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Possibility of LRO of long term operated Alloy 690 SG tubings

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1. Introduction

According to the trend of long-term operation of nuclear power plants for more than 40 years, SRO/LRO is drawing attention of some researchers.[1-3] Alloy 690 has a ratio of Cr and Ni close to Ni_2Cr, an intermetallic compound that causes SRO/LRO, so it is a material to be handled more carefully than alloy 600. In addition, the variables affecting LRO are not well understood, and there are not many studies on the reaction rate according to time and temperature. It is necessary to evaluate the LRO potential of the alloy 690 steam generator tubings of Kori unit 1, which was replaced in 1998 and operated for 166,000 hours only considering the simple calendar time until retirement in 2017.

In this paper, the LRO possibility of the alloy 690 SG tubing material of Kori unit 1 is analyzed. These analysis can be used as important data to evaluate the material integrity of steam generator tubings of other domestic nuclear power plants that are preparing for long-term operation.

2. SRO/LRO mechanism

Long Range Order (LRO) in Ni alloys is that Ni and Cr prefer definite positions relative to each other over the entire lattice, and Short Range Order (SRO) represents that the positions of Ni and Cr are non-random statistically, but the atoms are not in definite positions relative to each other over the entire lattice.

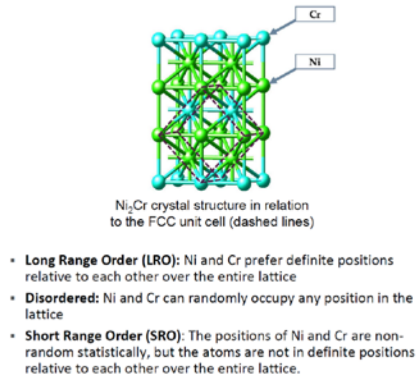

Fig. 1. Schematic of ordering structure of Ni alloys. [4]

LRO refers to the Ni_2Cr (γ') phase that can be formed in Ni-Cr alloys such as alloy 600 or alloy 690 and its weld materials, as shown in Figure 1.

Above the critical temperature, typically between 525-575 °C, it should be seen that LRO no longer occurs because nucleation decreases. When the temperature of ordered alloy rises above the critical temperature, a disordering reaction, which is the reverse reaction of the ordering reaction, occurs. In this case, the ordered Ni_2Cr phase loses its ordered structure and disperses excess chromium.

SRO can further transform into LRO with prolonged exposure to temperatures below 550 °C. The LRO follows a process of nucleation and growth, which can be homogeneous or heterogeneous, and the rate of nucleation increases with decreasing temperature. The growth of nuclei is slower at lower temperatures because it relies on atomic diffusion.[5]

Roman Mouginot reported that SRO may occur in alloy 690 during the initial stage of degradation, furnace cooling, solution heat treatment (SA) after water cooling, deformation or irradiation.[6] Marucco also reported that LRO is generated near the Ni_2Cr composition (30.7 wt.% Cr) and the Ni_3Cr composition (22.8 wt.% Cr) in the other literature.[7]

In the case of LRO, the formation is slow even near the basic composition, so at least 30,000 hours are required, and in general, about 60,000 to 100,000 hours of degradation time is required.[5]

When SRO/LRO occur, the lattice contraction is expected to cause a change in the mechanical properties of the material, and the lattice contraction can reach 0.25%, leading to a dimensional change of the structure.[5]

George A. Young et al. evaluated the LRO rate by heat-treating a model Ni-Cr alloy for up to 10,000 hours.[8] He confirmed that LRO could develop over a wide range of compositions, indicating that alloy 690 is not free from LRO. However, with some accelerating factors excluded, it has been concluded that LRO can be problematic for parts exposed to high temperatures (325°C) for more than 10 years.

3. Evaluation of ordering reaction

It is difficult to observe this ordering reaction experimentally and sometimes it is difficult to distinguish it from other metallurgical changes. As such, SRO/LRO of Ni-based alloys should be treated as important because they can be linked to dimensional change and internal stress increase, starting from lattice shrinkage, and material brittleness.[8]

During SRO, the size of the order domain does not exceed several nanometers, making it difficult to directly observe.[7] A differential scanning calorimetry (DSC) test applied to alloy 600 showed an exothermic ordering reaction at about 520 °C.[9]

The possibility of LRO can be also evaluated in an indirect way by increasing the microhardness. In the case of Ni_2Cr + 7Fe alloy, LRO occurs at a temperature below 450 °C and hardness may increase, but at 333 °C, LRO was not observed even after heat treatment for 10,000 hours.[4]

The degree of lattice contraction can be evaluated by the X-ray diffraction method.[4] Since the ordered phase has a smaller lattice parameter than the disordered phase, the degree of ordering can be known by evaluating it.

4. Possibility of Alloy 690 tubings in Kori unit 1

The alloy 690 tubings of the steam generator used in the Kori nuclear power plant unit 1 was replaced in 1998 and exposed to a primary side operating temperature of about 320°C for about 19 years (estimated EFPY 15.2) until decommissioned in 2017. Based on the analysis as shown in Figure 2, it is evaluated as the boundary time at which LRO may occur.

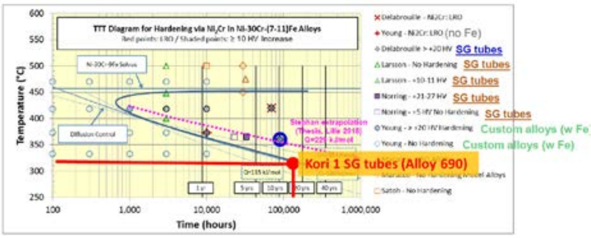

Fig. 2. Possibility of alloy 690 tubings in Kori unit 1(Reproduced based on Ref.4)

However, based on the data in Figure 3, it is judged that the time and temperature are not sufficient conditions for LRO to occur in the Alloy 690 heat transfer tube of the Kori unit-1.

5. Summary

- There is a lot of interest in whether alloy 690 will cause such LRO/SRO at the operating temperature of a nuclear power plant and not embrittle the material.
- LRO / SRO may be formed when alloy 690 is heat treated in the temperature range of 360 - 450 °C.
- The possibility of LRO/SRO seems low in Kori 1 alloy 690 material operated at 320 °C for 19 years.

ACKNOWLEDGEMENTS

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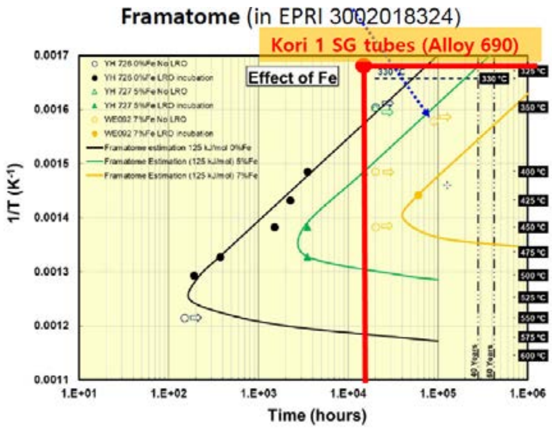

Fig. 3. Time and temperature are not sufficient for LRO of Alloy 690 tubings in Kori unit 1(Reproduced based on Ref.4)

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