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Which resolution can be achieved in practice in neutron imaging experiments? – A general view and application on the Zr - ZrH_2 and ZrO_2 - ZrN systems

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Abstract

Current methodical developments improve the spatial resolution of neutron imaging facilities. Objects with dimensions down to several microns should be detectable. However, the minimum object size detectable depends not only on the facility hardware like detector resolution or neutron optics, but also on the attenuation contrast. In this paper the relation between illumination time needed, neutron contrast of the objects and their minimal size detectable is derived and an analysis of the minimal dimension of an object can be detected in neutron radiography and tomography is discussed at two examples: zirconium hydride ZrH_2 in Zircaloy-4 as a high contrast system and zirconium nitride ZrN in zirconium oxide ZrO_2 as a low contrast system. It is concluded which minimal sizes of the precipitates can be detected in realistic times.

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Keywords: neutron imaging, resolution, Zircaloy-4, zirconium hydride, zirconium nitride, zirconium oxide

1. Introduction

Precipitates with dimensions in the order of magnitude of 1 to 10 µm play important roles for material properties. Zirconium alloys are applied e.g. for nuclear fuel claddings or pressure tubes. Under operation, accidents or long term storage conditions their mechanical and chemical properties are strongly influenced by two characteristic

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micro-structural features: The formation of nitrides in the oxide layer on zirconium alloy strongly accelerates the further oxidation kinetics. The nitrides can be formed if zirconium reacts in air or steam-nitrogen atmospheres. It can happen in spent fuel accidents or in the late phase of a severe reactor accident after failure of the pressure vessel. Fig. 1 gives an example of a typical ZrO_2/ZrN microstructure near the metal-oxide interface. The sizes of these nitrides are several micrometers. Another example is zirconium hydride in zirconium alloys (see Fig. 2). Hydrogen is absorbed by zirconium alloys as result of corrosion under operation conditions or by the reaction with steam at high temperatures under accident conditions. Typical dimensions of these rod shaped precipitates are 1 - 2 µm thickness and 10 - 20 µm length. They are decreasing the fracture toughness of the metal and provide the risk of the so called delayed hydride cracking and/or the fragmentation of the fuel rods during emergency cooling of an overheated reactor, both resulting in fission gas release and redistribution of fuel. Usual these hydrides are oriented in circumferential direction. Under certain stress conditions the can be re-oriented in radial direction. In this case they degrade the mechanical properties much stronger.

Neutron imaging is used by several groups to study the hydrogen in zirconium (for instance Yasuda et al. 2002, Lehmann et al. 2003, Grosse et al. 2008, Grosse et al. 2011, Wang et al. 2013, Smith et al. 2015, Tremsin et al, 2015) . Currently, nominal spatial resolutions of typical 25 to 80 µm were used for these investigations (pixel sizes between 13.6 and 40 µm). Integral information about the hydrogen content averaged over the corresponding gauge volume was obtained. Fig. 3 shows as an example the 3D reconstruction of the hydrogen distribution measured in a zirconium ally cladding tube exposed to a loss of coolant simulation test.. It can clearly be seen that the cladding tube brokes at a hydrogen enrichment. However, for deeper understanding of the precipitation, re-solution and re-orientation processes, knowledge about formation, amount, re-distribution and re-orientation of the individual precipitates in the bulk of the materials at the temperatures at which it occurs would be very helpful. In-situ neutron imaging with high spatial resolution could provide this information because of the penetration depth of neutrons in matter allowing dedicated sample environments.

However, currently the spatial resolutions of these methods which are in the order of 20 µm are not yet sufficient. Actually, a lot of efforts are made at several neutron imaging beamlines to improve the spatial resolution of the method (e.g. Tremsin et al. 2011, Hussey et al. 2015, Trtik et al. 2015, Bingham et al. 2015, Trtik et al. 2016a). Spatial resolution down to 1 µm seems to be reachable. The application of neutron optics together with high resolution detector systems seems to allow reducing the spatial resolution even below the 1 µm limit.

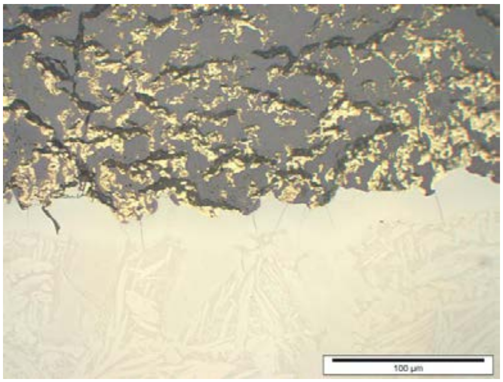

Fig. 1. Optical microstructure of the oxide layer on Zry-4 after 40 min reaction in 20 % steam + 80 % N_2 atmosphere at 1100°C (yellow: ZrN, bright gray: Zry-4, dark gray: ZrO_2, black: micro-cracks).

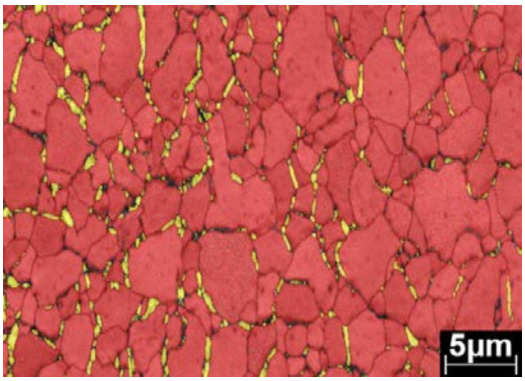

Fig. 2. EBSD pattern of a hydrided Zircaloy-4 sample (yellow: Zirconium hydrides (yellow), red: Zircaloy-4 (Pshenichnikov et al. 2016)

On the other hand, resolution in the neutron imaging experiment is a multidimensional parameter (two or three lateral dimensions for radiography or tomography, respectively, resolution of contrast, time and neutron wavelength) as mentioned in (Griesche et al. 2016). These parameters cannot be varied independently. A decrease of the minimal size detectable by neutron imaging is penalized by an increase in the illumination time per radiograph and/or a