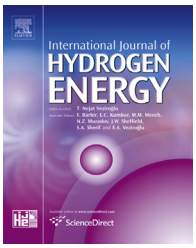
Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/he



Multi-phase modelling of intergranular hydrogen segregation/trapping for hydrogen embrittlement

Sathiskumar Jothi [a,*], *T.N. Croft* [a], *L. Wright* [b], *A. Turnbull* [b], *S.G.R. Brown* [a]

[a] *College of Engineering, Swansea University, Singleton Park, Swansea SA2 8PP, UK*
[b] *National Physical Laboratory, Teddington, Middlesex TW11 0LW, UK*

ARTICLE INFO

Article history:
Received 4 April 2015
Received in revised form
18 August 2015
Accepted 24 August 2015
Available online 26 September 2015

Keywords:
Hydrogen embrittlement
Microstructures
Metallic polycrystalline material
User element
Finite element analysis

ABSTRACT

Premature failure in polycrystalline materials due to hydrogen absorption affects a wide range of applications, including clean energy systems, hydrogen storage systems and rocket engines. A good understanding of the diffusion and trapping processes within such materials can inform material choices and component design to reduce the likelihood of such failures. Grain boundary segregation of hydrogen can often lead to intergranular hydrogen embrittlement (IHE). In order to understand the effects of hydrogen on intergranular and transgranular fracture in polycrystalline material it is important to first understand hydrogen diffusion and trapping in the general context of grain boundary segregation engineering (GBSE). Hydrogen diffusion is affected by local microstructural features including intergranular second phase precipitates, grain boundary (GB) thicknesses and geometrically necessary dislocation (GND) density. A multi-scale multi-phase model is presented here that has been developed to study GBSE with respect to hydrogen diffusion and IHE. The results of various multi-scale GBSE models with and without traps (including the effects of microstructure, intergranular precipitate phases and GB thickness) are compared and discussed, and the effects of microstructural parameters such as hydrogen segregation factor and GND trapping density on hydrogen diffusion are investigated.

Introduction

Hydrogen is a common fuel in rocket engines and these rocket engines are made of polycrystalline materials. It is well known that hydrogen causes embrittlement in many polycrystalline materials, including high nickel content polycrystalline materials, and catastrophic failure can occur in hydrogen fuel rocket engine components [1–19]. Hydrogen induced intergranular and transgranular embrittlement in polycrystalline materials are well established as causes of catastrophic brittle failure and have been studied extensively for more than five decades [1–13]. Apart from rocket systems, hydrogen diffusion, segregation and trapping in metallic polycrystalline materials are also significant as causes of hydrogen embrittlement in clean energy fuel storage systems used as power

* *Corresponding author.*
E-mail address: s.jothi@swansea.ac.uk (S. Jothi).
http://dx.doi.org/10.1016/j.ijhydene.2015.08.093

Nomenclatures

EBSD	electron backscattering diffraction
FE	finite element
GB	grain boundary
GBAZ	grain boundary affected zone
GBSE	grain boundary segregation engineering
GI	grain interior
GND	geometrically necessary dislocation
HIC	hydrogen induced cracking
IHE	intergranular hydrogen embrittlement
MC	metal carbide
MC	Monte Carlo
MPDII	micro precipitate double layer intergranular interfaces
NPDII	nano precipitate double layer intergranular interfaces
SEM	scanning electron microscopy
SIMS	secondary ion mass spectrometry
TEM	transmission electron microscopy
TJ	triple junction
UEL	user element

sources in the aerospace and automotive sectors. Physical microstructural factors such as grain size, grain boundaries, dislocations, grain boundary segregation of impurities, carbides and non-metallic particles have been identified as playing important roles in this phenomenon [11,13,20–49].

For example, segregation of phosphorous and sulphur assists hydrogen embrittlement in nickel based super alloys [45,46]. Some of these microstructural features such as grain boundaries (GBs) may also promote faster diffusion of hydrogen due to the locally disordered atomic structure [13,22,23,32–40,43,44]. GBs may also act as hydrogen trap (or segregation) sites when the probability of atomic hydrogen jumping into GB sites (capture) is greater than that of atomic hydrogen jumping out of GB sites (escape) [50–53]. Grain boundary segregation engineering (GBSE) is fundamental to understanding intergranular hydrogen embrittlement [22,41,44,50,53]. Dislocation sites may also act as traps for hydrogen atoms (providing a deeper potential energy well) and hydrogen atoms jumping into these sites may reside for longer times than in lattice sites [50–53]. These trapping effects can lead to an overall decrease in the rate of hydrogen transport over time [11,12,54–58]. The processes of hydrogen transport through/near such microstructural features are important for understanding the different modes of hydrogen induced cracking (HIC) in polycrystalline nickel [21,22] for example.

Intergranular and transgranular failures are two different modes of HIC. HIC commonly occurs along grain boundaries where high concentrations of hydrogen can exist [21]. These concentrations highlight the importance of including the random grain boundaries in any numerical model designed to predict embrittlement. Detailed description of the importance of grain boundary diffusion of hydrogen in nickel can be found in Harris [22,50]. In nickel components combinations of micro polycrystalline and nano polycrystalline nickel microstructures are often encountered. These are complex composites comprising irregular polygonal grains, random grain boundaries and triple junctions. The detailed nature of random irregular micro and nano polygonal grains and random irregular grain boundaries is significant when considering the processes of hydrogen diffusion and segregation mechanisms that lead to embrittlement failure in nickel [22–24].

Manufacturing processes such as casting, electro deposition and welding of polycrystalline materials and post-manufacturing processes such as hot rolling, cold rolling, heat treatment and hardening using thermo-mechanical processing produces residual plastic strain and GB misorientation which is directly associated with (geometrically necessary dislocation) GNDs. In order to understand GBSE for hydrogen transport based on elastic and plastic strains developed by manufacturing processes, a trap model and multi-scale multi-phase microstructural model has been developed to study the effect of GB misorientation, GND density and trap density on hydrogen segregation mechanisms in polycrystalline material. GB misorientations accommodate GNDs [32,44,50,59]. Hydrogen diffusion and segregation may increase or decrease depending on GB misorientation, GND density and GB energy, and GNDs can act as trapping sites for hydrogen in polycrystalline materials. It is also reported that hydrogen diffusion and segregation is influenced by GNDs, dislocation density, triple junctions (TJs), GB misorientation and GB connectivity [32,59,60]. The GND density may also vary depending on the GB misorientation types, angle and the GB energy. Previously hydrogen diffusion in FCC polycrystalline material has been studied using a Secondary ion Mass Spectrometry (SIMS) technique and it was reported that high energy GBs accelerate the diffusivity of hydrogen [25,44,53,59,61].

Raabe et al. observed double nano layers in grain boundary interfaces with thicknesses of 10–15 nm in polycrystalline steel material. Impurity segregation and nanoprecipitation in this intergranular double layer was also reported [41,42]. Microprecipitation in intergranular double layers in nickel-based polycrystalline alloys have been observed in this work as shown in Fig. 1. This indicates that it is important to take into account this intergranular double layer interface when studying the GBSE of impurity segregation and trapping in polycrystalline materials.

In the present study, several multi-scale multi-phase simulations with heterogeneous random irregular polygonal grains possessing random intergranular double layers as a second phase have been performed. Various microstructures containing intergranular double layer precipitate interfaces and Grain boundary affected zones (GBAZs) have been generated. A trap model based on the McNabb trap model has been implemented in a FORTRAN user element (UEL) subroutine within an ABAQUS-based multi-scale multi-phase microstructural model. Monte Carlo (MC) simulation has been performed for microstructural grain geometry development and trap density calculations based on GB misorientation. A coupled MC Finite Element (FE) multi-scale multi-phase continuum trap model is used to investigate hydrogen trapping and GB segregation mechanisms in polycrystalline material. A MC model has also been developed to investigate