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Enabling high resolution strain mapping in zirconium alloys

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ABSTRACT

High Resolution Digital Image Correlation (HRDIC) has been established recently as a novel displacement mapping technique during mechanical loading experiments to quantify strain localisation down to the level of individual slip traces. This is achieved by the creation of a nano-scale gold pattern naturally formed during remodelling of a thin gold layer that was sputtered onto the region of interest. To date, the gold remodelling is carried out in a water vapourisation environment, which excludes the technique to be applied to materials that readily form noticeable oxide layers in such environments. The current paper describes a recently developed gold remodelling technique using a styrene-argon environment at substantially lower temperatures than the water-vapour based technique. The material used in the present work is a zirconium alloy where we first demonstrate the problem of oxide formation during remodelling in water vapour and the benefit of the modified remodelling procedure. The error associated with the spatial drift was assessed for different interrogation window sizes followed by detailed analysis of the subsequent strain maps produced using the styrene remodelled patterns after tensile deformation to nominal applied strains of ~3.5% and ~7.0%. The level of detail captured demonstrate the suitability of styrene-argon-based remodelling for materials like Zr alloys with the strain maps showing clear strain patterning on both a transgranular and single grain scale with the possibility of quantifying strain across a single slip trace.

1. Introduction

Digital Image Correlation (DIC) is a widely used technique to measure full-field surface displacement and hence produce strain maps at spatial resolutions from the macro to the nano scale [1–13]. The spatial resolution is dependent on the speckle pattern that is applied to the surface of the sample and recently Dong and Pan [14] provided an overview of the different speckle patterns for different spatial resolutions and the fabrication techniques used to apply them, along with an assessment of the methods with the aim of providing practical guidelines for DIC users. While DIC has long been established as a technique to determine macroscopic 2D strain fields [16,17] recent advances in High Resolution Digital Image Correlation (HRDIC) technique has opened up quantification of heterogeneous deformation from slip band formation [17]. In principle, the measurement of local shear strain enables one to assess and compare the true level of strain heterogeneity for different alloy chemistries, microstructures and the effect of microstructure inhomogeneity. Previous studies using optically based DIC have focussed on strain localisation at the microscale, reporting maximum strain concentrations ranging between 2 and 3.5 times the average macroscopic strain [18,19]. Di Gioacchino and Quinta da Fonseca [4] developed a high spatial resolution strain mapping methodology using SEM imaging in combination with DIC that provides detailed understanding of the deformation behaviour at the sub-grain scale through quantification of the shear strain within individual slip traces, leading to a much more detailed characterisation of the strain heterogeneity. The dramatic improvement in spatial resolution demonstrated that maximum shear strain concentrations are usually 10–20 times greater than the macroscopic strain [20]. In addition, recording the shear strain associated with slip traces and combining this information with EBSD-based orientation imaging enables one now to undertake truly quantitative slip trace analysis.

The gold remodelling technique for applying a speckle pattern to the surface of a material was first developed by Luo et al. [21] on glass slides, by flowing nitrogen gas through an iodobenzene reservoir and over thin gold films to form nano-sized gold islands. The technique was further developed by Di Gioacchino and Quinta Da Fonseca [20] for HRDIC studies to observe the plastic deformation patterns on the

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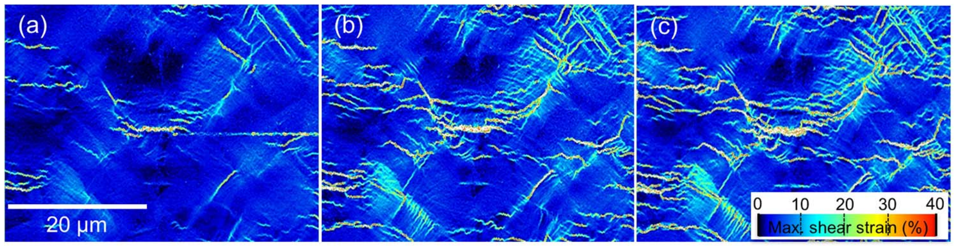

Fig. 1. Maximum shear strain map for Zircaloy-4 after speckle pattern formation using gold remodelling in a water vapourisation environment at (a) 4.9%, (b) 8.2% and (c) 9.5% compression.

surface of metals. In this case, a thin gold film deposited on flat mechanical test samples is exposed to water vapour at 300 °C for 90 min, which results in the formation of a nanoscale speckle pattern with well-defined features that conserve their shape during plastic deformation. During exposure to heat and water vapour, the thin gold film de-wets and forms either distinct speckles or mesh-like pattern morphologies that provide high contrast compared to the substrate when imaged by Backscatter Electron Imaging (BEI) in a SEM.

However, there are a number of materials, which readily form either protective or non-protective oxides in high temperature steam environments. Zirconium, a material used by the nuclear industry for fuel cladding due to its low neutron cross-section [22], has a particularly high affinity to oxygen and readily forms a protective oxide layer of 1–2 µm in high temperature steam [23–25]. As a result, previous attempts to use the water vapourisation gold remodelling technique for HRDIC in a zirconium alloy (Zircaloy-4) have proved unsuccessful. The remodelling process created an oxide layer on the surface of the sample, that masked the underlying plasticity, with the subsequent strain maps demonstrating oxide cracking, Fig. 1.

Quantifying the level of strain localisation is important for zirconium alloys for a number of reasons. Firstly, the low temperature phase of zirconium has a hexagonal-close packed (hcp) crystal structure, and like titanium and magnesium alloys, can deform on a number of different slip systems, which makes plasticity studies particularly interesting. The ease of slip has a strong dependency on the crystallographic orientation with prismatic ⟨a⟩ slip reported to be the easiest slip mode in zirconium compared to basal ⟨a⟩ slip [26] and the more difficult to activate slip modes that contain a ⟨c⟩ component [27–29]. Secondly, the primary application of zirconium alloys (structural material for nuclear fuel assemblies) means that they are subjected to neutron irradiation, which is known to affect deformation mechanisms and lead to severe slip localisation [30–34].

The deformation of zirconium alloys has previously been studied at the micro-scale using both DIC of optical images [3] and SEM images after pattern application using the micro-lithography technique [7]. Héripré et al. [7] found local strain concentrations of 4 times the applied strain and high strain localisation at boundaries between grains with low and high Schmid factors for easy slip during uniaxial tension experiments on zirconium grade 702. Similar findings were reported by Padilla et al. [3] during compression testing revealing strain hot spots that were correlated with grains favourably oriented for easily activated prismatic slip. Strain patterning was visible at both length scales [3,7], with distinct low and high strain bands at ± 45° to the loading direction. It is important to note that these strain measurements were at a maximum spatial resolution of 2 µm, limiting sub-grain information.

In the present study, we have optimised an alternative method for remodelling a thin gold film based on the use of styrene [35] as a remodelling reagent in order to avoid oxide formation during remodelling. The suitability of the method for use for 2D strain localisation mapping during plastic deformation using HRDIC is assessed in a zirconium alloy, where previous attempts using water vapour gold remodelling have been unable to resolve fine-scale deformation.

2. Experimental Procedure

2.1. Material and Mechanical Loading

The gold remodelling and HRDIC–based slip localisation studies were carried out on the nuclear-grade zirconium alloy, ZIRLO™ (Zr-0.9Nb-0.9Sn-0.1Fe). The recrystallised ZIRLO™ sheet material was provided by Westinghouse. Tensile specimens for mechanical testing and speckle characterisation were machined from 0.7 mm thick sheet material by electric discharge machining. The tensile specimens had a flat dog bone geometry with a 26 mm gauge length and 3 mm gauge width. The surface for gold remodelling was prepared by grinding and polishing to #800 grit paper, followed by diamond polishing to 1 µm and subsequent hand polishing for 30 min on an OPS cloth in a solution of 4:1 OPS to hydrogen peroxide. After polishing, a series of micro hardness indents were made on the surface of the specimen to act as fiducial markers and subsequently mapped by EBSD on a FEI Quanta 650 Field Emission Gun (FEG)-SEM equipped with an Aztec EBSD system and a Nordlys II detector. EBSD was performed at an accelerating voltage of 20 kV. An area of $0.25 \times 0.25\ mm^2$ was analysed with a step size of 0.5 µm to provide a sufficiently detailed grain orientation map. The data (confidence index > 0.1) were analysed using HKL Channel 5™ software [36]. Subsequently, styrene remodelling was carried out before tensile loading the specimen incrementally with strain rate of about $6.4 \times 10^{-5} s^{-1}$ using a Kammrath-Weiss 5 kN Tension-Compression microtester. As the working distance for optimal imaging conditions in the SEM was < 6 mm and the microtester requires a larger working distance when installed inside the microscope, the experiments were carried out ex-situ. Hence, after each deformation step the sample was removed from the microtester and mounted in the SEM for image acquisition.

2.2. Application of Speckle Pattern

A thin gold layer of 25–40 nm was deposited on to the surface of the sample using an Edwards S150B sputter coater at a deposition rate of 5–8 nm/min. The parameters for the sample remodelled in water vapour (Fig. 1) were 200 °C for 3 h using the method detailed in [4]. The alternative methodology suitable for temperature sensitive/readily oxide forming materials was developed using styrene as an alternative solvent to water vapour and has been proven to be successful for HRDIC on a magnesium alloy [35]. This method requires the remodelling chamber to be isolated to remove oxygen from the process. Once the gold layer has been deposited onto the polished sample surfaces, the samples are placed into the chamber shown schematically in Fig. 2. The remodelling process begins by initially flowing argon gas through a styrene reservoir, in turn creating a mixture of argon and styrene vapour. A hot plate, which has a flat surface to provide a homogeneous temperature distribution across the sample, is positioned in the centre of the remodelling chamber. The temperature can be monitored and adjusted using an external control unit. The Argon-Styrene flow is passed directly over the sample surface and subsequently remodels the gold layer into fine speckles. The chosen styrene remodelling