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Investigation of crack propagation resistance of 304L, 316L and 316L(N) austenitic steels in liquid sodium

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HIGHLIGHTS

- The fracture behavior of 3 austenitic steels has been tested in liquid sodium on notched tensile specimens after pre-wetting in oxygenated sodium (200 wppm).
- Austenitic steels are shown to have decreasing crack propagation resistance in liquid oxygenated sodium for some experimental conditions.
- Crack initiation occurs after significant plastic deformation.
- Evidences of brittle fracture are observed on the fracture surface.
- The effect of impurities on LME susceptibility is estimated to be unlikely.

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ABSTRACT

In order to assess the susceptibility of candidate structural materials to Liquid Metal Embrittlement (LME), the fracture behavior of three grades of austenitic steels was investigated in oxygenated (200 wppm) liquid sodium in the temperature range [473–673 K] on notched axisymmetric tensile specimens. The tests were carried out in an inert glove box at very low concentrations of dioxygen and humidity (<1 ppm) to prevent further contamination after pre-exposure in oxygenated liquid sodium. A decrease in crack propagation resistance of the three austenitic steels (304 L, 316 L(N), 316 L) is observed in oxygenated liquid sodium (200 wppm) from 573, 623 and 673 K respectively after pre-wetting in oxygenated sodium. This reduction is correlated with a ductile to brittle change of the fracture surface. This effect observed with the three austenitic steels is attributed to the onset of LME after significant plastic deformation.

1. Introduction

Liquid sodium has a range of potential applications such as coolant medium in nuclear reactor or solar thermal power plants. The issue of sodium interaction with structural materials has been an important topic of investigation since the early days of its use. The alloying elements of structural materials dissolve in ultrapure liquid sodium leading to subsequent mass transfer in the presence of temperature gradients and high flow conditions. In addition, the long-term evolution of material crucially depends on its interaction with the dissolved oxygen content. Indeed, it is known that dissolution kinetics are enhanced by traces of oxygen. Corrosion in liquid

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sodium is then mitigated via an active control of the oxygen concentration below a few wppm [1]. Carbon transport processes between structural materials or parts of sodium loops lead to carburization/decarburization issues [2]. Therefore, long term effects of exposure in loops have been predominantly studied in terms of material microstructure evolution and the subsequent mechanical properties degradation [3]. The question of a possible sensitivity to liquid metal embrittlement (LME) of austenitic steels in liquid sodium is also of interest although such a case has never been reported to our knowledge on a component of an operating facility. While several LME cases in liquid sodium were observed on ferritic or martensitic steels through sensitivity tests carried out in laboratory's conditions [4,5], the effect of sodium on the fracture mode of austenitic steels is regarded as negligible [6,7]. This stems from results of tensile testing carried out ex-situ after pre-exposure in vacuum test vessels. These tests, although extremely demanding due to the long-term pre-exposure required, were not carried out in a liquid sodium environment, as already mentioned, and were only targeting the detrimental effects on steel's microstructure of long term high temperature liquid sodium exposure. On the other hand, while the effect of sodium on 304 steel fatigue crack behavior was shown to be negligible at 823 K [8], intergranular cracking of the 304 austenitic steel was also observed during tertiary creep in sodium at the same temperature [9]. Similar behavior was also reported in sodium contaminated by sodium hydroxide [10]. Overall, the effect of sodium on the fracture mode seems to have a different outcome when sensitivity tests are carried out in-situ. Indeed, the 304 L austenitic steel was later on found to have an interfacial LME fracture mode in liquid sodium at lower temperature (between 473 K and 673 K) [11,12].

It is not sufficient to carry out in-situ experiments, one has also to make sure that LME prerequisites are fulfilled. Indeed, in a recent study of LME sensitivity on austenitic steels (15-15Ti and 316 L(N)) by in-situ small punch tests (SPT), a small effect of sodium on mechanical resistance was found for these steels, associated mainly with a ductile fracture mode [13]. Some scatter as a function of strain rate and temperature was also observed in these data. It is to be recalled that sodium doesn't wet native steel's oxides during short time exposure [14] while this is a requirement to correctly induce crack initiation in LME. This scatter might be attributed to the somewhat ineffective wetting procedure inducing uncertainty about the actual wettability state of the specimen during these tests. This stresses out the importance of carrying out mechanical tests in well-controlled conditions notably good wetting for example obtained by pre-exposure in oxygenated sodium [15].

The present study investigated the LME susceptibility in liquid sodium of three austenitic steels, 304 L, 316 L and 316 L(N) in well-defined wetting and static flow conditions (i.e using the recent insight obtained by the analysis of wetting by pre-exposure to liquid oxygenated sodium) to enable reproducible crack initiation independent of the inherent fluctuations of short term corrosion kinetics. After the wetting procedure, the residual sodium on the sample has to be prevented from contamination. Indeed, the role of minute amount of impurities in the gas gathered by the highly reactive liquid sodium can also be a source of unwanted contamination. Water vapor is a main concern because of its tendency to strongly react with sodium to form sodium hydroxide. In the absence of di-oxygen and di-hydrogen in the gas phase, the contamination by sodium hydroxide decomposes upon raising the temperature, thereby releasing hydrogen and oxygen in sodium [10]. High purity of the test and handling environment is therefore required to avoid such effects. In order to isolate the liquid sodium of such potential contamination by gas phase, we developed a new experimental setup able to fully isolate the specimens wetted with liquid sodium from contact with water vapor as well as di-oxygen during transfer and mechanical loading. The LME susceptibility of the three steels was investigated as a function of temperature from 373 K up to 673 K.

2. Materials and experiments

2.1. Description of the testing devices

In mechanical testing experiments, one usually does not have the capability to wait for the required time to dissolve the native oxide even if the sodium chemistry is in the reducing range (typically [O]<3 wppm for nuclear reactor [2]) with the exception of creep testing which is a long-term mechanical test carried out in dedicated loops. With other types of mechanical testing, specimens need to be prepared in a pre-wetted state to allow reproducible crack initiation conditions followed by sodium supply at the crack tip by capillarity during the crack propagation phase. In a previous work [5], the key step was the realization that one can induce sodium to wet the surface via the buildup of a ternary oxide in oxygenated sodium (Na_xCrO_2, $x \leq 1$). This change of the sodium chemistry leads in a few tens of hour to a very reproducible wetting state via a process of reactive wetting [15]. Therefore, the experimental procedure is divided into three steps: pre-wetting in liquid oxygenated sodium, transfer under inert conditions and mechanical testing in liquid sodium in a dedicated testing apparatus.

The pre-wetting equipment is an inert glove box equipped with a sodium static pot able to reach high temperature while remaining perfectly tight. The cover gas is Ar with a continuous recycling in a purification bypass able to lower the concentration of dioxygen and water content to less than 1 ppm, which is necessary to limit contamination during handling and preparation of the test. This experimental apparatus is further described in Refs. [5,16]. In order to provide the same conditions during subsequent handling and testing, an argon inert glove box was designed and adapted to a MTS 20/M uniaxial electromechanical testing machine (Fig. 1a). The load line is coupled to the glove box by flexible bellows ensuring gas tightness. The glove box gas is argon and an online purification process allows to keep dioxygen and water vapor contents lower than 1 ppm. The measured humidity dew point inside the glove box is constantly lower than −80 °C (equivalent to less than 1 ppm of water vapor). The overall height of the glove box is 0.8 m leaving ample space for the load line and several monitoring equipment. The load line has a special design to ensure electrical insulation from the glove box and heating is produced via a low voltage high intensity current (Fig. 1b). A control loop using an optical pyrometer measurement in the glove box regulates the temperature.

2.2. Materials and experimental procedures

In this work, three grades of austenitic stainless steel have been investigated, namely AISI 304 L, 316 L and 316 L(N). Their chemical compositions are listed in Table 1. EBSD mapping analysis of the initial microstructures revealed that the three materials have weak crystallographic texture and equiaxed grains in the Rolling-Transverse Direction (RD-TD) plane. The average grain sizes are estimated at 25, 30 and 130 μm for 304 L, 316 L and 316 L(N) steel respectively (Σ3 annealing twins excluded). Transmission Electron Microscopy (TEM) analysis of the as received materials reveals a low density of initial dislocations typical of an annealed microstructure. An example of EBSD map and TEM characterization for 316 L(N) grade is given in Fig. 2.

Axisymmetric notched specimens with a 4 mm diameter and a 15 mm gauge length were machined from 10, 15 and 30 mm-thick plates of 304 L, 316 L and 316 L(N) respectively. The sampling