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In-situ micro-cantilever bending test in environmental scanning electron microscope: Real time observation of hydrogen enhanced cracking

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ABSTRACT

A novel approach of in-situ micro-cantilever bending tests is introduced, integrating nanoindentation and environmental scanning electron microscopy (ESEM) to elucidate hydrogen embrittlement (HE) in FeAl. Bending tests were performed in vacuum (~5×10^{-4} Pa) and in ESEM with water vapor (180 Pa, 450 Pa) conditions, which introduce H in-situ into the cantilevers during the test. Micro-scale In-situ SEM testing provides a full control of all the parameters involved in HE as well as avoids the proximity effect from the free surface, which is always criticized in nano-scale in-situ TEM experiments. Both hydrogen induced cracking and hydrogen reduced flow stress were observed.

Hydrogen-enhanced or hydrogen-induced failure has been a topic of debate for the past decades. Different experimental observations and theoretical simulations proposed different models. One model is the hydrogen-enhanced decohesion (HEDE), which was proposed by Troiano in the 1960s [1], which simply postulated, that the hydrogen accumulated at the crack tip reduces the cohesive bond energy between atoms, thus decreasing the work needed for fracture to occur. The HEDE mechanism was found to agree with the observations that the crack tip opening angle decreases with rising hydrogen pressure in Fe-3Si (wt.%) and Ni single crystals [2]. Later, the HEDE mechanism has been expanded to the grain boundary and phase boundary area [3,4] based on the experimental observations that hydrogen will weaken the boundary toughness, thus promoting intergranular fracture. The next proposed model is the hydrogen-enhanced localized plasticity (HELP), proposed mainly based on the in-situ observation of dislocation motion inside environmental TEM cells [5–8]. From these observations, the solid solution hydrogen increases the dislocation mobility or decreases the stacking fault energy, thus improving the cross-slip. These mechanisms will introduce local plasticity and strain softening, and thus explain the hydrogen-enhanced plastic failure. Another alternative plasticity-based mechanism is the adsorption-induced dislocation emission (AIDE) reported in [9], which states that the formation energy of dislocations at crack tips is reduced by hydrogen adsorbed at the crack surface, after which crack propagation is prompted by dislocation motion. A recently proposed model, the defactant model, describes the HE based on the thermodynamic viewpoint [10,11]. Analogous to the action of surfactants with surfaces, the segregation of H to defects (vacancies, dislocations, and boundaries) with an excess value of Γ will lower the defect formation energy.

Obviously, in order to experimentally depict and distinguish between these mechanisms, proper experimental setups are required to allow firstly in-situ charging H and secondly a full observation of H effect on the deformation process and microstructure evolution. We solved these challenges by the miniaturization of the experimental setup, where a full control on H charging, microstructural evolution as well as H affected zone is accessible.

In this paper, a novel approach was applied for bending micro-sized cantilevers in a setup with the combination of nanoindentation system and ESEM to meet the above mentioned requirements. Using a specimen scaling in micrometer size allows us to narrow down the parameters involved in HE under full control as well as to avoid the proximity effect from the free surface, which is always criticized in in-situ TEM experiments. The novelty of our approach is to utilize the ESEM as a hydrogen charging instrumentation. Given water vapor as the default environment in ESEM option, the test material is wisely chosen to the one that will be embrittled by moisture-produced hydrogen, i.e. the FeAl intermetallic alloy with B2 crystal-structure.

The detrimental effect of the environment on FeAl alloy was attributed to hydrogen embrittlement, where the atomic hydrogen was produced by the reaction of aluminum with water vapor [12–15]:

$$2Al + 3H_2O \rightarrow Al_2O_3 + 6H^+ + 6e^-$$

One can use the reaction kinetics to calculate the H concentration: $C_{[H]} = C_0\,[1 - \exp(-kt)] = K_a K_r K_d P_{H2O} (C_{Al})^{1/3}\,[1 - \exp(-kt)]$, using K_a, K_r and K_d which are the rate constants for reactions of adsorption, oxidation, and dissolution of H, together with the P_{H2O} water partial pressure and C_{Al} the Al concentration in the alloy. Unfortunately, the

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available kinetic data for this are very limited and the experimental work to estimate them are lacking. Even though the hydrogen embrittlement of FeAl in a moisture-containing environment is widely agreed, the mechanism of hydrogen-assisted fracture is still not well understood. We explored the interaction of water vapor with FeAl alloy utilizing the novel approach proposed, and analyzed the HE effect further.

The single crystalline Fe-Al was grown by a modified Bridgeman technique [16]. Cantilevers were cut from this specimen, by a focused ion beam (Helios Nanolab DualBeam FIB, FEI Inc.) system with ~2 × 4 × 8 (μm) dimensions using 93 pA at 30 kV as the final current, in order to maintain a good surface quality. The cantilever beams have an initial orientation of [4,3,23] parallel to normal direction and [28,1,5] parallel to loading direction, as shown in Fig. 1a. After ex-situ electron backscattered diffraction (EBSD) characterization, the beams were subjected to the load in-situ using the PI-85 Pico-indenter system (Hysitron Inc.) inside the ESEM (Quanta FEG 650 ESEM, FEI Inc.) in displacement-controlled mode with a 2.5 nm/s loading rate. The dwell time between loading and unloading steps was 20 s. Bending tests were performed both under vacuum (~5×10^{-4} Pa) and under water vapor (180 Pa, 450 Pa) conditions. The latter provides an in-situ H charging environment. The beams were bent to 1 μm displacement in 180 Pa and to 5 μm displacement in 450 Pa ESEM conditions. As shown in Fig. 1b, the cantilever is initially covered by a thin oxide layer created by the chemical reaction of aluminum with the moisture in air. During bending, small cracks will form on the oxide layer, which provide a pathway for water molecules to contact with fresh FeAl matrix. The H produced by the chemical reaction of H_2O is transported to the tension area through diffusion. A minimum of three tests were conducted under each condition. The microstructure after bending was characterized using EBSD and TEM (JEM-2100, Jeol Inc.).

The stress–strain curves shown in Fig. 2a were obtained from the force–displacement measurements. The bending stresses are calculated using Eq. (1), in which *F*, *y*, *w*, *t* refer to the forces, moment arms, width and thickness of the beams, respectively [17]. The corresponding strain, ε, was corrected by normalizing the displacement, *d*, with the moment arm, *y*. Due to the asymmetrical geometry of the beams, they were not bent around the same defined curvature radius, so the normalization of the displacements does not give the exact strains. However, the good agreement of the elastic stiffness among the different beams suggests that the moment arm is a reasonable normalization measure.

$$\sigma = \frac{4Fy}{w \cdot t^2} \quad (1)$$

Fig. 2 shows three different sets of experiments in which cantilevers were bent to 1 μm and 5 μm displacements under three different environmental conditions, i.e. in vacuum, in ESEM with 180 Pa and 450 Pa water vapor pressures. EBSD analysis was performed on the beams bent to 5 μm and TEM on the beams bent to 1 μm. For a better comparison, the flow curves tested in ESEM mode were offset by 0.1 unit on the strain axis.

The flow curves show a clear effect of the hydrogen from three main phenomena. 1) A reduction of yield strength (YS) was observed in ESEM mode. 2) The flow stresses (FS) of all the beams bent in water vapor environment decreased compared with those bent in vacuum. The reduction of FS increases with increasing water vapor pressure. 3) A continuous decrease of bending stress after 0.45 strain was detected, which is due to the propagation of cracks nucleated from the transition corner between the beam and bulk, as shown in Fig. 2c_1.

The reduction of YS as one of the effects of hydrogen on FeAl has been pointed out previously in [18,19]. In their experiments, the YS of Fe-40Al was found to decrease with decreasing strain rate when tested in air, while in vacuum the YS was independent of strain rate. One possible reason for the hydrogen-reduced YS is that hydrogen aids the heterogeneous nucleation of mobile dislocations at the surface. Reducing activation energy for homogeneous dislocation nucleation by hydrogen has been recently pointed out via in-situ electrochemical nanoindentation on FeAl single crystals [20]. After further bending of the beam to 0.45 straining, cracks are nucleated from the triple corner of the cantilevers, where the beam experiences the highest tensile stress. Further propagation of the crack in a stable manner, the FS decreases continuously until the final 5 μm displacement is reached. The crack nucleation on the sample surface, when tested in air, has been observed previously in Fe-40Al alloy, while internal crack initiation was found when tested in O_2 atmosphere [21]. In our experiments no crack nucleation was observed in the tests performed in air (Fig.2b).

The local misorientation gradients are shown by Kernel Average Misorientation (KAM) maps, which qualify the average misorientation of one EBSD point with respect to a defined set of neighbors (the 3rd neighbor shell is used here), as shown in Fig. 2b_2 and c_2. The confidence index (CI) is higher than 0.1 for all the points shown here and points with CI lower than 0.1 are simply colored in black. The low CI values for the bottom part of the cantilever in Fig. 2c_2 were due to the out-of-plane bending induced by the crack propagation from the corner to the outmost surface, which makes this region deflected from the 70° plane for EBSD. Hence, in order to interpret the results, we compare

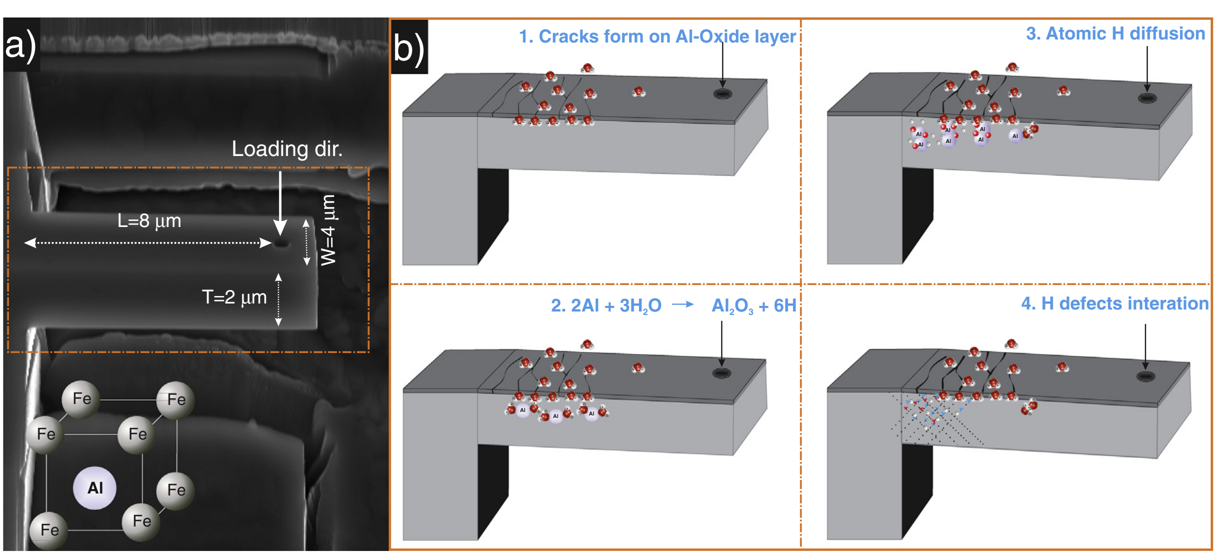

Fig. 1. Side view of a FIB-milled FeAl single crystal cantilever before bending a); the principle of in-situ hydrogen charging within ESEM mode b).