Annihilation behaviour under electron irradiation of athermal $\omega$-phase crystals formed by cooling at 131K in a $\beta$ Ti-Mo alloy

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Abstract. Formation of athermal $\omega$-phase crystals due to cooling to 131 K has been directly observed in a $\beta$-type Ti-15mass%Mo alloy. The athermal $\omega$-phase crystals easily disappear by electron irradiation during the in-situ observation at 131 K. Incubation phenomenon of the annihilation is also recognized. The annihilation behaviour was investigated based on the dependence on electron irradiation conditions and incubation phenomena. It is concluded that the annihilation mechanism is concerned with interactive effects of temperature rise due to electron irradiation and collective oscillation resulted from inelastic scattering of electron beam.

Introduction
Negative temperature dependence of electrical resistivity in $\beta$-Ti and Nb alloy systems has been reported, and the origin has been postulated on the formation of athermal $\omega$-phase crystals due to cooling [1-3]. The present authors have reported that a direct observation of the formation of postulated athermal $\omega$-phase crystals has been achieved in a Ti-15mass%Mo alloy cooled to 131 K using electron microscope technique [4, 5]. At the same time, it has been also mentioned that (1) atomic structure of athermal $\omega$-phase crystals consists with atomic structures of $\omega$-phase crystals formed by aging and stressing, and (2) the athermal $\omega$-phase crystals easily disappear by electron irradiation during the in-situ observation, which means that an inverse, $\omega$-$\beta$ phase transformation occurs due to electron irradiation.

The occurrence of phase transformations caused by electron irradiation under different conditions has been reported in many kinds of alloy systems, and it is considered that the phase transformations are resulted from one of factors such as radiation damage, radiation enhanced and induced diffusions, and thermal diffusion due to temperature rising, or their interactive effect(s) [6, 7]. In the previous paper, the main factor for the annihilation caused by electron irradiation has been discussed on temperature rise and radiation enhanced diffusion [5]. Radiation damage has not been considered as a main factor, since the used electron microscope was operated at 200 kV and the threshold voltage for atomic displacement for Ti and Ti alloys is over 400 kV [8]. But, a reasonable conclusion has not been taken.

In the present paper, a predominant factor on the onset of annihilation mechanism of athermal $\omega$-phase crystals due to electron irradiation at 160 kV and 200 kV has been discussed, because a clarification of the predominant factor might be important to understand one of the factors of the onset of $\omega$-$\beta$ phase transformation, and the relationship between electron irradiation and phase transformations. The
discussion has been done based on experimental results about (1) a dependence of onset times of 
annihilation of athermal \( \omega \)-phase crystals upon electron irradiation conditions, and (2) the existence of 
incubation time to start the annihilation.

2. Experimental procedures
A Ti-15mass%Mo alloy was used for the specimen, which was solution-treated at 1073 K for 3.6 ks in 
vacuum, followed by water quenching. Thin foil specimens for electron microscope observations were 
prepared using twin-jet electropolishing method. A specimen holder, EM-31100 was used for \textit{in-situ} 
observation at 131 K, and for controlling specimen temperatures. A JEM 2010F electron microscope 
operated at 160 kV and 200 kV was used. Dark field images and high resolution electron microscope 
(HREM) image observations were carried out at 131 K. Results of \textit{in-situ} dark field image 
observations during cooling and heating processes were recorded using a TV-VTR system. Electron 
irradiation conditions were adjusted by changing acceleration voltages to 160 and 200 kV, and 
electron beam currents of approximately 20 and 5 pA/cm\(^2\) were also adjusted by changing condenser 
apertures and condenser lens.

3. Experimental Results
In order to consider a predominant factor for the annihilation behaviour due to electron irradiation, the 
annihilation behaviour under four different irradiation conditions has been investigated. Specimens 
were cooled down to 131 K and held in the temperature for 10.8 ks without electron irradiation, and 
athermal \( \omega \)-particles of approximately 40 nm in diameter were irradiated (\textit{in-situ} observed). Their 
annihilation behaviour was recorded using a TV-VTR system, and their onset times of the annihilation 
(incubation times) and life times (times until disappearing) were measured by playing back the tapes. 
When \textit{in-situ} observation was carried out while a specimen was cooled down from room temperature 
to 131 K, the formation of athermal \( \omega \)-particles was not observed [5]. Figure 1 shows an example of 
an athermal \( \omega \)-particle and the process of annihilation due to electron irradiation with the condition of 
160 kV and 5.7 pA/cm\(^2\) [5]. It is noted that until irradiating for 255 sec, the size change of athermal \( \omega \)-
particles is hardly recognized. After that, the annihilation due to electron irradiation started. In this 
case, the irradiation time for the onset of annihilation was decided as 255 sec. This result indicates that 
an incubation phenomenon occurs during the annihilation behaviour.

Similar observations have been performed under four different irradiation conditions. On each 
condition, annihilation behaviour has been recorded several times and each onset time was decided.

![Figure 1](image.png)

**Figure 1** Annihilation behaviour of an athermal \( \omega \)-particle formed by cooling at 131 K 
for 10.8 ks due to the irradiation of 160 kV and 5.7 pA/cm\(^2\).
Those results are summarized in Figure 2.

![Figure 2](image)

**Figure 2** Electron irradiation condition dependence of the onset time for annihilation of athermal $\omega$-particles formed by cooling at 131 K for 10.8 ks.

It is found that the incubation times depend on irradiation conditions. The following two results are revealed: (1) In the case of the same acceleration voltage, the onset times of annihilation under the irradiation with lower electron beam current density, 5 pA/cm$^2$ is more than two times longer than those under the irradiation with 20 pA/cm$^2$. (2) In the case of the same electron beam current density, the onset times of annihilation under the irradiation at 200 kV is slightly shorter than those under the irradiation at 160 kV. In the latter case, a significant difference is hardly recognized since the difference of acceleration voltages is smaller compared with the difference between beam currents. It is recognized that both of acceleration voltage and beam current affect the annihilation behaviour.

4. **Discussion**

The annihilation mechanism of athermal $\omega$-particles due to electron irradiation has been considered. The present study has been carried out under the following conditions: (1) under acceleration voltages lower than the threshold voltages of atomic displacement in Ti and its alloys, and (2) at a lower temperature, 131 K. Also the atomic structure of athermal $\omega$-phase is confirmed to consist with de Fontaine’s model [9] using electron diffraction technique and HREM method [4]. Therefore, it is thought that the origin of the annihilation is not due to knock-on phenomenon but due to the effect of energy transfer from electron beam; that is, inelastic scattering between electron beam and atoms. Temperature rise of metallic and semiconductor materials due to electron irradiation has been investigated theoretically and experimentally [10, 11]. Theoretical consideration indicates that the amount of temperature rise ($\Delta T$) is proportional to $I_0\rho/\kappa$, where $I_0$ is the beam current at the specimen, $\rho$ atomic density and $\kappa$ thermal conductivity of the specimen [10]. A temperature rise in the present study was estimated using the experimental conditions. Thermal conductivity of some Ti-Mo system alloys is approximately 7 W/mK [12] (5 W/mK in Ti-6Al-4V at 131 K), and the beam current was $2 \times 10^{17}$ e/cm$^2$s. The amount of temperature rise was 1.46 K. This result leads us to conclude that the temperature rise due to electron irradiation in the present work is not the predominant origin, because it is known that athermal $\omega$-particles formed at 131 K were disappeared at temperature over 149 K by heating-up experiments [13].

It is also found that the annihilation of athermal $\omega$-particles occurs only in a local area irradiated by electron beam [4]. Therefore the annihilation is strongly affected by encountering of irradiated electrons with atoms of crystal lattice. Therefore, this annihilation might be able to divided into two
primitive processes: (1) the first process occurs the moment electrons encounter atoms, and (2) the second process occurs after the first one. The conduction of heat, which is treated in theoretically corresponds to the second process. The first process is thought a phenomenon in tremendously short period. However, irradiated electrons interact with atoms inelastically and these electrons transfer their energy to atoms of crystal lattice continuously. As the result, free energy of the irradiated area might be gradually increased. Two collective oscillation mechanisms are described as the energy transfer mechanisms in a textbook: one is plasmon oscillation between electron cloud around atoms and electron beam, and the other one is phonon oscillation between atoms of crystal lattice and electron beam [14]. The latter one corresponds to heat, treated in the theoretical estimation. By means of these mechanisms, the continuously transferred energy from electron beam is accumulated in the irradiated area. However, energy dispersion due to the second process occurs simultaneously. Experimental results indicated that the incubation periods exist in the annihilation process and depend on the irradiation conditions. In the present study, it is considered that the supplied energy by electron irradiation is lager than the dispersed energy due to the second process. And the moment the free energy of athermal $\omega$-particles increased by the accumulated energy reaches to a critical value, the annihilation starts. Therefore, the origin of annihilation of athermal $\omega$-particles due to electron irradiation is considered as not only thermal activation due to temperature rise but also the energy transferred by inelastic scattering between electron beam and atoms of crystal lattice.

5. Summary

Formation of athermal $\omega$-phase crystals due to cooling to 131 K is recognized in a Ti-15Mo alloy. The athermal $\omega$-phase crystals easily disappear during the in-situ observation even at 131 K. The origin of the annihilation was investigated based on temperature rise due to electron irradiation and the incubation time for the annihilation. It is considered that the annihilation mechanism is concerned with interactive effects of temperature rise due to electron irradiation and collective oscillations resulted from inelastic scattering of electron beam to atoms of crystal lattice.

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