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Comparisson of Recrystallization Kinetics and Grain Growth in Polycrystalline Shape Memory Alloys

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Abstract: The non-ferrous SMAs (shape memory alloys) have, normally, two problems that hinder the use in industrial scale: the natural aging and grain growth. The first degrades the memory effect, while the second, observed during the alloy's mechanical processing, modifies the phase transformation temperatures. Thus, the study of recrystallization kinetics is important for enabling the control of hardened state as a function of treatment time without allowing the exaggerated grain growth. The objective of this study is to determine the recrystallization kinetics in different SMAs (Cu-14Al-4Ni, Cu-12Al-0.5Be and Ni-42Ti), based on an empirical law of J-M-A (Johnson-Mehl-Avrami), as well as their activation energies for grain growth process according to the empirical Arrhenius law.Quantitative evaluations of the grain growth kinetics over a wide range of indicated DSC (differential scanning calorimetry) temperatures have been performed. The results show that the alloy less susceptible to aging in temperatures below the recrystallization peak is the Ni-42Ti, because it presented the highest activation energy, followed by the Cu-14Al-4Ni. The equations that describe the recrystallization kinetics follow the empirical law of J-M-A. The recrystallization kinetics accompanied by hardness variation was an important tool, working as an advisor for selection of treatment time as a function of temperature.

Key words: SMAs, NiTi, CuAlNi, CuAlBe, recrystallization kinetics, grain growth.

Nomenclature

DSC	differential scanning calorimetry
SMA	shape memory alloy
J-M-A	Johnson-Mehl-Avrami
Y_{REC}	recrystallized fraction
E_A	activation energy
D	final grain diameter
D_0	initial grain diameter
T	temperatures
t	time
K, n	kinetics parameters from JMA's equation
k_o	kinetics parameter Arrhenius equation
exp	exponential

1. Introduction

The SMAs (shape memory alloys) are a group of metal materials capable of recovering the original shape (shape memory effect) by imposing a temperature field, due to induced phase transformations in the material and the thermoelastic properties of superelasticity [1]. The most commonly used alloys are Ni-Ti, Cu-Al-Ni, Cu-Al-Be and Cu-Al-Mn alloys. In these alloys, Ni-Ti alloys have better thermoelastic properties, such as shape recovery and superelasticity [2]. Although, the properties of Ni-Ti are higher than the ones of Cu-Al-Ni and Cu-Al-Be, but they are interesting because of the lower cost of machining and processing.

In order to amplify this spectrum of multi-applicability of these alloys, studies on the kinetics of recrystallization and aging [3] are necessary in order to adapt the possible combinations, often of the relationship between formability × properties [4]. In this way, it allows the industry to control the state of the alloy as a function of the treatment time (recrystallized fraction), adapting it to the specifications related to the manufacture of the different products, without changing the kinetics of transformation.

As it is very difficult to quantify the recrystallization through direct methods, such as the microstructure's images, it is plausible to use indirect methods, associated to the values of hardness and grain growth, which change during the recrystallization [5].

The recrystallization can be understood as a phase transformation in a solid solution which occurs by nucleation and grain growth. The kinetics of phase transformation of heterogeneous systems is very complex to be treated analytically, hence the need to use empirical methods to study the phenomenon, as proposed by Zener and Johnson-Mehl-Avrami [6].

For the study of the kinetics of recrystallization, this work will be carry out dynamic analyzes in DSC

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(differential scanning calorimetry), to determine the transformation temperatures and the recrystallization time. Then, different samples will be annealed close to recrystallization temperatures, at predetermined time. After treatment, micro hardness tests will be done to plot the Property vs. Treatment Time curves.

Based on the values of micro hardness, treatment time and recrystallized fraction, the recrystallization kinetics will be determined for the property analyzed, according to the Formation Law predicted by Johnson-Mehl-Avrami [6]. For the characterization of grain growth process, analyses were performed in an optical microscopy; all treated samples were examined for statistical metallography, and grain sizes were measured. After measurement, the diagrams $\ln[-\ln(1-Y_{REC})] \times \ln t$ and $\ln(D-D_0)\times(1/T)$ were plotted to determine the parameters of the JMA's equation (Eq. (1)) and the activation energy of the process, according to the empirical law of grain growth of Arrhenius (Eq. (1)), respectively. Finally, $\ln(D-D_0)\times(1/T)$ curves were plotted to establish the kinetics and, based on these diagrams, the activation energy for grain growth was estimated for the alloys analyzed.

$$Y_{REC} = 1 - \exp(-Kt)^n \quad (1)$$

$$D^2 = D_0^2 + k_0 \cdot t \cdot \exp(-E_A/RT) \quad (2)$$

Once the recrystallization kinetics for each specific alloy has been determined, it will be possible to define the ideal treatment conditions (time and temperatures) in order to match the mechanical properties of the desired alloys characteristics. This study will be able to compare the behavior of recrystallization and the aging probability of Cu-14Al-4Ni, Cu-12Al-0.5Be and Ni-42Ti alloys (% by weight), helping to choose which alloy has the better application.

2. Experimental Setup and Results

The Cu-12Al-42Ti-0.5Be and Ni alloys have been received in Ø3/8″ bars and hot rolled in increments of 5% up to 20% of strain. The Cu-14Al-4Ni alloy was vacuum melted in an induction furnace of 24 kVA, using a silicon carbide crucible. The melted alloy was homogenized at 1,223 K for 15 min and was water-cooled at 298 K. So, the ingot was 2% hot-rolled, in order to standardize the thickness and the microstructure. X-ray fluorescence analysis was performed at Cu-14Al-4Ni to verify the chemical composition, shown in Table 1.

2.1 Heat Treatment

DSC (differential scanning calorimeter) was performed to discover the transformation temperatures. Small blocks were cut in small blocks and were soaked at temperatures near the phase transformations and peak of the DSCs (943, 983, 1,023 and 10,063 K for Cu-14Al-4Ni, 763, 788 and 823 K for Cu-12Al-0.5Be and 1,053, 1,083 and 1,118 for Ni-42Ti.

The samples were soaked at those temperatures for 1, 2, 4, 12, 24 and 120 min, to promote recrystallization and grain growth at different time and temperatures. All the experiments were repeated for three times.

Table 1 Chemical composition of Cu-14Al-4Ni.

Element	Cu	Al	Ni
Wt%	82.17	13.87	3.96

2.2 Vickers Micro Hardness Testing

A Vickers micro hardness tester was used in determining the micro hardness of the samples. The tests were ground in polished samples using a load of 300 g for 10 s, performed for 10 times in each sample, always at the grain's center.

The comparative curves of the medium Vickers micro hardness as a function of treatment time, as well as comparative curves of estimated average recrystallized fractions in function of the treatment time are shown in Fig. 1. The property values were correlated with the recrystallized fraction, for the determination of the recrystallization kinetics.

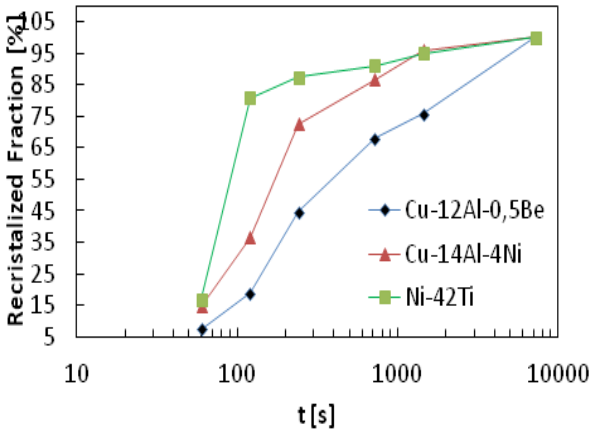

Fig. 1 Estimated medium recrystallized comparative curves of the recrystallized fractions as a function of treatment time.

Micro hardness in all alloys decreases over time of treatment, featuring the recrystallization process.

From the diagrams of Figs. 2-5 it was possible to determine the JMA's equation's parameters shown in Table 2, which describes the kinetics of the recrystallization temperature for each treatment.

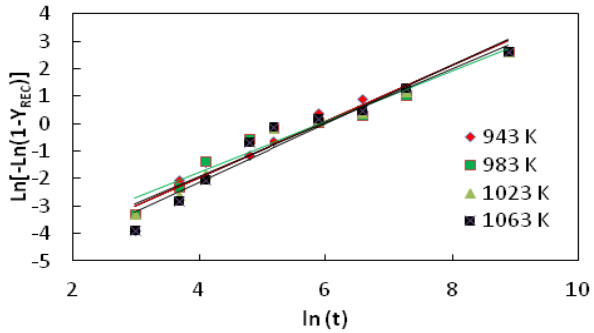

Fig. 2 Diagrams ln[-ln(1- Y_{REC})] × ln*t* for Cu-4Ni-14Al alloy.

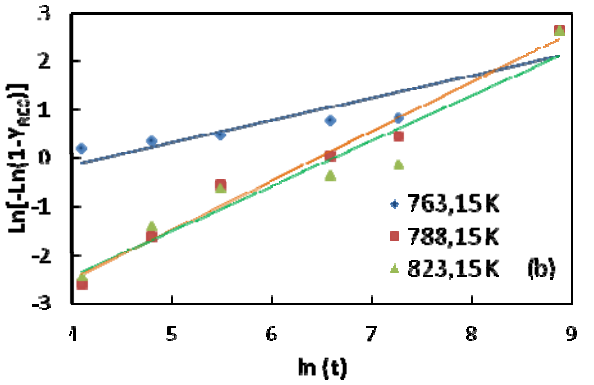

Fig. 3 Diagrams ln[-ln(1- Y_{Rec})] × ln*t* for Cu-12Al-0.5Be alloy.

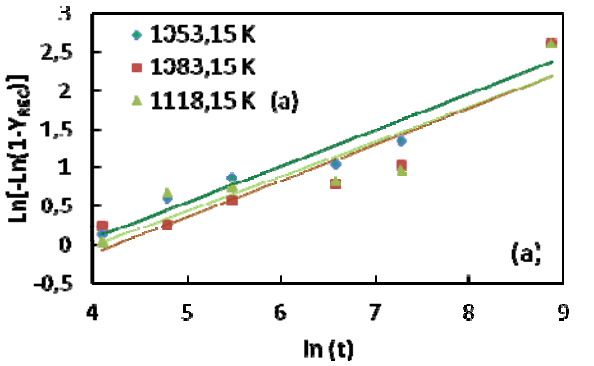

Fig. 4 Diagrams ln[-ln(1-Y_{REC})] × ln*t* for the Ni-42Ti alloy.

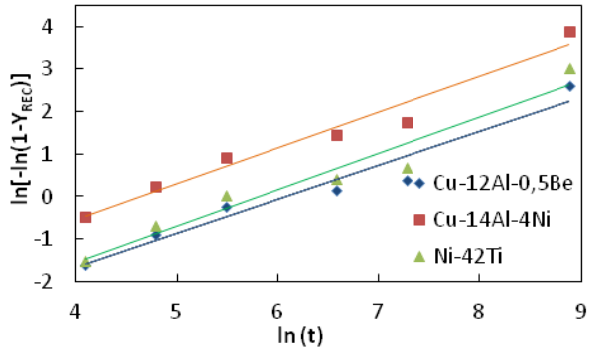

Fig. 5 Comparative media among the 3 alloys' diagrams ln[-ln(1-Y_{REC})] × ln*t*.

Table 2 Kinetics parameters from JMA's equation.

SMA	*T* (K)	*K*	*n*	Correlation factor (R^2)
Cu-14Al-4Ni	943.15	0.0017	0.8692	0.9622
	983.15	0.0026	0.9205	0.9536
	1023.15	0.0026	1.0185	0.9645
	1063.15	0.0024	1.0702	0.9463
Cu-12Al-0.5Be	763.15	0.0139	0.4573	0.8165
	788.15	1.0127	1.0127	0.9734
	823.15	1.0127	1.0127	0.9168
Ni-42Ti	1053.15	0.4689	0.4689	0.9373
	1083.15	0.4703	0.4703	0.8635
	1118.15	0.4496	0.4496	0.8270

2.3 Grain Sizes Measures

For the Cu-14Al-4Ni alloys, under all conditions studied (treatment temperature), high dispersionwas checked in grain size (Fig. 6).

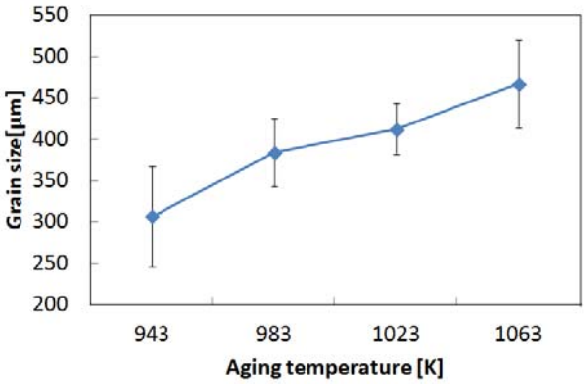

Fig. 6 Grain sizes in function of aging temperature for the Cu-14Al-4Ni alloy.

This dispersion occurs because, during solidification, the mold wall was cold, and the molten metal, in contact with the cold wall solidifies rapidly, causing columnar grain growth at the center of the ingot. This generates a heterogeneity of composition and morphology (due to the different melting points of the components) even when the ingot is thermo mechanically processed, partially recrystallized.

In addition, grain growth in Cu-14Al-4Ni alloy system is favored by low levels of energy activation of the growth process. At elevated temperatures (above 873 K) the secondary recrystallization is easily observed (due to the high atomic diffusion through grain boundaries), characterized by the slope of

diagram ln $(D\text{-}D_0)\times(1/T)$(Fig. 7) and the morphology of some grains. The Cu-12Al-0.5Be and Ni-42Ti alloys have less dispersion in relation to the size of the grains (Figs. 8-11) compared with Cu-14Al-4Ni due of the homogeneity of their manufacturing process.

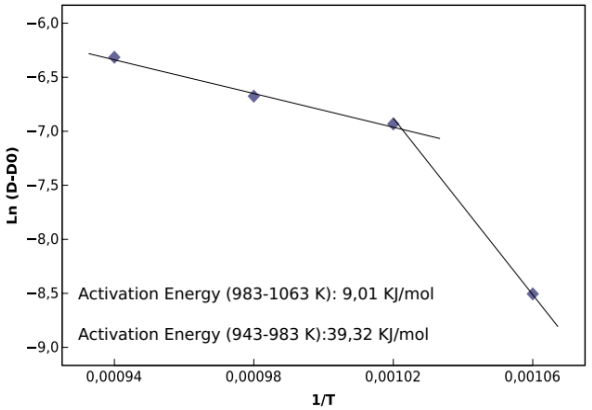

Fig. 7 Diagram ln$(D\text{-}D_0)\times(1/T)$, for the determination the activation energy of the grain growth process for the Cu-14Al-4Ni alloy.

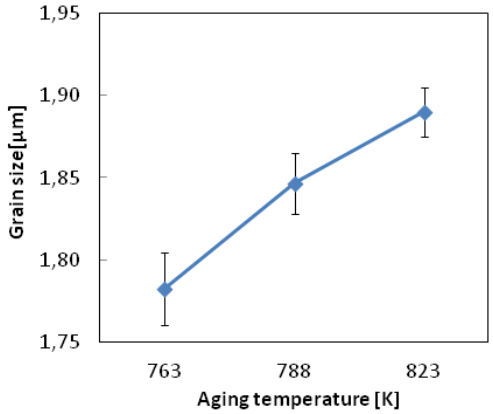

Fig. 8 Grain size as a function of aging temperature for the Cu-12Al-0.5Be alloy.

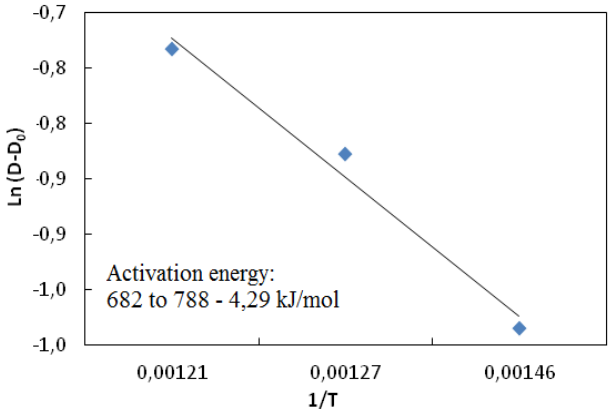

Fig. 9 Diagram ln$(D\text{-}D_0)\times(1/T)$ to determine the activation energy of the grain growth process of the Cu-12Al-0.5Be alloy.

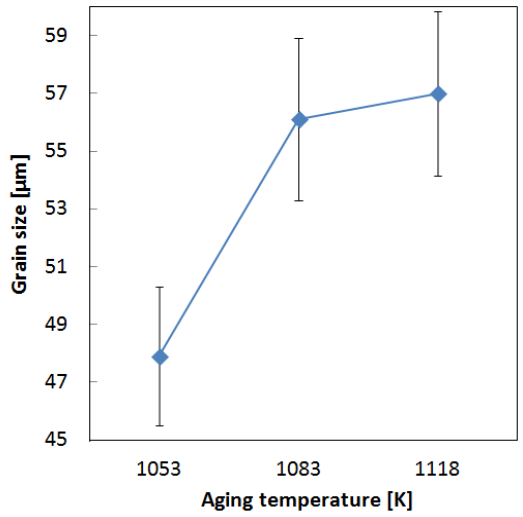

Fig. 10 Grain size as a function of aging temperature of the Ni-42Ti alloy.

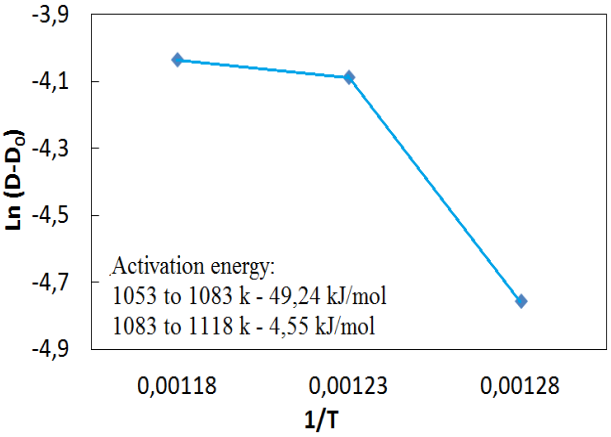

Fig. 11 Diagram ln$(D\text{-}D_0)\times(1/T)$ for the determination of the activation energy of the grain growth process of the Ni-42Ti alloy.

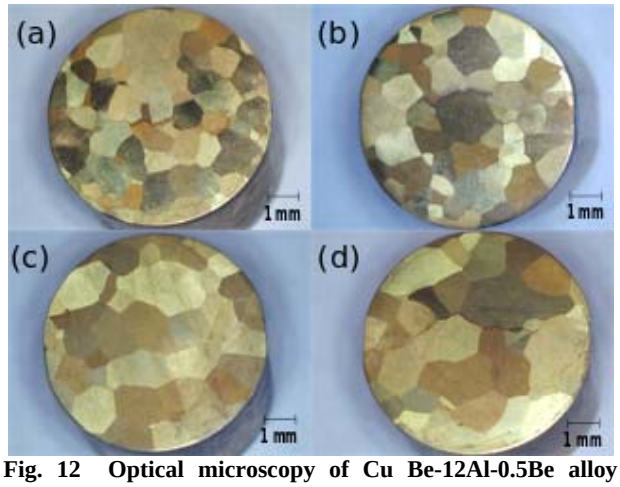

Fig. 12 Optical microscopy of Cu Be-12Al-0.5Be alloy subjected to aging treatment at 823 K, for 2, 12, 24 and 120 min from (a) to (d).

The results show Cu-14Al-4Ni alloy is extremely sensitive to variation of temperatures, having dual grain growth kinetics. Through the diagram $\ln(D-D_0)\times(1/T)$, the empirical activation energy process was estimated by two growth areas (Fig.7), as at the Ni-42Ti alloy (Fig. 11). The Cu-12Al-0.5Be alloy has a single growth area, shown in Fig. 9. Fig. 12 shows the increased grain of the Cu-12Al-0.5Be alloy.

3. Conclusions

The recrystallization kinetics followed by the change of hardness despite the limitations of this process, proved to be an important tool, working as a guideline for selection of treatment time as a function of temperature.

The equations describing the recrystallization kinetics follow, with reasonable approximation, especially for Cu-14Al-4Ni and Ni-42Ti alloys, the empirical J-M-A's law.

Considering the grain growth process, the alloy less susceptible to aging at temperatures lower than the recrystallization peak is the Ni-42Ti, because it had the highest activation energy, followed by the Cu-14Al-4Ni.

Due to the difference in color found between the grains (Fig. 12) and the fact that it does not have double recrystallization kinetics (common in Cu-based SMAs), the results suggest that the Cu-12Al-0.5Be alloy is heterogeneous.

The dual grain growth kinetics presented by this alloys, similar to what occurs with other alloy systems, may be, in the first domain, of lower temperature, associated to primary recrystallization (involving a higher activation energy), and, at the second domain (at higher temperatures), to secondary recrystallization, produced by intense atomic diffusion through contour (involving a lower activation energy).

Acknowledgements

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