, Ti, Nb, or Ta (γ′-phase). Dislocations cannot enter the precipitates easily; planar defects, such as anti-phase boundaries and complex stacking faults, arise if they do. These microstructural faults result in a considerable strengthening of the material. This is the primary source of the excellent high-temperature performance of these super alloys [13]. They are much used in the hot section of turbine engines, where they operate close to the melting temperature $T_m$ (above 0.9 $T_m$).

Using high-energy X-ray diffraction, Collins et al. [13] studied the phase transitions of several Ni-base super-alloys while cooling from above the solution temperature of the γ′-phase (1180°C). Although the experimental setup is simple, with the recorded patterns and a subsequent diffraction analysis conducted from first principles, the team was able to display the kinetics of the complete phase transitions in a set of super-alloys in great detail and with high accuracy (Figure 3), revealing volume fractions and lattice mismatch.

Another example of the power of high-energy diffraction is the study by Jones et al. on the elasto-plastic behavior of a certain type of carbides (Ti$_x$SiC$_2$), where the deformation of grains within the material is either elastic or undergoes deformation along slip planes, depending on the grain orientation in respect to loading direction [14]. Although the experimental arrangement is simple, the access to a large section of the reciprocal space gave the authors the possibility to look at different crystal lattice planes in different loading directions simultaneously. By this, they could see elastic and plastic deformation of different grains at the same time.

It is not a surprise that diffraction has a key role in determining dynamic structural parameters like strain, phase transitions, texture, volume fractions, and so on. Imaging, on the other hand, can capture structural features on the microscopic level where different microstructural features show up, like cracks, voids and grain- and phase-boundaries. The technique is also capable of determining strain; here, one needs to record the complete topology (2D or 3D) of the object dynamically throughout the deformation process and calculate the local displacement field (2D or 3D) by comparing incremental image sets. The technique has been developed for visible light—called digital image correlation (DIC)—and can be applied to X-ray imaging or three-dimensional tomography data using the same concept (digital volume correlation or DVC).

Vertyagina et al. [15] undertook a study of the behavior of hard, brittle ceramics under load of a conical indenter pressing into its surface, similar to what is used for the measurement of Vicker’s hardness. Understanding the observed indentation cracking of the structural ceramic under study is important, since the resulting damage mimics the type of wear these materials are suffering. Applying DCV analysis to the acquired in-situ tomography data revealed, for the first time, the magnitude of cracks along the different orientations in respect to the indenter. Shear components of indentation cracks and opening displacements of approximately 1 μm could be determined, as well as lateral cracks of the same magnitude.

SiC-SiC ceramic matrix composites are candidate materials for fuel cladding in Generation-IV nuclear fission reactors. In-situ observation of mechanical damage within a SiC-SiC tube has been undertaken by Saucedo-Mora et al. [16]. Applying DVC to a set of tomographic data during axial tensile loading of the tube exposed the displacement (Figure 4), which was then used to calculate the strain field in the specimen under different load conditions.

Forming, welding, joining

Controlled deformation, welding, and other methods of joining are ways to shape parts of components during the manufacturing process. These methods cause severe transformations to the material’s micro-
structure, either due to thermal treatment, plastic deformation, or a combination of both.

Collins et al. [17] devised an experiment where a sheet of automotive steel can be deformed in two perpendicular directions at different rates. This biaxial deformation mimics the process during bodywork manufacturing much closer than the uniaxial loading geometry mostly applied in academic research. The experiment revealed that the accumulation of lattice strain during deformation, as a function of azimuthal angle, is highly sensitive to strain path. For a ratio between tensile and radial strain of 1:1.5, while lattice strain accumulates most rapidly in the tensile direction during early stages of plastic deformation, the lattice strain is shown to distribute almost perfectly isotropically for the observed orientations when plastic strain is high. This was found to be in contrast to strain paths where the ratio was smaller than 1.5, demonstrating that lattice strain magnitudes remain highest in the direction parallel to the tensile axis with the highest applied load (Figure 5).

During the hot forming stages of the steel-making process, the dominant mechanism of damage nucleation and evolution can vary, depending on a number of parameters, including strain rate, forming temperature, material composition, and microstructure [18]. One very significant industrial case is the hot forming of free-cutting steels (FCS). Here, the presence of inclusions which improve machinability also promotes damage at high temperatures by acting as void nucleation sites and stress raisers. Puncreobutr et al. employed in-situ tomography during tensile testing of FCS at a temperature of 1000°C to quantitatively measure the occurrence, distribution, and coalescence of different classes of damage, namely internal and surface connected voids. Side-by-side FEA calculations were used to correlate the void distribution with the mechanical strain during deformation [18, 19].

Linear friction welding (LFW) is a welding method that exploits the heat and plastic deformation materials are subjected to during the process of fast periodic motion under compressive forces. The microstructure of LFW is characterized similarly to other welding methods by a thermo-mechanically affected zone (TMAZ) close to the weld line, a heat affected zone (HAZ) further from the weld line, and the unaffected parent material microstructure further still [20]. Song et al. used X-ray diffraction measurements to determine the two in-plane strain components, which they used to validate a thermo-mechanical model and derive an analytical function which can describe the residual stresses for general FLW geometries [20].

Operation, component and device characterization

The two examples of characterization of operational components we cover here both exploit a rarely used, yet powerful, in-situ diffraction technique. The stroboscopic EDXD method we employ on I12 can measure strain components perpendicular to the incoming beam direction and, at the same time, offer time resolution of several 10 microseconds for a periodic process. This technique relies on the geometry and characteristics of the multi-element Ge-detector in use. The detector has 23 individual Ge-detection crystals arranged in a semi-annular ar-
ray with an azimuthal angle-coverage of 0° to 180°; this makes up 11 orthogonal element pairs in total (plus one additional element) and allows the simultaneous measurement of 11 sets of orthogonal q-vector components. The detector amplifiers can be synchronously gated in phase with an external periodic process so that diffraction signals can be added up in a stroboscopic manner at high signal-to-noise ratio, as the detector itself is practically noise-free.

Baimpas et al. [21] used this technique to determine the elastic strain in the connecting rod of a combustion engine during operation (Figure 6). The exposure time for each revolution was 0.3 ms, with total accumulated exposure time amounting to 24 s during 80 min overall engine running time. The accuracy of determining the peak center position was better than 40 ppm, yielding a figure of the strain in the con-rod with the engine running at ~2000 rpm to be compressive at 630 microstrain (630 × 10⁻⁶).

Stroboscopic EDXD has also been used by Mostafavi et al. [22] to obtain, for the first time, the strain measurement of a ball bearing under dynamic contact. The main aims in this study were to measure the...
dynamic strain that the ball exerts on the outer raceway of the bearing while the bearing was rotating and compare it to the static strain that a ball exerts on the outer raceway when it is stationary.

**Damage during operation, crack, corrosion, fatigue**

Crack development and subsequent propagation during fatigue are common types of damage that occur during operation of periodically loaded components such as air foils or compressor disks of aeroengines. To mimic the fatigue life of such a component in the lab, low-cycle fatiguing (LCF) is applied, where, at each cycle, the material undergoes severe plastic deformation. Chapman et al. [23] used in-situ tomography to image the crack growth of an aero grade Ti-alloy during LCF testing. In all instances of crack initiation, the mechanism on the fracture surface was by facet formation, on the scale of primary grain size (15–50 µm diameter). Crack initiation and crack growth rates have been extracted quantitatively under different conditions of atmosphere.

Fracture behavior on a quasi-brittle material is driven by a different process than for a ductile sample, such as the previously discussed Ti-alloy. During the fracture of a partially cracked component made from a quasi-brittle material, a fracture process zone develops due to distributed micro-cracking ahead of the crack tip [24]. In-situ tomography, combined with DVC on a poly-granular graphite sample, has been carried out to measure the 3D displacements in the cracked specimen during fracture. The crack-opening displacements, where quantitatively mapped and allowed the authors to measure the extent of the damaged zone ahead of the crack-tip. The results could be simulated using a cohesive-zone model where one essentially needs to overcome a critical energy in the damaged zone to open the crack further.

Davenport et al. [25] studied atmospheric corrosion of stainless steel, which is used as container material for radioactive waste of intermediate level. The initial corrosion attack is likely to be in the form of pits, and there is concern that stress corrosion cracking may develop from these pits. It is therefore important to inform existing simulations for the development of long-term damage with experimental data for the kinetics for the formation of corrosion-initiating pits. In particular, the influence of relative humidity needs to be properly accounted for. The authors use in-situ tomography to study the effect of alloy microstructure and humidity fluctuation on the development of corrosion pits. The results were used to refine models of corrosion propagation, which are currently under development.

**Decommissioning and recycling**

At the end of their lifetime, some materials can be recycled to enter again the cycle of processing, manufacturing, and so on. Due to the difficulty of complete material separation of a multi-element component, there is an issue of contamination and, consequently, the material properties of the recycled base material will differ from the initial properties. The influence of recycled-grade Al base material on the microstructure of Al-alloy casts has been discussed earlier [11].

Uranium metal from reprocessing of spent nuclear fuel is a contributor to nuclear waste of intermediate level (intermediate-level waste: ILW). For its long-term storage, processes of U-oxidation in the vicinity of cladding material need to be well-understood. During corrosion, metal oxides and hydrogen gas are produced. As hydrogen accumulates, the corrosion of uranium may switch to produce uranium hydride (UH₃) instead. This pyrophoric compound reacts vigorously with oxygen to make UO₂ and, as such, is thought to exist only fleetingly. Stitt et al. [26, 27] investigated the risk posed by an accumulation of pyrophoric uranium hydride. For these experiments, UH₃ was stored underwater after being artificially formed on a uranium rod and encased in the grout used at nuclear waste facilities. The sample underwent periodic testing at 0, 3, and 10 months, employing tomography to reveal the topological changes at the corrosion surface (Figure 8) and diffraction to specify the corrosion product, with the U-slab remaining inside the original grout.
cladding. The findings are significant for ILW packaging. They suggest potential for generation of hydrogen—a flammable gas and precursor to uranium hydride formation—by corrosion of uranium and other reactive metals in grouted ILW and its accumulation in the waste packages.

After this short review of work accomplished over several years by the many users of beamline I12, we can say that engineering science is a challenging field, as rich and complex as fundamental sciences and with one particular feature: it is engineering that enables our lifestyle.

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