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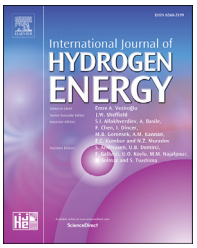

Effect of nickel on hydrogen permeation in ferritic/pearlitic low alloy steels

Hans Husby [a,*], *Mariano Iannuzzi* [a,b,1], *Roy Johnsen* [a], *Mariano Kappes* [c], *Afrooz Barnoush* [a]

[a] *Department of Engineering Design and Materials, Norwegian University of Science and Technology (NTNU), 7491 Trondheim, Norway*

[b] *GE Oil & Gas and NTNU, 1338 Sandvika, Norway*

[c] *Instituto Sabato, San Martín, Argentina*

ARTICLE INFO

Article history:
Received 8 October 2017
Received in revised form
25 December 2017
Accepted 26 December 2017

Keywords:
Oil & gas
Low alloy steel
Nickel
Hydrogen
Permeation
Embrittlement

ABSTRACT

Nickel offers several beneficial effects as an alloying element to low alloy steels. However, it is, in the oil and gas industry, limited by part 2 of the ISO 15156 standard to a maximum of 1 wt% due to sulfide stress cracking resistance concerns.

Hydrogen uptake, diffusion, and trapping were investigated in research-grade ferritic/pearlitic low alloy steels with Ni contents of 0, 1, 2 and 3 wt% by the electrochemical permeation method as a function of temperature and hydrogen charging conditions.

Qualitatively, the effective diffusion coefficient, D_{eff}, decreased with increasing Ni content. The sub-surface lattice hydrogen concentration, C_0, decreased with increasing Ni content in all charging conditions while the trend between the sub-surface hydrogen concentration in lattice and reversible trap sites, C_{OR}, and Ni content varied with the charging conditions. Irreversible trapping, evaluated by consecutive charging transients, was not observed for any of the materials. Lastly, the possible influence of an increasing fraction of pearlite with increasing Ni content is discussed.

Introduction

Low alloy steels (LAS) are widely used in the oil and gas industry due to their excellent combination of mechanical and technological properties and cost [1]. Part 2 of the ISO 15156 standard [2] regulates the use of carbon steel (CS) and LAS in H_2S-containing environments. The standard restricts the allowable nickel (Ni) content to maximum 1 wt%, due to controversial concerns regarding sulfide stress cracking (SSC) resistance [1,3]. Despite extensive investigations from the mid-1960s to late 1980s, the engineering community has yet to reach consensus as to whether the cap on Ni is scientifically justified. ISO 15156 allows the use of steels that exceed the strength, hardness, and composition requirements if successfully qualified as per the procedures described in Annex B of the specification. In practice, however, the 1 wt% Ni limit excludes LAS families with superior mechanical and technological properties that contain above 2–3 wt% Ni, such as ASTM (American Society for Testing and Materials, West Conshohocken, PA) A508 Grade 4, 10GN2MF2 and UNS K32047, from sour service applications [4]. In this regard, Ni improves LAS hardenability, fatigue life and toughness, and lowers the ductile to brittle transition temperature with a moderate penalty on weldability [1]. Qualifying LAS with Ni contents

* *Corresponding author.*
E-mail address: hans.husby@ntnu.no (H. Husby).
[1] Currently at Curtin University, Perth, Australia.

https://doi.org/10.1016/j.ijhydene.2017.12.174
0360-3199/

above 1 wt% could benefit the development of sour reservoirs with severe temperature and pressure conditions [1]. A comprehensive overview of the effects of nickel on LAS performance can be consulted elsewhere [1].

Hydrogen embrittlement (HE) is a generic term referring to the embrittlement of a material caused by the presence of atomic or nascent hydrogen [5]. HE groups various environmentally assisted cracking mechanisms such as hydrogen stress cracking (HSC) and SSC [5]. HSC results from the combination of tensile stress and atomic hydrogen in the metal [6]. Hydrogen uptake, diffusion, and trapping have been shown to affect cracking resistance [7–11]. In this regard, SSC is considered a form of HSC in the presence of H_2S [12]. Ni affects these properties, both directly as an element in solid solution, and indirectly due to its refining effect [13] on the microstructure. As part of a broader effort to quantify Ni's role on the overall HE susceptibility of LAS, this work focuses on the effect of Ni in solid solution in the body center cubic (bcc) ferrite phase on hydrogen permeation.

LAS are primarily used in the quenched and tempered (QT) condition. The effect of Ni on hydrogen permeation in such steels has been investigated previously by, e.g., Wilde et al. [14] and Yoshino and Minozaki [15]. In both cases, the effective diffusion coefficient, D_{eff}, decreased with increasing Ni content. D_{eff} is the apparent diffusion coefficient found from fitting a hydrogen permeation transient to Fick's second law as described in section Analysis of the results. However, no conclusions can be drawn about the effect of Ni present in solid solution in the ferrite phase from the results on QT steels.

Beck et al. [16] investigated hydrogen permeation in pure Fe-Ni alloys at temperatures from 27 to 90 °C. The authors assumed purely ferritic microstructures up to 8 wt% Ni, which was later questioned by others [17,18]. D_{eff} decreased and the hydrogen content increased with increasing Ni content. Dresler and Frohberg [17] performed permeation experiments at 25 °C on Fe-Ni alloys, which, according to the authors, had ferritic microstructures up to 5 at% (~5 wt%) Ni. D_{eff} decreased slightly with increasing Ni content. However, hydrogen concentrations were not estimated. Likewise, consecutive permeation transients to evaluate whether trapping was reversible or irreversible were not performed in either of the investigations. Moreover, all CS and LAS contain interstitial carbon, which may influence hydrogen permeation alone [19] and in combination with Ni in solid solution.

Quantifying the effects of Ni as a solid solution element in the ferrite phase on hydrogen uptake, diffusion, and trapping is essential to gain a fundamental understanding of the HE performance of nickel-containing LAS. This work describes the electrochemical hydrogen permeation testing of ferritic/pearlitic research-grade LAS whose chemistries differed only by their Ni contents.

Experimental

Materials

Research-grade LAS plates that varied only in their Ni content were fabricated for the project. The nominal Ni concentrations were 0, 1, 2 and 3 wt%. The actual chemical compositions are shown in Table 1. The alloys were vacuum induction melted in an alumina crucible at 1600 °C. They were fully "killed" (i.e., deoxidized) and fine grain treated by aluminum addition. Calcium was added for inclusion shape control.

Impurity levels were analyzed by glow discharge mass spectroscopy. Calculated *X*- (Bruscato) and *J*- (Watanabe) factors [20], presented in Table 1, reflect the ultra-low level of impurity elements present in the samples. As suggested by Kohno et al. [21], the materials can be considered immune to temper embrittlement for the purpose of this work.

Heat treatment

Materials were delivered as plates with a thickness of approximately 1 cm after casting and hot-rolling. The rolling operation resulted in plates with banded microstructures in the as-received condition. All samples were subsequently homogenized by prolonged stepwise heat treatments to eliminate the observed banding. Samples were first heated to 1200 °C for 7 days. Furnace cooling to 500 °C (i.e., below the lower transformation temperature, A_{c_1}) and reheating to 930 °C (i.e, above the upper transformation temperature, A_{c_3}) twice to refine the microstructures, followed the homogenization step. Coupons were encapsulated in quartz glass, providing a vacuum atmosphere to minimize oxidation and decarburization of the samples during homogenization. Because Ni has a strong grain refinement effect [13], a third re-austenitization step, followed by controlled cooling down to 600 °C, at a rate slower than that obtained by furnace cooling, was applied to the 2 and 3 wt% Ni samples to obtain comparable microstructures for all Ni contents. The full heat-treatment process is shown in Fig. 1.

Characterization of microstructures

The degree of banding in the as-received materials was documented in a 2 wt% Ni spare sample. The as-received material was heated to 930 °C followed by furnace cooling to 500 °C, before cooling with the furnace door open to air until reaching room temperature. Micrographs were taken normal to the rolling direction by scanning electron microscopy (SEM) using secondary electron imaging. The removal of banding by the homogenization treatment was confirmed by microstructure investigations in the SEM normal to the rolling direction in samples of all Ni contents. Likewise, the microstructures of the tested samples, one of each Ni content, were documented in the SEM after permeation testing.

Table 1 – Chemical compositions of research-grade LAS. Analyzed by manufacturer with methods specified in ASTM E1019-11/CO [22] and ASTM E1479-99/CTP3101/ICP [23].

Alloy	Ni [wt%]	Mn [wt%]	Si [wt%]	C [wt%]	*X*-factor	*J*-factor
0 wt% Ni	0.00	1.30	0.24	0.17	0.47	6.99
1 wt% Ni	0.97	1.30	0.24	0.17	0.48	7.05
2 wt% Ni	1.85	1.28	0.23	0.17	0.43	6.56
3 wt% Ni	2.86	1.30	0.24	0.17	0.59	9.09