Materials and Design 89 (2016) 759–769

Contents lists available at ScienceDirect

Materials and Design

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

# Influence of thermo-mechanical embrittlement processing on microstructure and mechanical behavior of a pressure vessel steel

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ARTICLE INFO

*Article history:*
Received 11 June 2015
Received in revised form 30 September 2015
Accepted 5 October 2015
Available online 9 October 2015

*Keywords:*
SA508-IV
Thermo-mechanical embrittlement processing (TMEP)
Precipitates
Reactor pressure vessel steel
Ductile to brittle transition temperature (DBTT)

ABSTRACT

A thermo-mechanical embrittlement processing (TMEP) consisting of thermal aging and cold strain could cause the deterioration of reactor pressure vessel (RPV) steels in the form of an increase in the ductile to brittle transition temperature (DBTT) and a decrease in the upper-shelf energy (USE). In this study, the TMEP was employed to investigate the microstructure and the evolution of mechanical behavior of the SA508-IV pressure vessel steel. In the microstructure of the as-received state, Cr and Mn atoms replace Fe atoms and form alloying cementites, $(Fe,Cr)_3C$ and $(Fe,Mn)_3C$, through in-situ nucleation. Due to the slower diffusion coefficient, Cr precipitates in the outer layer of the Mn clusters. In the subsequent embrittlement process, needle-shaped $Mo_2C$, fine copper-rich precipitates (CRPs) and P-rich precipitates are formed, which play a great role in the mechanical behavior evolution. Mechanical test results show that a series of changes in mechanical behavior occurred. It has been found that DBTT fits a linear function of the square root of embrittling time at 520 °C ($t^{1/2}$) from 10 h to 90 h degradation and the degree of embrittlement reaches saturation after 90 h.



## 1. Introduction

A main objective for the nuclear industry is to enhance the safety and the reliability of nuclear power plants and to extend their operation lifetime. The reactor pressure vessel (RPV) steel plays the most important role in safety rankings of the whole nuclear power plant system. The SA508-IV Ni–Cr–Mo low alloy steel has excellent mechanical behavior [1], which possesses higher strength and fracture toughness by adding Ni and Cr elements [2]. Neutron irradiation has been considered as the main factor responsible for the embrittlement of RPVs [3]. As the experiments under neutron irradiation are not easy due to the radioactivity, the thermo-mechanical embrittlement processing (TMEP) method has been chosen to study the performance of RPVs.

Neutron irradiation is generally known as a major reason for the embrittlement of RPV steels because of the damage produced in collision cascades consisting of self-interstitial atoms and vacancies. The neutron irradiation embrittlement of RPV steels is known to be related to the presence of residual elements [4], such as Phosphorus, which can also be obtained from long-term tempering [5,6]. Copper-rich precipitates (CRPs) are also considered as the reason for irradiation embrittlement [7]. In the temperature range of 300–500 °C, temper embrittlement has been considered as the main factor responsible for the loss of toughness and the premature failure of steels [8,9], which is a non-hardening embrittlement caused by the grain-boundary segregation of impurity elements, mainly phosphorus [2].

Currently, the nature and mechanisms of irradiation embrittlement, especially the changes in the microstructure and mechanical behavior of materials, are the subjects of major studies. However, as neutron irradiation is harmful, the thermo-mechanical process, thermal aging and cold strain were employed to deteriorate the RPVs to simulate the effect of neutron irradiation. To be specific, the temper-embrittlement process could be combined with cold strain, which is called the TMEP procedure. The heat treatment can produce a larger concentration of vacancies and vacancy clusters that are supposed to accelerate the formation of CRPs based on a solute-vacancy exchange diffusion mechanism [10]. After that, the cold strain may cause the interaction of dislocations and precipitates, and then the steel could be embrittled by the large numbers of dislocation loops forming behind them.

In the previous study, the pre-strain followed by aging process was applied to study the microstructure evolution of RPV steels. Ghosh et al. [11] have found that the 50% pre-strain prior to aging could trigger the precipitation by decreasing the activation energy values for Cu precipitation from 182 to 126 kJ/mol. Kamada et al. have conducted a series of experiments to simulate the irradiation embrittlement of nuclear RPV steels [12]. The Fe-1 weight percent (wt.%) Cu alloys with and without pre-deformation in the solid-state solution were thermally aged at 773 K (500 °C) for various times. After that, the evolution of hardness,

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http://dx.doi.org/10.1016/j.matdes.2015.10.024
0264-1275/

conductivity, and microstructure were investigated. It has been found that the pre-deformation enhanced Cu precipitation and caused precipitation at dislocations.

Although the microstructure evolution of the above methods was close to the results of the neutron irradiation state, the mechanical tests could not obviously show the embrittlement of RPVs. Wu et al. [13] studied the TMEP process, applying the 8% uniform cold strain following the heat treatment, to simulate the effect of neutron irradiation on the mechanical properties of a pressure vessel weld metal. The fracture tests were carried out using blunt notch four-point-bend specimens in slow bend over a range of temperatures. The ductile–brittle transition temperature (DBTT) was shown to increase by approximately 100 °C as a result of the degradation.

In this study, the TMEP was applied to the new type of pressure vessel steel A508-IV. It has been stated by Fatehi et al. [14] that the precipitation of Cu in steels is usually observed in the temperature range of 400–650 °C. Thus, the current work was conducted at 520 °C using the TMEP deterioration process. It was focused on studying the embrittlement effect of the hardening and grain-boundary P segregation, to compare with the in-service SA508-III and the neutron irradiation embrittlement. The microstructure evolution, which leads to the embrittlement, will be studied in details and the resultant degradation of the mechanical behavior will also be discussed.

## 2. Experimental procedure

### 2.1. Testing materials and specimens

The chemical composition of the SA508-IV Ni–Cr–Mo low-alloy steel is listed in Table 1. As shown in Fig. 1, the specimens were subjected to a heat treatment of 1.5 h at 920 °C, followed by water quenching (WQ) to room temperature (RT), re-heating to 650 °C for 30 h and furnace cooling to RT. Specimens without the further TMEP treatment are termed as "the as-received state". The degradation procedure has been worked out with aging, followed by the 10% uniform cold strain (Fig. 1). The cold strain is applied by the compressive stress at the speed of 0.5 mm/min using the 100 t computer-controlled electro-hydraulic servo universal testing machine. The corresponding specimens are termed as "the degraded state".

The above two states of materials are all fabricated into tensile specimens and Charpy-impact test specimens. The tensile specimens are bars with a gage length of 25 mm and gage diameter of 5 mm. The dimension of Charpy-impact test specimens is 10 mm × 10 mm × 55 mm with the notch direction transverse to acting surfaces applied by the compressive stress (Fig. 2).

### 2.2. Mechanical tests

Tensile tests at room temperature were carried out using a computer-controlled 50 kN testing machine with a loading rate of 0.5 mm/min.

Charpy-impact tests were conducted using a 300 J pendulum impact test machine in a temperature range from −160 °C to RT. The test results were used to plot ductile to brittle transition temperature (DBTT) curves.

In order to assess the age-hardening effect, Rockwell C Hardness (HRC) values were measured for each state of samples under a load of 150 kg at 9 positions, and the results are averaged to assure the repeatability.

**Table 1**
Chemical composition of SA508-IV Ni–Cr–Mo alloy steel in wt.%.

| C | Si | Mn | P | S | Ni | Cr | Cu | Mo | Al | Fe |
|---|---|---|---|---|---|---|---|---|---|---|
| 0.15 | 0.36 | 0.34 | 0.011 | 0.008 | 3.26 | 1.66 | 0.041 | 0.46 | 0.005 | Balance |

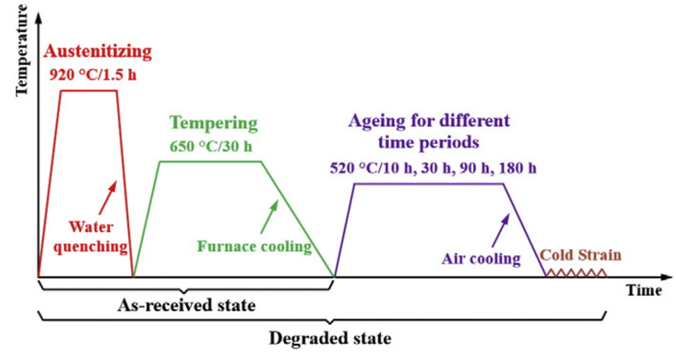


**Fig. 1.** Schematic of the degradation process.

### 2.3. Microstructure analysis

After standard grinding and polishing, the samples were etched by 5 volume percent (vol.%) nital. The microstructures were examined using the optical microscopy (OM) and scanning electron microscopy (SEM).

Transmission electron microscope (TEM) was employed to examine the fine details of the microstructure and to obtain the diffraction pattern of the selected phases. The TEM specimens were prepared using a double-jet electrolytic thinning technique. The images were taken under both TEM and scanning transmission electron microscope (STEM) mode.

## 3. Results and discussion

### 3.1. Microstructural observation

#### 3.1.1. Undegraded state

Figs. 3(a) and (b) are SEM micrographs of the undegraded (as-received) samples of SA508-IV. Fig. 3(a) shows that it has an overall martensitic structure. In Fig. 3(b), it could be found that there are some coarse carbides precipitated along grain boundaries. According to the research of Lee et al. [15], they should be the $M_3C$-type carbides. Then based on the study of Hashimoto et al. [16], the carbides distributed along grain and/or lath boundaries could be defined as $M_{23}C_6$ and Cr-, Ni-rich $M_6C$. The fine $M_2C$-type carbides are also dispersed inside laths, similar to the results mentioned in Refs. [13] and [14]. In Fig. 4, it can be seen that the plate-shaped Cr-rich $M_{23}C_6$ carbides have a diameter of about 142 nm [Fig. 4(a)], and the rod-shaped $M_6C$ is about 129 nm in length [Fig. 4(b)]. Besides, a few MC precipitates mainly distribute along dislocations (see Fig. 5).

In the tempering process, the alloying elements diffuse quickly and re-locate inside α-Fe and $Fe_3C$ phases. It can be obviously seen from the STEM distribution diagram of elements (Fig. 6) of the undegraded

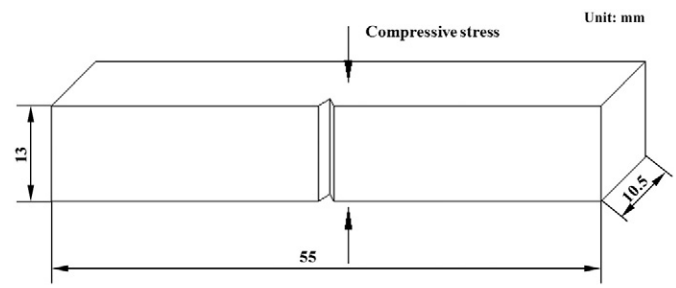


**Fig. 2.** Position relationship of the compressive stress and notch location.