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Dynamics of domain walls in Ti–Ni–Cu alloy

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Abstract. The internal friction for Ti–Ni–Cu shape memory alloys (SMAs) was measured in the Hz and kHz range. The peak temperature in the Hz range is independent of the measuring frequencies. Only in the tens of kHz range does the peak temperature shift with the frequencies, showing a thermally activated progress with $\tau = \tau_0 e^{-B/T_c - T}$, characteristic of viscous motion of domain walls, instead of the Arrhenius relation. Taking into account the change of the density of the domain walls during the phase transformation, we modified the Q^{-1}-formula by using a model of viscous motion of domain walls, obtaining a good result in agreement with the experimental data. Additionally, corresponding parameters, which play a key role in the shape memory effect, such as the viscosity coefficient and the effective pinning force constant, were obtained.

1. Introduction

The internal friction (Q^{-1}) associated with the first-order phase transition in the low-frequency range is known to be of static hysteresis type and independent of the measuring frequencies [1–4]. Huang *et al* [5] found a frequency-dependent peak for KDP and TGS crystals in the higher-frequency range, and it has been determined that the peak is associated with viscous motion of domain walls. As we know that shape memory alloys undergo first-order martensitic transformation, during the phase transformation the phase interfaces and domain walls will merge. The mobility of these domain walls is important in the shape memory effect because it affects the fatigue time and response speed of the shape memory effect. So Shen *et al* [6] studied the dynamics of Cu–Al–Zn–Ni shape memory alloys and also found that the internal friction peak temperature shifts with frequency only in the tens of kHz range, showing a thermally activated progress with $\tau = \tau_0 e^{-B/T_c - T}$, characteristic of viscous motion of domain walls, instead of the Arrhenius relation. By computer simulation, they obtained corresponding parameters such as the viscosity coefficient and effective pinning force constant, which play a key role in the shape memory effect.

In this paper, we also found a peak shift for Q^{-1} only for frequencies in the tens of kHz range for Ti–Ni–Cu shape memory alloy. Taking into account the change of the density of the domain walls during the phase transformation, we modified the Q^{-1}-formula on the basis of the model of viscous motion of domain walls and obtained a fairly good simulation result in agreement with experimental data.

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2. Experiments

A shape memory alloy with the composition $Ti_{50}Ni_{25}Cu_{25}$ was prepared by vacuum induction melting and then homogenized at 900 °C for 24 hours. Samples with sizes of $3 \times 3 \times 33$ mm^3 and $1 \times 1.5 \times 40$ mm^3 were prepared for the Q^{-1}-measurement by spark cutting and then aged at 550 °C for 36 hours. The internal friction was measured by a torsion pendulum apparatus in the Hz range and a piezoelectric composite oscillator in the 50–150 kHz range. X-ray diffraction (XRD) and transmission electron microscopy (TEM) were applied to study the structure, the average density of the interfaces and the spontaneous strain in the samples.

3. Results and discussion

Shi [7] has carried out extensive research into the internal friction of Ti–Ni–Cu SMAs in the Hz range and has revealed that the peak temperature is independent of the measuring frequencies. Figure 1 shows a typical result which was measured on heating at $\mathrm{d}T/\mathrm{d}t = 1$ K min^{-1} and $f = 0.46$ Hz. The transformation temperatures were obtained as $A_s = 44$ °C and $A_f = 73$ °C. We also observed a frequency-dependent internal friction peak below A_f in the 50–150 kHz range on heating (as shown in figure 2), just like that in reference [6]. Wang and co-workers [5, 6, 8] pointed out that this behaviour of the internal friction is associated with the viscous motion of domain walls, and they deduced the Q^{-1}-formula to be

$$Q^{-1} = \frac{2N[\varepsilon_{ij}^{(s)}]^2}{Jk} \frac{\omega\tau}{1 + \omega^2\tau^2} \tag{1}$$

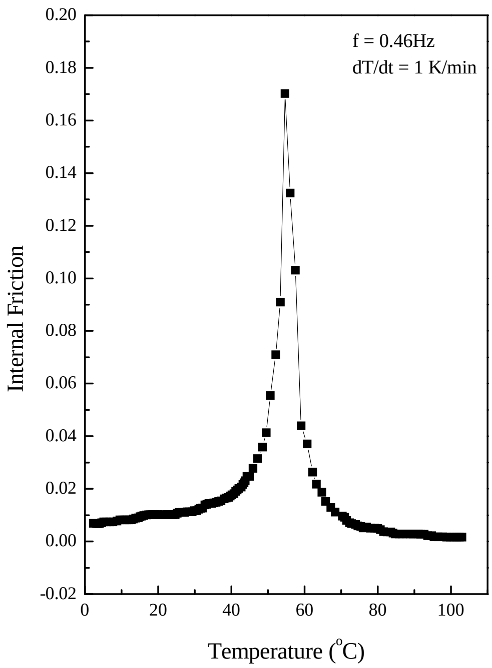

Figure 1. Q^{-1} versus T at 0.46 Hz and $\mathrm{d}T/\mathrm{d}t = 1$ K min^{-1} for Ti–Ni–Cu alloy. $A_s = 44$ °C; $A_f = 73$ °C.

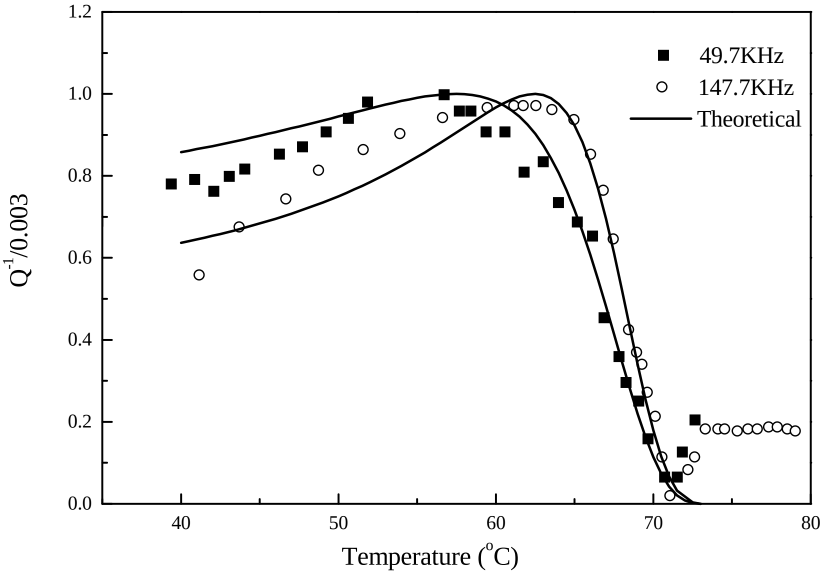

Figure 2. Q^{-1} versus T for Ti–Ni–Cu alloy. $A/k = 1 \times 10^{-5}$ s and $B = 20$ K, from simulation.

where N is the average number of parallel interfaces per unit perpendicular distance, $\varepsilon_{ij}^{(s)}$ is the spontaneous shear strain, J is the compliance, k is the effective force constant, $\tau = \Gamma/k$ is the relaxation time and Γ is the viscosity coefficient.

The expression for Γ obtained by Comb and Yip [9] with constants A and B is

$$\Gamma = A\mathrm{e}^{-B/T_c-T}. \tag{2}$$

For a second-order, or a weakly first-order, phase transition [10],

$$N = N_0/(T_c - T) \qquad [\varepsilon_{ij}^{(s)}]^2 \propto (T_c - T)$$

so $N[\varepsilon_{ij}^{(s)}]^2$ is nearly independent of T. But this does not hold in a martensitic transformation because it is so strongly a first-order phase transition. It is necessary to find a new way to establish the relation between N and T. The best way is by experimental observation. Also San Juan and co-workers [11] pointed out that, during a thermoelastic martensitic transformation, the volume fraction of transformed martensite $n(T)$ can be expressed as

$$n(T) = \int_{T_s}^{T} \mathrm{IF}_{\mathrm{Tr}}\omega^l \, \mathrm{d}T \bigg/ \int_{T_s}^{T_f} \mathrm{IF}_{\mathrm{Tr}}\omega^l \, \mathrm{d}T \tag{3}$$

where $\mathrm{IF}_{\mathrm{Tr}}$ is the transient contribution of the internal friction. The temperature range (T_s, T_f) is assumed to be large enough to reach the background regions on both sides of the peak. Thus we can use figure 1 to calculate $n(T)$, because this internal friction peak is mainly a transient one [11] and it is reasonable to suppose that N is proportional to $n(T)$. Figure 3 shows $n(T)$ calculated with $l = 1$ [12–14]; $n(T)$ decreases from 1 to zero as the temperature changes from A_s to A_f. So we find the function for N, by simulation, to be $N \propto \cos[0.082(T - T_c)]$.

However, the characteristic of the first-order phase transition, i.e. the phase transition occurring between A_s and A_f, should be taken into account. So we considered that T_c in Γ has a uniform distribution between A_s and A_f with

$$\int f(T_c) \, \mathrm{d}T_c = 1$$

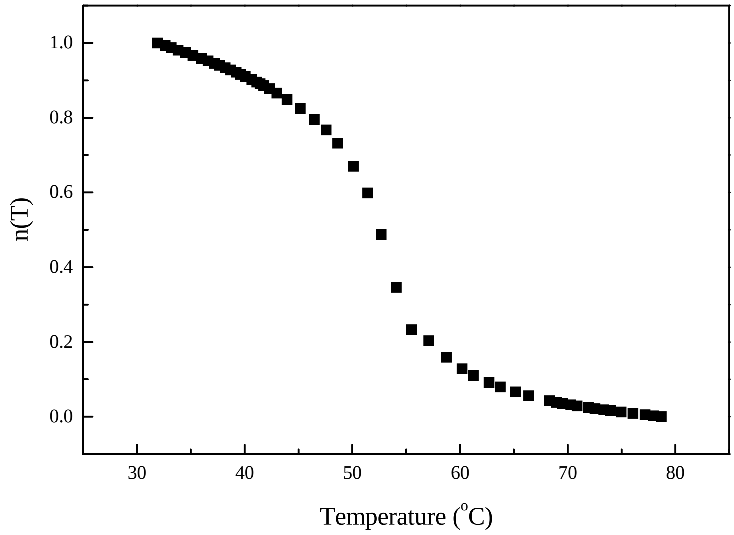

Figure 3. The calculated volume fraction of transformed martensite $n(T)$ versus T for Ti–Ni–Cu alloy.

and the Q^{-1}-formula was modified to the form

$$Q^{-1} = \int \frac{2N(T, T_c)[\varepsilon_{ij}^{(s)}]^2}{Jk} \frac{\omega\tau}{1 + \omega^2\tau^2} f(T_c)\, \mathrm{d}T_c. \tag{4}$$

For a first-order phase transition, $\varepsilon_{ij}^{(s)}$ changes slowly except for one finite abrupt change, so we consider it as a constant here. Now, Q^{-1} can be simulated with the new formula. The simulation result is also plotted in figure 2, and it shows a fairly good fit to the experimental data. We obtained $A/k = 1 \times 10^{-5}$ s and $B = 20$ K. In order to get the effective force constant k, we calculated the spontaneous shear strain, density of interfaces N and modulus J^{-1} on the basis of the results from x-ray diffraction, TEM observation and internal friction measurements.

The results of the x-ray diffraction measurements for the Ti–Ni–Cu alloy show that $a = 0.2908$ nm, $b = 0.4322$ nm and $c = 0.4450$ nm in the orthorhombic B_{19} martensitic phase whose principal axes are parallel to the [100], [011] and $[0\bar{1}1]$ directions of the B_2 cubic parent phase with $a_0 = 0.3039$ nm. In the principal-axes coordinate system of the B_{19} phase, we can obtain the strain due to the martensitic transformation on the basis of the XRD results and the Neumann principle:

$$\varepsilon_{11} = (a - a_0)/a_0 = -0.0431 \qquad \varepsilon_{22} = (b - \sqrt{2}a_0)/\sqrt{2}a_0 = 0.0056$$
$$\varepsilon_{33} = (c - \sqrt{2}a_0)/\sqrt{2}a_0 = 0.0354 \qquad \varepsilon_{ij}(i \neq j) = 0.$$

So, the spontaneous shear strain in the B_2 cubic coordinate system is

$$\varepsilon_{ij}^{(s)} = 2\sum_{k,l=1}^{3} a_{2k}a_{3l}\varepsilon_{kl} = -0.0298 \tag{5}$$

where $a_{11} = 1$, $a_{22} = a_{33} = a_{32} = \sqrt{2}/2$, $a_{23} = -\sqrt{2}/2$, $a_{21} = a_{12} = a_{31} = a_{13} = 0$. The average density of interfaces N is obtained as 6.8×10^6 m^{-1} on the basis of TEM observation. The modulus J^{-1} is 6.14×10^4 MPa according to the formula $J^{-1} = 4\rho l^2 f^2$, where

$\rho = 5.66 \times 10^3\,\mathrm{kg\,m^{-3}}$, $l = 3.3\,\mathrm{cm}$ and $f = 49.7\,\mathrm{kHz}$. The effective pinning force constant k and the viscosity coefficient Γ can be obtained from the results mentioned above and formulae (2) and (4). They are $k = 1.24 \times 10^{16}\,\mathrm{N\,m^{-3}}$, $\Gamma = 1.24 \times 10^{11} \exp[-20/(346 - T)]\,\mathrm{kg\,m^{-2}\,s^{-1}}$.

4. Conclusions

An internal friction peak associated with viscous motion of domain walls was measured in the 50–150 kHz range for Ti–Ni–Cu shape memory alloy. The Q^{-1}-formula was modified and the viscosity coefficient and the effective pinning force constant were obtained by computer simulation using the modified formula.

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