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Letter to the Editor

Suppression of ambient temperature creep by eutectic phase for hexagonal close-packed metal

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ABSTRACT

Zr–Si alloys were designed to contain eutectic phase surrounding the parent phase to suppress creep behavior of claddings. Creep tests conducted at 294–573 K showed that creep behavior was inhibited and that the creep failure time of new Zr alloy became longer than that of a conventional alloy: Zircaloy-4. Results show that the eutectic phase can suppress creep at operating temperatures prevailing in current nuclear power plants.

1. Introduction

Zirconium alloys have been used as components of nuclear power plants because of their good mechanical properties and small absorption cross-section for thermal neutrons at operating temperatures [1–9]. Therefore, their creep properties have been investigated carefully at 536–1873 K for about half a century. However, the present author reported that hexagonal close-packed (HCP) materials show creep behavior with a stress exponent of 3 and a grain-size exponent of zero at less than their 0.2% proof stresses ($\sigma_{0.2}$) at $T < 0.3\ T_M$ (T_M, melting temperature) [10–12]. Dislocations are aligned as straight with no tangles in this condition. Therefore, it is claimed that the dislocations move freely inside of grains. Subsequently the dislocations pile up at grain boundary and are absorbed into it, bringing about the creep phenomenon [11]. In addition, because the apparent activation energy is one-fourth that of dislocation core diffusion [1], i.e., about 20 kJ/mol [12], the dislocations might be accommodated by a non-diffusion process, i.e., slip-induced grain-boundary sliding (GBS) [13,14], to control the creep rate.

However, it has been considered that creep does not occur because the elastic or low-temperature dislocation creep regime is spread widely at operating temperatures according to deformation mechanism maps of Zr and its alloys in this temperature region [1,2]. Therefore, the creep behavior described above was ignored when the maps were constructed. Confirming and suppressing the creep behavior at operating temperatures of nuclear power plants is necessary to ensure the safety of nuclear power plants. This paper describes Zr–Si alloys designed to improve the creep properties of Zr alloy by the eutectic phase ($Zr_3Si + \alpha$-Zr) covering the parent phase (α-Zr). Moreover, Zircaloy-4 was used as a reference material in this study because it is a material used currently in existing water reactors. In this study, the samples were recrystallized to ascertain the effects of the eutectic phase on mechanical properties.

2. Experimental procedure

Zr-0.5 and 1.5 Si [weight %] alloys were manufactured using 99.9% Zr and 99.999% Si by three-time arc melting in argon atmosphere. Then they were annealed at 873 K for 3 h at 2×10^{-4} Pa before subsequent tests. Scanning electron microscopy (SEM) was conducted to ascertain the alloy microstructure. Fig. 1(a) and (b) respectively show backscattered electron images of Zr-0.5 and 1.5 Si alloys, where bright regions correspond to the parent phase and dark regions are the eutectic phase. The images show that the eutectic phase covers the parent phase clearly and that the phase thickness increases with increasing Si content. In addition, the parent phase grain sizes were about 15 μm for each sample, as evaluated by the intercept length. On the other hand, Zircaloy-4 was recrystallized in same condition. Its grain size was about 30 μm. An optical micrograph (OM) of the surface is also presented in Fig. 1(c).

Next, tensile tests were executed at a constant cross-head speed corresponding to an initial strain rate of $1 \times 10^{-4}\ s^{-1}$ to evaluate 0.2% proof stresses ($\sigma_{0.2}$) and tensile strength (σ_B) for all samples. Then, creep tests were performed with dead-load creep frames at 294–573 K in air. The gauge length, width, and thickness were 6.0, 1.2, and 0.50 mm, respectively. Tensile and creep strains were measured using strain gauges mounted directly on the specimen surface at 294 K and an eddy current displacement sensor at >423 K.

3. Experimental results and discussion

Fig. 2 shows $\sigma_{0.2}$, σ_B and failure strain (ε_f) as a function of Si contents at 294 K. Results show that $\sigma_{0.2}$ and σ_B increased but ε_f

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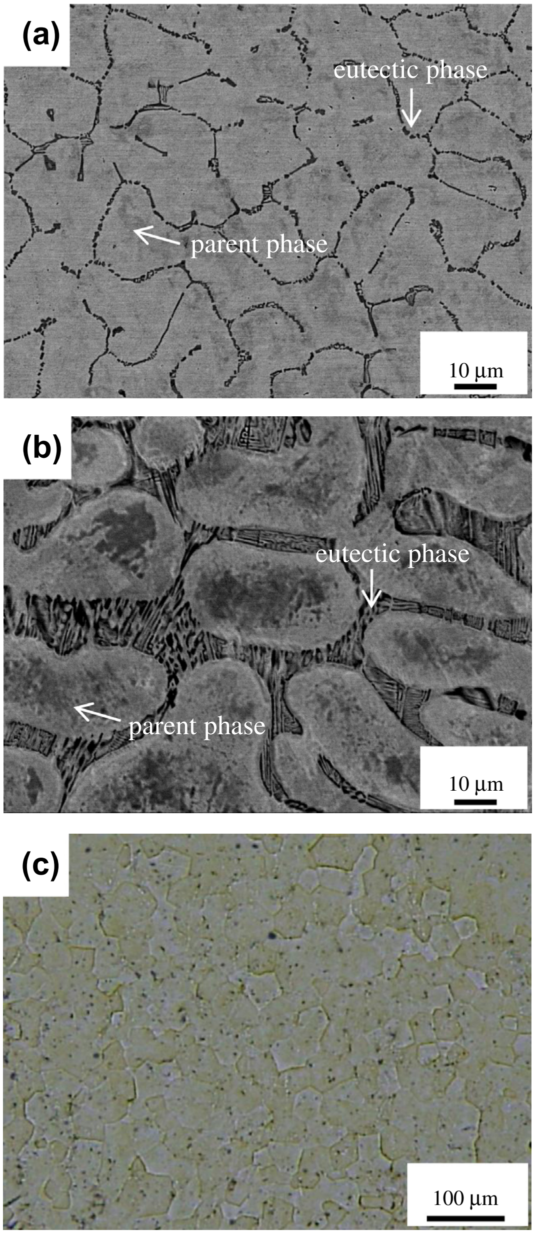

Fig. 1. Backscattered electron images of Zr- (a) 0.5 and (a) 1.5 Si. Darker regions correspond to the eutectic phase, whereas the brighter regions are the parent phase; (c) shows an OM image of Zircaloy-4.

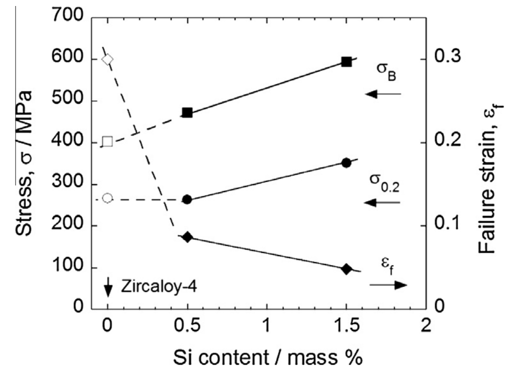

Fig. 2. Relation between $\sigma_{0.2}$, σ_B and ε_f and Si contents of Zr–Si alloys as closed symbols. These values of Zircaloy-4 are also shown as open symbols. The strength increased but the ε_f decreased concomitantly with increasing Si contents.

decreased with increasing Si content. These parameters of Zircaloy-4 are shown as open symbols in Fig. 2. Comparison to data of the alloy shows that Zr–Si alloys were strengthened, but they become brittle with increased amount of the eutectic phase.

After the tensile tests, SEM observations were performed for Zr–Si alloys. Fig. 3 shows backscattered electron images near the fracture surface for Zr-(a) 0.5 and (b) 1.5 Si, which shows cracks only in the eutectic phase. Moreover, Fig. 3(c) and (d) depict top views of fracture surfaces of Zr-0.5 and 1.5 Si, respectively. Because dimples were not observed for each sample, fracture occurred along the eutectic phase by propagation and conjugation of the cracks, confirming that Zr–Si alloys are more brittle than Zircaloy-4.

Creep curves of Zr–Si alloys and Zircaloy-4 at (a) 294 and (b) 573 K are portrayed in Fig. 4. Although remarkable creep behavior was observed for Zircaloy-4 at both temperatures, it was suppressed dramatically for Zr–Si alloys at the same stress level. The creep strain decreased concomitantly with increasing Si content.

Using the creep data, a double logarithmic plot of the modulus-compensated applied stress and strain rate is depicted in Fig. 5. It shows a stress exponent of about 3 for Zr-1.5 Si alloy. The n value is the same as that of ambient temperature creep [11,12]. The apparent activation energy is only 16 kJ/mol, as shown in an Arrhenius plot at $\sigma/E = 0.003$ (Fig. 6). Therefore, the observed creep is controlled not by a diffusion process but by dislocation motion and slip-induced GBS, similarly to HCP metals [11,12].

Previous reports have described that deformation twin influences creep behavior in α-Ti alloy at $T < 0.25\ T_M$ [15]. However, its grain size of about 200 μm was larger than that of Zr–Si alloys and other HCP metals [10–12]. According to those earlier reports [10–12,15], dominant intragranular deformation mode might change by grain size: fine-grained samples creep by dislocation whereas coarse-grained samples creep by twin. Therefore, it was confirmed that dislocation motion generates deformation in Zr–Si alloys with grain size of 15 μm.

After creep tests at 573 K, SEM observations reveal the effects of the eutectic phase on creep at σ = 193 MPa and ε = 0.015 in Zr-1.5 Si alloy. The accelerated creep region was not observed, and backscattered electron images of the Zr–Si alloy show cracks (Fig. 7). However, it did not conjugate like the tensile specimen (Fig. 3). It can be inferred that the eutectic phase is effective for creep suppression at $T < 573$ K.

Moreover, the creep failure time of Zr–Si alloys lengthens by at least 10^4 s compared to that of Zircaloy-4 at 573 K (Fig. 8). The creep is brought about by dislocation motion and slip-induced GBS, as described in HCP metals [12–14]. Therefore, it is considered that the creep suppression is caused by retarding dislocation motion at grain boundaries [16].

4. Conclusion

Tensile, creep tests and SEM observations were performed to assess the mechanical behavior of Zr–Si alloys at 294–573 K. The new alloys' strength became greater than that of Zircaloy-4. Creep strain was suppressed and decreased concomitantly with increasing Si contents for Zr–Si alloys. The creep failure time therefore became much longer than that of Zircaloy-4. Moreover, SEM observations revealed cracks only in the phase, confirming that the eutectic phase retards dislocation motion, thereby giving high strength. Results demonstrate that the eutectic phase can suppress creep and improve the properties effectively at operating temperatures prevailing in nuclear power plants.