Micro Architectures Materials and Hydrogen Embrittlement

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ABSTRACT

Hydrogen embrittlement is a classical problem in bulk materials while it is rather untouched for advanced materials such as micro-architectured materials. This can be a barrier to industrial adoption of these materials where hydrogen is present as a popular source of energy. In this study, we developed a numerical scheme to assess the hydrogen degradation in metallic micro-architectured materials. The numerical scheme is built on the concept of elastoplastic homoge nization and two hydrogen embrittlement theories, i.e. hydrogen enhanced decohesion (HEDE) and hydrogen enhanced localized plasticity (HELP). The use of homogenization allows for explicit de nition of a unit-cell, drastically improving the computation time. The hydrogen degradation loci of two speci c micro-architectured materials, that is cubic (with 10%, 20% and 30% relative densities) and body-center cubic (with 20% relative density), are numerically characterized. Additionally, the in uence of unit-cell topology, relative density, and trap hydrogen on the degradation of homogenized macroscopic material is determined. Finally, a unique failure locus is provided for generic cubic unit-cell with arbitrary relative densities. This degradation law is in- dependent of the relative density and can be interpreted as a material property, contributing to the material design charts.

KEYWORDS: Fracture Mechanics, Structural Integrity, Hydrogen Embrittlement, Nondestructive Evaluation

1.0 INTRODUCTION

Micro-architectured materials are a class of porous multiscale materials formed by the periodic assembly of a unit-cell (micro to millimeter length-scale) in the design space. The unit-cell architecture can be arti cially tailored to achieve a speci c property such as negative Poisson's ratio, high acoustic damping, or energy absorption. The porous structure of these materials provides a great weight saving potential. This, in addition to their tailored properties, make micro-architectured materials interesting candidates for a wide range of industries. These materials were dif cult to fabricate, but the recent advancements in additive manufacturing took this hindrance away and accelerated their industrial adoption. Yet, accurate prediction of the failure and lifetime of these materials is crucial for their wide-spread application. In this respect, we may classify the literature on micro-architectured materials into two groups. Some studies focus on the con- ceptual design and physics governing the behavior of these materials, e.g. [1-17], while others investigate the failure (quasi-static and fatigue) under compression, e.g. [18-29] and [30-47], and (more recently) under tension, e.g. [48-56]. Most of these failure-related studies highlighted the role of manufacturing imperfections, and some thoroughly investigated the topic, e.g. see [57-75]. These imperfections (like void, strut waviness and surface roughness) often arise from additive manufacturing processes, and noticeably in uence the material behavior. This topic has gained attention, and some recent studies are solely devoted to the analysis of defects in micro-architectured materials, e.g. [66-80]. We argue that additive manufacturing defects are not the only imperfections that a micro-architectured material may encounter. As we move towards a more sustainable future, fossil fuels are being replaced by alternative sources of energy. Hydrogen is one of the main alternatives, especially in aerospace and automotive industry. These industries employ micro-architectured materials in their products, potentially facing a new challenge, that is hydrogen embrittlement in metallic micro-architectured materials. To the authors' knowledge, this form of defect in micro-architectured materials has not been addressed in the literature. Hydrogen embrittlement is a well-known issue in bulk metallic materials, which has been studied both numerically, e.g. [1-22], and experimentally, e.g. [23-47]. More recently, the research on this topic has extended to additively manu-factured bulk materials, e.g. see [48-57]. There exist two prevailing theories for hydrogen embrittlement, namely the hydrogen enhanced decohesion (HEDE) mechanism and the hydrogen enhanced localized plasticity (HELP) mechanism. The former assumes that hydrogen reduces the cohesive strength of material lattice, and the latter assumes that hydrogen enhances dislocation activity, leading to localized plasticity and reduced global plastic. So far, it is still unclear how the presence of

hydrogen affects the macroscopic behaviour of micro-architectured materials [58-66]. In this study, we address this question. To capture hydrogen embrittlement in micro-architectured materials, we develop a computational framework with explicit denition of material unit-cell with periodic boundary conditions (PBC). We combine this model with hydrogen diffusion equations and update the materials and maximum stress based on the HEDE and HELP mechanisms. We then use elastoplastic homogenization to compute the macroscopic material behavior [67-77]. The developed model is generic and applicable to any arbitrary micro-architectured material. We organize the paper as follow. In Section 2, we derive the governing equations for hydrogen diffusion, discuss how hydrogen concentration affects material's constitutive behavior, and review elastoplastic homogenization theory [68-85]. We then calibrate the material model and elaborate on the numerical scheme and its implementation in Sections 3 and 4, respectively. In Section 5, we apply the model to two micro-architectured materials, namely cubic and body-centered cubic (BCC), to characterize their macroscopic hydrogen degradation laws. We also investigate the influence of relative density, unit-cell architecture (topology) and trap hydrogen on strength degradation of the candidate materials. Finally, we conclude on the results and provide suggestions in Section 6.

2.0 METHODOLOGY

The theories discussed in Sections 2.1–2.3 describe the behavior of a homogeneous solid material. To assess the behavior of a multiscale material, we need to bridge between scales, i.e. from microscale to macroscale, using homogenization. Periodic micro-architectured materials are formed by the tessellation of a unit-cell in the design space. We investigate the behavior of these mate- rials by isolating a single unit-cell, also referred to as a representative volume element (RVE), subjected to periodic boundary condition (PBC). We then take the volume average of microscopic elds over the RVE domain to obtain the effective material properties. This approach is referred to as "averaging homogenization", and is well-established for the analysis of multi-scale materials, e.g. see [1-33] for more details. In this section, we follow a coupled chemo-mechanical analysis and derive the governing equations for hydrogen diffusion under quasi-static loading conditions. We then elaborate on Orian's theory to determine trap hydrogen concentration. The influence of hydrogen on mechanical properties, the stress and failure criterion are then introduced. Finally, we review the concepts behind elastoplastic homogenization for the analysis of multiscale materials. In the following, we use the theories elaborated in this section as tools to construct a numerical scheme and study the hydrogen embrittlement in micro-architectured materials. To analyze the in uence of hydrogen on micro-architectured materials, we first need to determine the properties of the base material. We choose AISI 4135 (B-15) steel as the base material, for which we have experimental and numerical data on its hydrogen- charged tensile specimens in the literature, e.g. [34-56]. Using the available data, we calibrated the material's true stress–strain response in the absence of hydrogen (Ctot = 0) in the form of Eq. (17), see Table 1. However, the material properties listed in Table 1 are insuf cient for calibration of the material failure because notch radius and consequently stress triaxiality affect the maximum strength of the sample. Within fracture community, it is common to characterize the material failure in terms of plastic strain and stress triaxiality, e.g. see [57-69]. Such relations are not explicitly provided when the measure is the maximum strength, while most studies on hydrogen embrittlement use stress-based criteria in their analyses, e.g. [66-78]. For that, we employ the failure strength data in [79-85] and perform numerical simulations on notched tensile specimens (see Appendix A) to characterize the failure locus of the material in terms of maximum strength and stress triaxiality. We record the applied displacement and reaction force at each time increment of the numerical test, and divide them, respectively, by initial length and cross-sectional area at the center of the specimen to obtain the nominal stress–strain curve. We stop the simulation once the maximum global stress (according to provided experimental data in [1-22]) is reached, and store the local stress and stress triaxiality of the element with maximum principal stress at the vicinity of notch. in bulk materials, e.g. see [23-47]. Here, we make connection between theories reviewed in Section 2 and develop an integrated numerical scheme for the analysis of hydrogen embrittlement in micro-architectured materials. We execute coupled chemo- mechanical analysis under quasi-static loading, in which the mechanical homogenization is updated with the input from implicit diffusion equations. The overall simulation process is shown in Fig. 1. We rst discuss the general procedure, and then explain the subroutines for hydrogen embrittlement (UHARD and USDFLD) in more detail. The details of elastoplastic homogenization scheme (1st and 2nd python script in Fig. 1) are provided in [46-64], and we skip further explanation here. We simulate uniaxial strain-controlled tensile tests by applying macroscopic strains to the material unit-cell with PBC. We then pass the model to Abaqus 2019 implicit solvers along with two Fortran subroutines, i.e. UHARD and USDFLD. We use UHARD to de ne hydrogen

concentration update the stress with the progression of hydrogen, and revise the value of maximum stress with respect to stress triaxiality. We also need to employ USDFLD to accessible variables at material points during the analysis. Once the simulation is complete, we read the output data and perform averaging according to elastoplastic homogenization rules using another python script [65-78].

Table 1
Mechanical properties of AISI 4135 in absence of hydrogen.

Elastic modulus (E)	Yield stress (σ_0)	Hardening exponent (N)	Ultimate tensile strain (ε_{ut})
195 *GPa*	1320 *MPa*	0.11	1.7 %

Table 2
Material parameters for hydrogen diffusion in AISI 4135.

Number of trap sites per lattice atom (α)	1
Number of NILS sites per lattice atom (β)	6
Dislocation density at zero plastic strain (ρ_0)	$10 \times 10^{-10} m^{-2}$
Lattice Parameter (a)	$2.86 \times 10^{-10} m$
Hydrogen binding energy (ΔH)	$-18\ KJ.mol^{-1}$
Number of lattice atoms per volume (N_L)	$8.64 \times 10^{28} m^{-3}$
Partial molar volume of hydrogen (V_H)	$2 \times 10^{-6} m^3.mol^{-1}$
Parameters related to dislocation density	$k_1 = 2 \times 10^{16} m^{-2}, k_2 = 10^{16} m^{-2}$

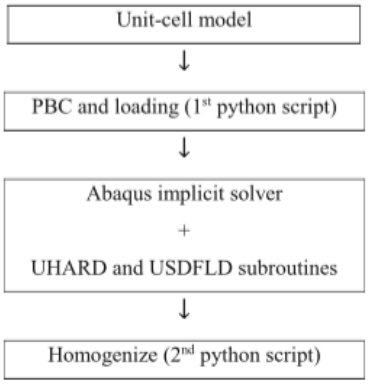

Fig. 1. Overview of the numerical scheme.

UHARD

- Include USDFLD subroutine to read or compute the required field variables, e.g. principal stresses, pressure stress, von Mises stress, etc.
- Compute NILS hydrogen concentration, eq. (10)
- Compute trap hydrogen concentration, eq. (12)-(16)
- Compute the initial yield stress in presence of hydrogen, eq. (18)

- Update ultimate tensile stress (SUT), eq. (19) and (24)
- Check the failure criterion:

IF ($SPMAX \geq SUT$) THEN
$SYIELD = 0$
$HARD(1) = 0$
ELSE IF
Compute $SYIELD$ from eq. (17)
$HARD(1) = \frac{\partial(SYIELD)}{\partial(PEEQ)}$
END IF

Fig. 2. Algorithm for UHARD subroutine.

3.0 RESULT

We study the mechanical response of hydrogen pre-charged micro-architectured materials under macroscopic tensile loads using the numerical code. We assume the pre-charging time has been suffcient to have a spatially uniform hydrogen concentration over the unit-cell, which is reasonable for high strength steel with a large hydrogen diffusion coefficient. This is especially the case for open-cell micro-architectured materials, where the bulk material is immersed in the hydrogen environment. Yet, additively manufactured micro-architectures may have defects (for example residual stresses at unit-cell joints), which can in uence the initial hydrogen distribution.

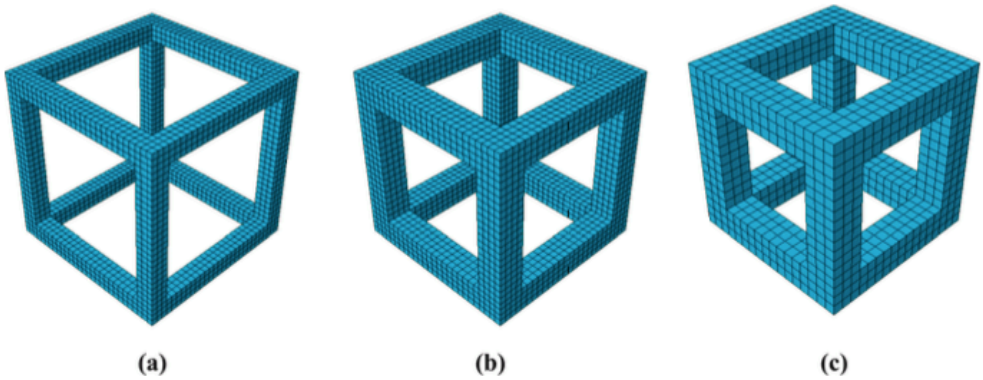

Fig. 3. Cubic unit-cell with: (a) $\rho_r = 10\%$, (b) $\rho_r = 20\%$, and (c) $\rho_r = 30\%$.

But the presence and severity of such defects are subjective to the choice of additive manufacturing method and its processing parameters. Thus, we exclude these defects and assume to have perfectly fabricated materials. We start the analysis with the total hydrogen concentration, i.e. both NILS and trap hydrogen, to evaluate whether the trap hydrogen affects the material behavior or not. Next, as a case study, we implement the numerical scheme on cubic unit-cell and characterize its mechanical performance in the presence of hydrogen. Finally, we implement the code on other unit-cell topologies to examine the role of unit-cell architecture.

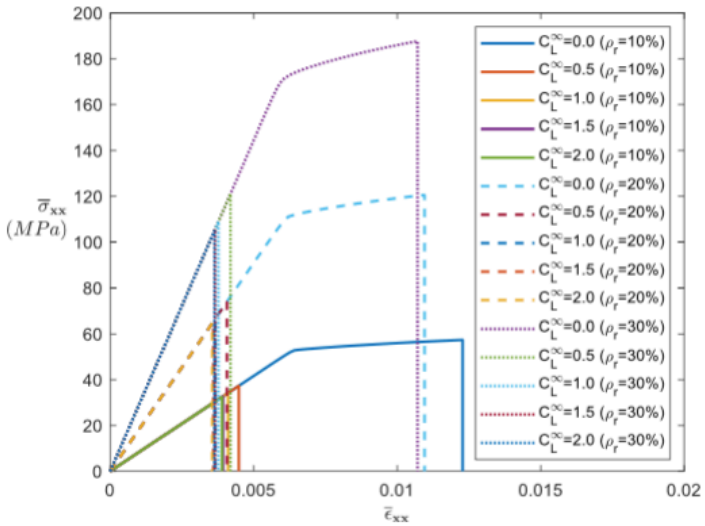

Fig. 4. Macroscopic stress–strain curves for cubic unit-cell with various relative densities (ρ_r) and initial hydrogen concentrations (C_L^∞). Plots for $\rho_r = 10$ %, $\rho_r = 20$ % and $\rho_r = 30$ % are represented with solid, dashed, and dotted lines, respectively. (For colors, refer to online version.).

To assess the in uence of trap hydrogen on maximum strength in micro-architectured materials, we ran simulations under two conditions: (1) Ctot = CL , and (2) Ctot = CL +CT . We noticed the results barely change in the presence of trap hydrogen. In fact, the low ductility of the base material (high-strength steel, AISI 4135) prohibits large scale plasticity. This is even more dominant in micro-architectured materials, as they often have lower ductility compared to their base material. Especially

in the presence of hydrogen, little to no plasticity develops before the failure of micro-architectured materials made of AISI 4135. This makes the contribution of trap hydrogen in the fracture process negligible. However, the situation may differ if we chose a more ductile metal, e.g. low- or mid-strength steel, as the base material. In that case, the concentration of trap hydrogen, especially reversible traps, may in uence the process of crack initiation and propagation. Yet, our conclusion about micro-architectured materials agrees with what Ayas et al. claimed about the bulk samples made of high-strength steel.

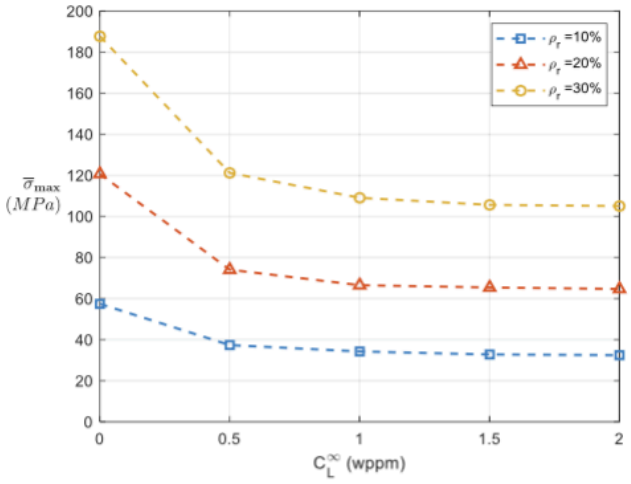

Fig. 5. Macroscopic maximum stress versus pre-charged hydrogen concentration.

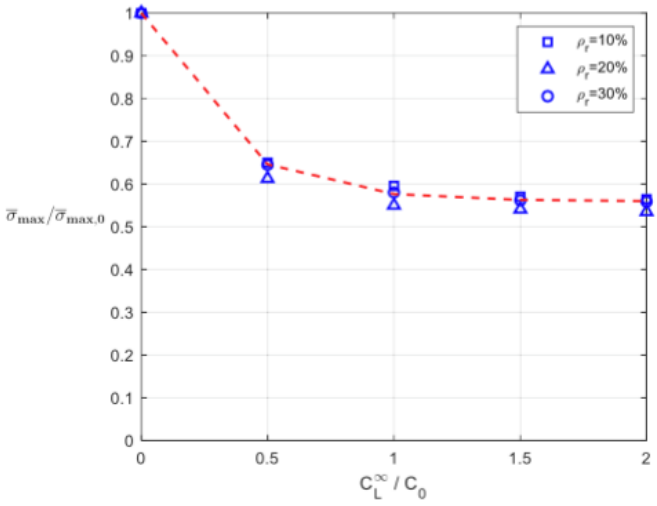

Fig. 6. Normalized failures locus of the cubic unit-cell made of AISI 4135 ($C_0 = 1wppm$).

4.0 CONCLUSIONS

We studied the role of hydrogen in strength degradation of micro-architectured materials. We began by analyzing the Reynold's transport equation for hydrogen diffusion problem and obtained the governing equations for hydrogen concentration under quasi- static loading. We then used the concept of elastoplastic homogenization to bridge the scales and derive the material effective properties. We implemented these theories in a numerical scheme for decoupled diffusion-deformation analysis, in which we update the homogenization algorithm with hydrogen concentration and ow stress through UHARD subroutine and implicit formulae. We applied the model to cubic (withρr = 10 %, 20 % and 30 %) and BCC (withρr = 20 %) unit-cells along with PBC and char- acterized their macroscopic hydrogen degradation laws. Each micro-architectured material has its unique failure loci depending on cell architecture, but one can provide a single expression to describe the behavior of a cubic unit-cell over a range of relative densities. Also, it turned out that the role of trap hydrogen in embrittlement of

micro-architecture materials is negligible, especially when the base material has low ductility. For load bearing applications and in presence of hydrogen, the cubic material outperforms the BCC because: (1) at equal hydrogen contents, its maximum strength is higher than BCC, and (2) it undergoes a less sever hydrogen degradation as compared to the BCC. On the other hand, the BCC material has higher ductility which is desirable for certain applications. Generally, micro-architectured materials made of high-strength steels, e.g. AISI 4135, are prone to brittle fracture in the presence of hydrogen. This agrees with what was observed in bulk specimens, e.g. The developed computational scheme is generic and applicable to any periodic micro-architectured material and is an efficient tool for assessment of hydrogen embrittlement. We proposed the design methodology and numerically characterized the macroscopic hydrogen degradation laws for cubic and BCC unit-cells. The experimental validation of the results on hydrogen embrittlement in architecture materials is suggested as a part of future studies.

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