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Void growth and coalescence in irradiated copper under deformation

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A B S T R A C T

A decrease of fracture toughness of irradiated materials is usually observed, as reported for austenitic stainless steels in Light Water Reactors (LWRs) or copper alloys for fusion applications. For a wide range of applications (*e.g.* structural steels irradiated at low homologous temperature), void growth and coalescence fracture mechanism has been shown to be still predominant. As a consequence, a comprehensive study of the effects of irradiation-induced hardening mechanisms on void growth and coalescence in irradiated materials is required. The effects of irradiation on ductile fracture mechanisms - void growth to coalescence - are assessed in this study based on model experiments. Pure copper thin tensile samples have been irradiated with protons up to 0.01 dpa. Micron-scale holes drilled through the thickness of these samples subjected to uniaxial loading conditions allow a detailed description of void growth and coalescence. In this study, experimental data show that physical mechanisms of micron-scale void growth and coalescence are similar between the unirradiated and irradiated copper. However, an acceleration of void growth is observed in the later case, resulting in earlier coalescence, which is consistent with the decrease of fracture toughness reported in irradiated materials. These results are qualitatively reproduced with numerical simulations accounting for irradiation macroscopic hardening and decrease of strain-hardening capability.

1. Introduction

Structural materials used for fission reactor cores [1] (or selected for ITER fusion reactor [2]) are subjected to high energy neutron irradiation and high irradiation dose, leading to significant evolutions of mechanical properties related to the creation of irradiation defects in the microstructure. 300 series austenitic Stainless Steels (SS) are used for Light Water Reactors (LWR) core internals, and are also foreseen for first wall/blanket and divertor of ITER fusion reactor. For the latter, copper alloys are also considered. Fracture toughness of these materials and its evolution with irradiation are required for design purposes, but also for ageing management, as experimental studies have shown a strong decrease of toughness with irradiation. Reviews of the degradation of austenitic stainless steels toughness with irradiation under LWR conditions can be found in Refs. [3–5]. A decrease of toughness (as measured through initiation energy release rate J_{Ic}) up to a factor ten is observed after a few dpa. Fracture surfaces of unirradiated SS exhibit transgranular dimples, indicating void nucleation [6,7], growth [8,9] and coalescence mechanisms [10]. Classically, voids nucleate by cracking or decohesion of inclusions or second-phase particles, then grow due to the plastic flow of the matrix material around them until strong interactions between adjacents voids appear, which correspond to the coalescence phase. Details about these mechanisms can be found elsewhere [11]. It should be noted that void growth and coalescence of concern here is due to plastic flow under mechanical loading post to irradiation, which is a mechanism clearly different from void growth from vacancy condensation appearing under irradiation and known as swelling [12,13]. Void growth and coalescence is still the predominant fracture mechanism of austenitic stainless steels irradiated in LWR conditions. Another mode of fracture - known as channel fracture - has also been reported for these steels under specific conditions (higher irradiation temperatures) but is not considered in this study [14]. Regarding fusion applications, lower irradiation temperatures and doses are considered, leading to less pronounced decrease of fracture toughness of austenitic stainless steels [15]. Materials selection of ITER reactor has required assessing in particular the fracture toughness of pure copper and copper alloys, irradiated and tested at relatively low temperatures (below 300 °C). Copper alloys have been recently selected to be used in the final ITER blanket system.

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Significant hardening (Fig. 1) and reduction of uniform elongation for low doses are observed for pure copper and copper alloys, as a result of the production of irradiation defects such as dislocation loops and Stacking Fault Tetrahedra (SFT). Fracture toughness of copper alloys has been shown to depend on temperature, and the decrease with irradiation strongly depends on alloying elements: significant decrease was observed for CuAl25 while no significant effect was observed for CuCrZr at low testing temperatures [16,17]. Fractographic observations [17,18] indicate that room temperature fracture mechanisms involved microvoid coalescence. A comprehensive review of mechanical properties of unirradiated and irradiated copper and copper alloys can be found in Ref. [19].

Empirical bounding curves of fracture toughness have been defined for engineering purposes (see Ref. [4] for irradiated austenitic stainless steels). Correlation of fracture toughness with tensile properties (evolution of yield stress and uniform elongation with irradiation) has also been proposed [20], while reduction of uniform (or total) elongation measured on tensile tests does not correlate in general with reduction of fracture toughness. Physically-based models of ductile fracture through void growth and coalescence are now widely used for unirradiated materials, following seminal contributions of McClintock [8] and Rice and Tracey [9] describing the behavior of voids under mechanical loading, and Gurson [21] and Thomason [22] homogenized models regarding void growth and coalescence, respectively. Recent reviews of these models can be found in Refs. [11,23,24]. Assuming that deformation mechanisms are similar (which hold true for low irradiation dose), such models can a priori be applied to irradiated materials, and decrease of fracture toughness J_{Ic} with irradiation can be rationalized as resulting from loss of strain-hardening capability (decreasing J_{Ic}) and hardening (increasing J_{Ic}) [25], the former effect being dominant. Applications of these physically-based models to irradiated materials is more limited. Early models have been proposed in Refs. [20,26] assuming microvoid coalescence. Physically-based models have been recently described (see Refs. [14,27] and references therein) to describe fracture toughness of irradiated austenitic stainless steels, accounting for various phenomena such as void initiation, growth and coalescence, channelling, irradiation-induced nanovoids [12]. More recently, void growth and coalescence has been assessed numerically [28] at the crystal scale using physically-based constitutive equations developed for irradiated stainless steel [29], showing accelerated growth and coalescence with irradiation.

Physically-based models aiming at predicting fracture toughness of irradiated materials [14,27] assume some physical fracture mechanisms which need validation through dedicated experimental observations. In particular, irradiation of austenitic stainless steels and copper alloys leads to a change at the crystal scale of deformation mechanisms from an homogeneous to heterogeneous one: dislocations sweep away irradiation defects in narrow channels, making subsequent motion of dislocations easier. Assessing the effects of these irradiation-hardening mechanisms on void growth and coalescence is therefore required. The objective of this study is thus to assess experimentally void growth and coalescence in an irradiated material. Pure copper has been selected as a model FCC material to describe austenitic stainless steels for LWR applications (both sharing similar evolution of mechanical properties with irradiation). Moreover, void growth and coalescence in irradiated copper is relevant for fusion applications. Section 2 describes the material and methods used in this study. Analytical models are also presented. Section 3 details the experimental and numerical results, that are discussed in Section 4.

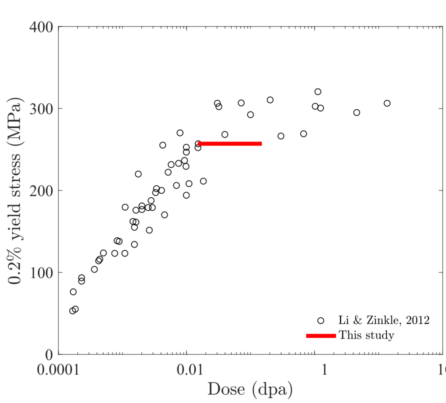

Fig. 1. Evolution of conventional 0.2% yield stress of pure copper with dose (data taken from Ref. [19] and references therein). The red line corresponds to this study considering the range of dose in the depth of the irradiated layer (see Section 2.2). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2. Material and methods

2.1. Material and irradiation

In addition to being relevant for fusion applications [2], pure copper has been used in this study as a model FCC material. Compared to austenitic stainless steels more relevant for fission applications (LWR internals structure, FBR claddings), copper has also a Face-Centered Cubic crystallographic structure, shows a high sensibility to irradiation with significant hardening for low doses, and saturation of mechanical properties below 0.1 dpa (compared to few dpa for SS), and high thermal conductivity [19]. The two last properties simplify irradiation with ions, the former by reducing irradiation time, the latter by allowing to achieve high flux (and therefore reducing also irradiation time) with a good monitoring of temperature. 75 μm foils have been supplied by Goodfellow® with a typical chemical composition: Cu $>99.9\%$, Ag 500 ppm, O 400 ppm, Bi <10 ppm, Pb <50 ppm, other metals <300 ppm. Initially in an hardened state, foils have been heat-treated (200 °C, 30 min, air cooling) to restore some ductility. The heat-treatment conditions are a compromise between ease of use (to manipulate tensile samples without damaging them) and ductility (to get homogeneous plastic strain (up to few percents) along the gauge length of tensile samples, *i.e.* before necking). Electron Back Scattered Diffraction (EBSD) revealed that the material shows no texture, with a significant number of twins, and a mean grain size of about 20 μm (Fig. 3b).

Two copper foils were irradiated, a third one was kept to get reference data. Irradiations were performed at JANNuS facility (CEA, Saclay) [30] with 2 MeV protons. Such energy ensures that a significant thickness of the material is irradiated while avoiding the $^{65}Cu(p,n)^{65}Zn$ nuclear reaction[1] making the samples radioactive. Each copper foil was fixed to a copper sample holder cooled by liquid nitrogen (Fig. 2a) in order to avoid any heating due to the energetic proton beam. A 20 mm diameter disk-shape region of the foils was irradiated, corresponding to the rastering of the millimetric ion beam [30]. Sample temperature was monitored to be below 20 °C throughout the irradiations. Flux and fluence obtained for each irradiation are given on Fig. 2b. SRIM-2013 software [33] was used to compute Displacement Per Atom (thereafter noted *dpa*), using Kinchin-Pease (KP) model [34] and a displacement energy of 30 eV for copper [35]. For irradiation n°1, ion-beam angle of 15° with the foil normal was also accounted for in the calculations. Dpa levels as a function of depth are shown on Fig. 2b where

[1] The threshold energy of $^{65}Cu(p,n)^{65}Zn$ is 2.17 MeV [31,32].