Analytical method and experimental verification for predicting static recrystallization fraction of Zr-1.6Nb-0.2Cu

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1. Introduction

Zirconium alloys have played a key role in the nuclear industry due to the fact that their neutron capture is low as well as excellent corrosion resistance to water or vapor in high temperature and high pressure environment. Excellent physical or chemical properties are required for applications including cladding, grid, and guide tube as components constituting fuel assembly. Basically, those properties of materials depend on the microstructure developed during manufacturing process, such as mechanical and thermal processes[1,2]. Therefore, controlling their microstructure is significant so that fuel assembly components can exhibit their performance.

During the thermal process, annealing, microstructural changes evolve to relieve the internal stress stored by the presence of defects such as vacancy and dislocation induced by cold-work. In this process, the defects are removed by recovery and then dislocation-free grains are formed within the deformed structure by recrystallization in order. These then grow and consume the old grains, resulting in a new grain structure with a low dislocation density[3]. The microstructural evolution usually can be shown as values of measured mechanical properties such as strength and hardness[4].

However, regardless of annealing condition, the behavior of recrystallization in materials is varying from activation energy for recrystallization because they have their own alloying elements and reduction degree.

In the present study, the recrystallization behavior of a certain zirconium alloy was experimentally interpreted with annealing parameter including kinetic and thermodynamic parameter and prediction of recrystallization fraction was analytically performed by using activation energy, and annealing parameter. With this method to predict the amount of recrystallization using the annealing parameter, the practical benefit is obvious when the desired mechanical properties and the degree of recrystallization should be achieved without much experimental effort during fabrication.

2. Experimental procedures

The chemical composition of the test material is Zr-1.6Nb-0.2Cu, which will hereafter be denoted as M25. The alloy was fabricated by VAR(vacuum arc remelting) of nuclear purity sponge zirconium with high purity elements. After the button-type ingot was melted at least four times, the solution treatment at 1020^{o}C for 20min was performed and then it was water quenched in room temperature. Hot rolling by 60% reduction of thickness was carried out after pre-heating to 640 ^{o}C for 30min. 2 times of repeated cold rolling gave 50% reduction of thickness per step except final cold rolling and, finally, resulted in 0.9mm thickness-strip. Among those cold rolling steps, each intermediate heat treatment was done at 580 ^{o}C for 2 or 3 hours in a vacuum of 1 x 10^{-5}Torr. The final reduction degree was 60% by final cold rolling.

Specimens for hardness test and microstructure observation were machined to 7x10mm^2 and sealed in quartz capsules filled with Ar(g). Following the final cold rolling, various heat treatment was performed as final annealing processes at 460~ 580 ^{o}C for 40~ 5000min for the isochronal and isothermal annealing, respectively. The heating rate was 5 ^{o}C/min and the specimens cooled down outside of a furnace after the annealing time. The final annealed specimens were mounted in a certain direction where normal plane to the rolling direction was shown and measured, and then mechanically polished up to 2000grit SiC paper. For microstructure observation with optical microscopy, the polished surface was etched using a solution of 10vol.% HF, 45vol.% HNO_3, and 45vol.% H_2O(distilled water). Optical micrographs were taken under polarized light. A semi-Vickers hardness tester was used to measure the hardness at a 9.8N load. At least 10 hardness measurements were performed for each specimen.

3. Results and discussion

Fig. 1 shows the optical micrographs of the normal plane to rolling direction of strip of cold-rolled M25 with various temperatures and times. Elongated grains exist in the most of 460^{o}C heat treated samples because the as-received material was deformed in the cold rolling process and those were not as fully as heat treated to become equiaxed. However, the grains gradually changed into equiaxed shape as time varying at the temperature, but deformed-shape still remained. It appears to be that the equiaxed recrystallized grains began to evolve at 520^{o}C after 300min. At 580^{o}C, micrographs clearly display a typical recrystallized structure having fully equaxed grains and there was no apparent grain growth as time varies.

However, it is difficult to establish the degree of recrystallization using optical micrographs only. A typical method for establishment of it is hardness

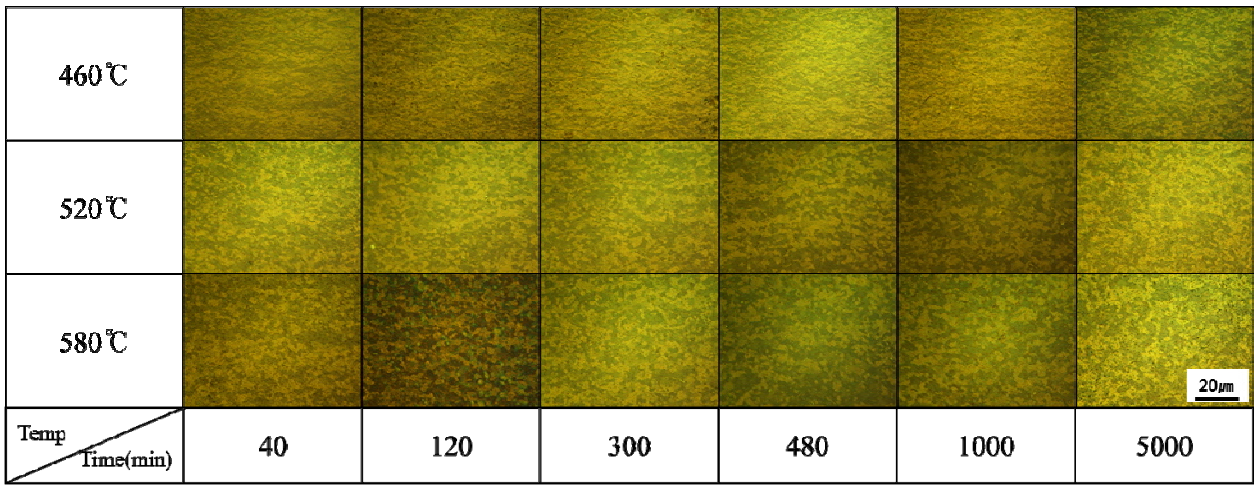

Fig. 1. Optical microstructure for annealed M25 as functions of time and temperature.

measurement and Fig. 2 shows the Vickers hardness values of M25 strip for various temperature and time conditions.

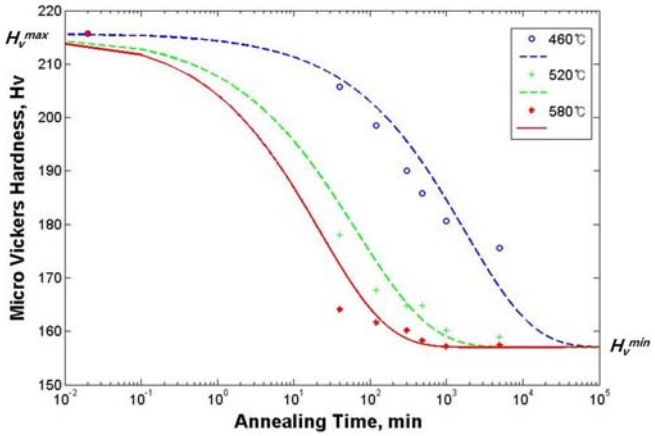

Fig. 2. Variations in the Vickers hardness values of the M25 annealed at various heating condition.

The hardness of the M25 strip decreased as the annealing time increased at each temperature and ranged from 215H_v to 157H_v. It appears to be that recovery and recrystallization in matrix were actively in progress across 100~10000min at 460°C and occurred before 1000min at 520°C, and finished before 500min at 580°C. Furthermore as temperature going up from 460°C to 520°C, its accelerated decrease occurred.

As shown Fig. 2, the decrease of the hardness H_v with increasing time follows an S-shaped curve, where the value of H_v starts from an upper plateau H_v^{max}, followed by a steep decrease, and ends in a second lower plateau H_v^{min}.

This kind of error function shaped curve was kinetically interpreted using JMAK(Jonson-Mehl-Avrami-Kolmogorov) with k and n of temperature dependent constant and exponent depending on the geometry and temperature, respectively[5]. However, through this analytical approach, it is insufficeint to apply time and temperature factor to an integrated parameter, finally for predicting hardness and recrystallization fraction quantitatively with one simple parameter. However, Steinberg *et al.*[6] introduced single technological fabrication parameter called annealing parameter A, normalized annealing time, capable of applying to precipitation and recrystallization.

$$A = t \cdot \exp\left(-\frac{Q}{RT}\right)$$

As shown in the equation, the parameter consists of annealing time, t, and temperature, T, with activation energy for recrystallization, Q and, hence, with this concept, it is possible to simply describe time and temperature factor as an integrated parameter A.

According to Steinberg *et al,* the shape of curves in Fig. 2 is typical for all properties including H_v that change by a process, function of time where the slope of decrease (dH_v/dt) is proportional to the actual value H_v and the difference $(1\text{-}H_v)$ when the process starts from H_v^{max}=1 and ends at H_v^{min}=0. In this case, one can write the differential equation as

$$\frac{dH_v}{dt} = -kH_v(1-H_v)$$

which may be written as

$$\frac{1-H_v}{H_v} = \exp(k \cdot t)$$

This mathematical description explains S-shaped curves with increasing time as presented in Fig. 2.

If one now plots the results as they are shown in Fig. 2 as versus 1000/T(Fig. 3), one expresses that

$$\ln \frac{H_v^{\max} - H_v}{H_v - H_v^{\min}} = -\alpha \frac{1}{T} + \ln \beta$$

where the values of α, slopes of lines, are found to be about 18,800K(Q/R) averaged including activation energy for recrystallization. The expression can also be expressed to

$$\frac{H_v^{\max} - H_v}{H_v - H_v^{\min}} = k \cdot t \cdot \exp\left(-\frac{Q}{RT}\right)$$

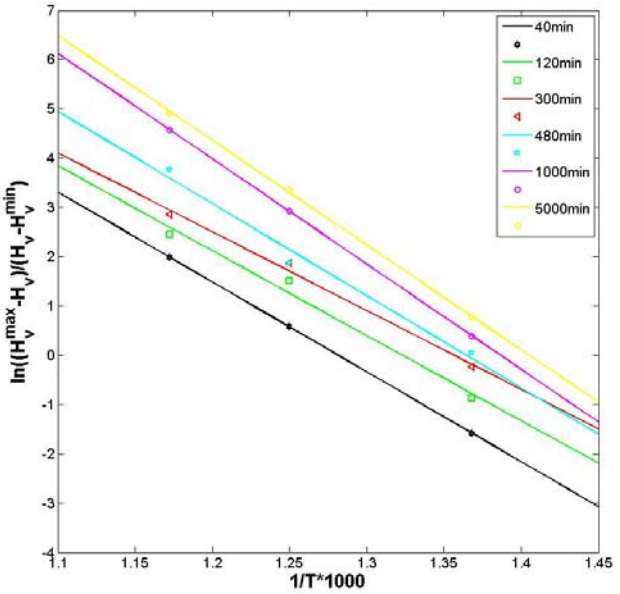

Fig. 3. Correlation between hardness and temperature according to the function $ln(H_v^{max}-H_v)/(H_v-H_v^{min}) = -\alpha \cdot 1/T+ln\beta$.

referred to "Sherby-Dorn parameter"[7] and the expression may now be interpreted as below description.

$$\frac{H_V^{\max} - H_V}{H_V - H_V^{\min}} = k \cdot A$$

In Fig. 4, experimental results with increasing annealing parameter, A, are represented and those are well fitted with just one straight line when the correlation of experimental hardness results and annealing parameter is used for the plotting for all annealing times and temperatures.

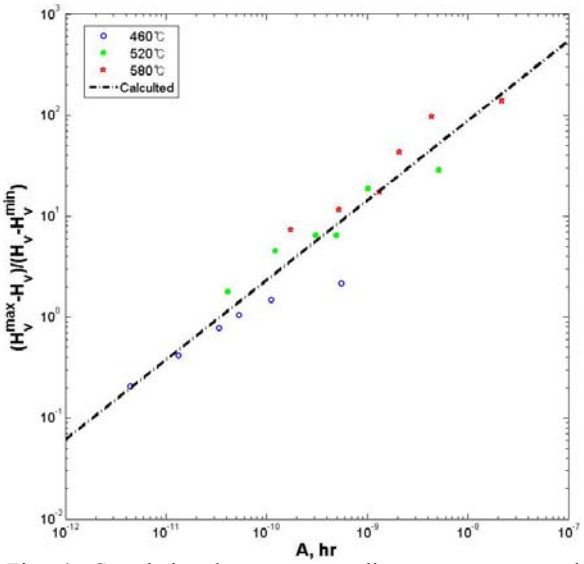

Fig. 4. Correlation between annealing temperature and time expressed as $A=t \cdot exp(-Q/RT)$ and the hardness expressed as $(H_v^{max}-H_v)/(H_v-H_v^{min})$.

For practical uses, those results can be shown as a correlation between H_v and A or f_{rex}(recrystallization fraction) and A. By algebraic transformation the correlation of hardness and A could be described as

$$H_V = H_V^{\min} + (H_V^{\max} - H_V^{\min}) \cdot \frac{1}{1+k \cdot A}$$

and

$$f_{rex} = \frac{H_V^{\max} - H_V}{H_V^{\max} - H_V^{\min}} \times 100,\%$$

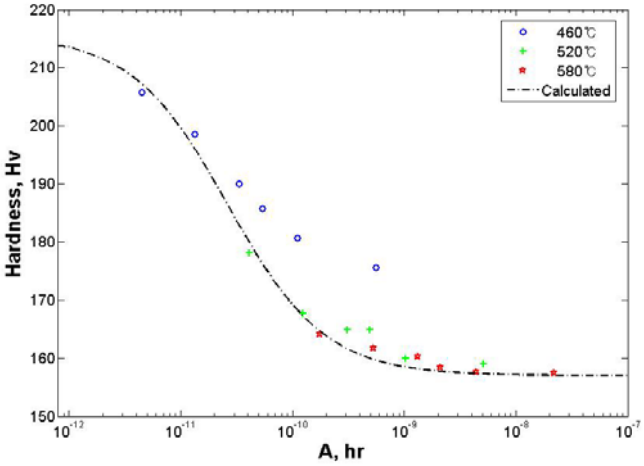

Fig. 5. Dependence of hardness on the normalized annealing time A(cold work:60%).

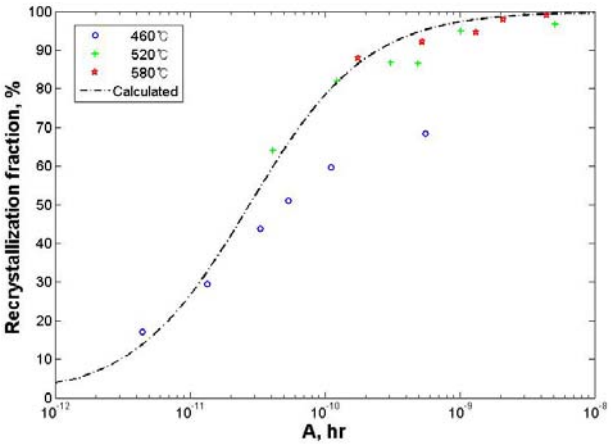

Fig. 6. Dependence of recrystallization fraction on the normalized annealing time A(cold work:60%).

Ultimately, with this correlation of hardness and A as shown in Fig. 5 and 6, analytical plottings of H_v versus A and f_{rex} versus A are possible and most of experimentally measured values fit well with each curve calculated, except 460°C results. It is assumed that 460°C was too low to evoke recrystallization in 60% cold-worked M25 and just microstructural recovery happened out of the lack of hardness drop.

Based on this approach, the combined influence of annealing time and temperature can be acquired as annealing parameter A and hardness and the amount of hardness can be quantitatively calculated through just certain results without much experimental effort during fabrication.

4. Conclusions

In this study, the recrystallization behavior of Zr-1.6Nb-0.2Cu(M25) strip was investigated isothermally and isochronally. Microstructure observation and hardness test were performed and as a result, hardness and recrystallization fraction were calculated using correlations introduced by Steinberg. By the correlations proposed, it will be possible for instance to optimize the needs for mechanical properties. This study proved that the degree of recrystallization can be acquired analytical method obtained from experimental approach.

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