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INTERNAL FRICTION IN TERNARY α-CuNiZn ALLOYS

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Abstract. - Internal friction measurements have been carried out for a number of face-centred cubic CuNiZn alloys, especially the alloys around the composition Cu$_2$NiZn. Damping measurements are carried out using an inverted Ké-type pendulum equipped with an optical detector. With an extrapolation in the ageing time relaxation data for the disordered structure were obtained. Also shear moduli and Young's moduli are reported on the same alloys.

Introduction. - The anelastic damping in disordered Cu$_2$NiZn below the critical temperature for ordering is measured. In earlier papers measurements in ordered material (Van Der Veen et al. 1979) as well as on a Cu$_2$NiZn single crystal were reported (De Rooy et al. 1980). For a detailed description of the structure types in this alloy reference should be made to Vrijen et al. 1976 and De Rooy et al. 1980. The main features will be outlined below.

In this alloy the order-disorder transformation takes place in two steps. The first step from a fully ordered A$_2$BC (L1$_0$) structure to a ternary ordered A$_3$B (L1$_2$) structure has a critical temperature of about 625 K. After this transformation the Cu and Ni atoms are disordered, while the Zn atoms are still on their own sublattice. The second step, from the partially ordered A$_3$B structure to the completely disordered structure has a critical temperature of about 770 K.

There is a little difference in atomic size of Cu and Ni. In comparison with the atomic radius of Zn, the atomic size difference of Cu and Ni can be neglected (the experimental c/a for A$_2$BC is < 1.0002). This gives rise to the expectation that there will be hardly any effect on the internal friction when the critical temperature for A$_2$BC-A$_3$B is passed by. Due to the difference in size of Zn compared with the other two elements it may be expected that the second transformation will be more drastic.

Since the internal friction measurements are related to the elastic moduli, also the shear modulus and the Young's modulus are measured.

In the following section the theory is summarized, especially the formulae needed to explain the results. In section two the sample preparation and experimental technique are described. In the third section the experimental results are reported, followed by a discussion in the last section.

1. Theory. - When suddenly a stress is applied to an anelastic solid, this solid will show creep. In a torsional pendulum this creep behaviour is demonstrated when we follow the creep angle $\gamma$ as a function of time, that obeys approximately the exponential equation:

$$\gamma = \gamma_0 (1 - e^{-1/\tau})$$  \hspace{1cm} (1)

When the applied stress is removed, the anelastic strain relaxes and so the angle changes according to the equation:
This is called the elastic after-effect. In equations 1 and 2 $\gamma_0$ stands for the angle at $t=0$, $\tau$ is the relaxation time for the process at the temperature $T$. The relaxation time obeys an Arrhenius type equation:

$$\tau = \tau_0 e^{Q/kT},$$

where $\tau_0$ is the frequency factor and $Q$ is an activation energy. In Cu$_2$NiZn two processes are involved, each with its own relaxation time. The total creep angle is than a superposition of the two angles:

$$\gamma_{tot} = \gamma^1 + \gamma^2.$$

In a dynamic experiment an alternating stress of the following form is applied to the solid:

$$\sigma = \sigma_0 \sin \omega t,$$

where $\sigma_0$ stands for the amplitude of the applied stress, $\omega$ stands for the circular frequency and $t$ is the time. If the phase lag of the strain in relation to the stress is $q$, than the strain is expressed as:

$$\epsilon = \epsilon_0 \sin (\omega t + q).$$

For very low frequencies the deformation can follow the applied stress almost instantaneously. For very high frequencies the after effect does not play any significant role. In both cases the phase lag will be very small. Between these limits there is a frequency range where the phase lag is more pronounced. When the relaxation behaviour is the same for creep and after effect, the phase lag between stress and strain is given by:

$$\tan q = \frac{\Delta}{\sqrt{1 + \Delta}} \cdot \frac{\omega \tau'}{1 + (\omega \tau')^2}$$

with $\Delta$: relaxation strength: $(g_u - g_r)/g_r$

$g_u$: elastic modulus at high frequencies

$g_r$: elastic modulus at low frequencies

$\tau' = \tau / \sqrt{1 + \Delta}$.

Equation (7) is a Debye type equation, that has a maximum at $\omega \tau$ is equal to 1. Then:

$$\tan q_{\text{max}} = \frac{\Delta}{2 \sqrt{1 + \Delta}}.$$

For small amplitudes the logarithmic decrement $\delta$ defined as $\delta = \log A_n - \log A_{n-1}$, with $A$ as amplitude, can be expressed in the phase lag by:

$$\delta = \pi \tan q.$$

We then arrive at the expression of the logarithmic decrement as function of frequency:

$$\delta = 2 \delta_{\text{max}} \frac{\omega \tau}{1 + (\omega \tau)^2}.$$

Rather than to investigate the damping as function of the frequency $\omega$ at fixed temperatures (i.e. $\tau$), it is more convenient to vary the temperature and keep the frequency constant. Another way of writing equation (10) is with the help of the hyperbolic secants function, namely:
\[
\delta = \delta_{\text{max}} \text{sech} \left( \ln \omega T \right).
\] 

According to equation 3 we can write for the term \(\ln \omega T\):

\[
\ln \omega T = \ln \omega \tau_0 + \frac{Q}{kT}.
\] 

At the peak temperature the relation \(\omega \tau_p = 1\) holds, i.e. \(\ln \omega \tau_p\) is equal to 0. Then it is possible to write the term \(\ln \omega \tau_0\) as a function of the peak temperature, according to:

\[
\ln \omega \tau_0 = -\frac{Q}{kT}.
\] 

Combining the equations (11)(12) and (13), we may write:

\[
\delta = \delta_{\text{max}} \text{sech} \left\{ \frac{Q}{k} \left( \frac{1}{T} - \frac{1}{T^*} \right) \right\}.
\] 

The measured experimental curve is again a summation of two processes:

\[
\delta_{\text{tot}} = \delta' + \delta^2.
\] 

The shear modulus is calculated from the time of oscillation \(T\) of the torsional pendulum:

\[
G = \frac{8\pi I l}{T^2 R^4},
\] 

where
- \(I\): moment of inertia of the pendulum
- \(l\): length of the specimen
- \(R\): radius of the specimen

2. Sample preparation. - The starting material is Cu(5N), Ni(4N), Zn(5N). The three elements are melted together in the appropriate amounts in a high vacuum furnace. After solidification the alloy is sealed in a quartz tube and homogenized for a week, 50 K below the melting point. The composition is checked using atom absorption spectroscopy. The Cu\(n\)Ni\(n\)Zn specimens are then rolled and drawn to wires of the chosen diameters. To obtain samples with large grains, the wires are heat treated for four days at 1225 K. Disordered specimens are obtained by quenching from this temperature into water. Ordered samples are obtained by an additional heat treatment of four days at 625 K and slowly cooled afterwards.

2.1 Experimental method.

2.1.1 Low frequency internal friction. - The low frequency internal friction measurements are performed with an inverted K\(\ddot{e}\) torsional pendulum. This pendulum is equipped with an optical detection system, essentially a laser interferometer. The specimen, which is placed in a vacuum chamber, is surrounded by a tube-type furnace. This vacuum chamber can reach an ultimate vacuum of \(10^{-2}\) Pa. The frequency is controlled by the dimensions of the sample and by shifting the weights on the inertia arm of the pendulum. In this way the frequency range covered is 0.5-35 Hz. The measurements are made in free decay. After a prefixed number of cycles the pendulum is started again automatically by two solenoids placed nearby the inertia arm. The normally used prefixed number of cycles is eight. The temperature is controlled by a proportional control unit connected with a linear heating device. The heating rate is 2 K/min. The temperature is determined by a chromel-alumel thermocouple placed as close as possible to the centre of the specimen. This equipment allows us to follow the damping almost continuously.

2.1.2 Elastic after-effect. - For relaxation times between 20 and 7000 s elastic after effect measurements are used. The elastic after effect measurements are performed with the same pendulum and satellite equipment as for the low frequency internal friction measurements. The modification is brought in by a different arrangement of the solenoids to the inertia arm and by an external current supply. It is now pos-
sible to have DC current through the solenoids, so the specimen can be twisted over a certain angle. When the specimen has been held in the twisted position for a certain time, the current is switched off instantaneously and the strain relaxation is recorded. These measurements are performed at different temperatures, which are controlled within 0.1 K.

2.1.3 The shear and Young's modulus. - The shear modulus can be obtained in two ways using the inverted Ké pendulum. Firstly from the low frequency internal friction measurements, where the shear modulus is connected with the frequency according to equation (16) and secondly from the after effect measurements, where the shear modulus is related to the twist angle. Both methods are used to determine the shear modulus. To find the shear modulus as a function of temperature, it is only necessary to follow the frequency as a function of temperature in the internal friction apparatus. With the after effect set-up discrete temperature steps must be taken.

The Young's modulus is measured with a stress-strain apparatus. A stress is introduced by lifting an extra weight to the wire specimen. Between the wire specimen and the extra weight a magnetic displacement device is mounted. The wire itself is placed in an inert gas atmosphere of He+3%H₂ to prevent oxidation. At each temperature about ten stress-strain measurements are performed. Each temperature step is 25 K.

3. Experimental results. - The actual damping peak is obtained by subtracting from the experimental curve a low and high temperature background. The low temperature background due to damping in the apparatus, while the high temperature background is due to grain boundary damping. The difference turns out to be asymmetric, indicating two damping processes to be present. A fitting procedure to the composed curve using the hyperbolic secants functions yields values for the peak temperatures. These temperatures together with the relaxation times, calculated from the natural frequency at the peak temperature are plotted in a ln τ vs. 1/T plot.

In order to obtain the elastic after effect in disordered material below the critical temperature for ordering, the relaxation time as a function of the ageing time is measured. When we now extrapolate back to zero time, the relaxation time for the disordered material is known (Goering et al.). As mentioned in section 1 the relaxation curve can be described by two relaxation processes, each with its own relaxation time. It is possible that in the disordered case the fastest relaxation time is so small, that the curve can be described only by a single process. The relaxation time is determined using equation (4) with the least square fitting. The data points corresponding to these relaxation times and temperatures are plotted in the same ln τ vs. 1/T plot as the low frequency internal friction results.

![Fig. 1 Alloy 50/25/25 relaxation time vs. 1/T.](image1)

![Fig. 2 Alloy 50/20/30 relaxation time vs. 1/T.](image2)
The results are compiled in figure 1 (Cu\textsubscript{50}Ni\textsubscript{25}Zn\textsubscript{25}) and in figure 2 (Cu\textsubscript{50}Ni\textsubscript{20}Zn\textsubscript{30}). In both figures two processes are clearly visible. The process with the faster relaxation time at a certain temperature is called the fast process and vice versa. The results of the shear and Young's modulus measurements are depicted in figure 3a and 3b.

Activation energies and pre-exponential factors can be obtained from figure 1 and 2. The activation energy corresponds to the first derivative of the relaxation time towards the inverse temperature, the pre-exponential factor to the intersection of the damping curve with the T-axis at T = \infty (equation (3)).

The calculated values and critical temperatures estimated from these plots are:

<table>
<thead>
<tr>
<th>Internal friction data of both alloys</th>
</tr>
</thead>
<tbody>
<tr>
<td>order</td>
</tr>
<tr>
<td>slow Q(eV)</td>
</tr>
<tr>
<td>(\tau_0) (s(^{-1}))</td>
</tr>
<tr>
<td>fast Q(eV)</td>
</tr>
<tr>
<td>(\tau_0) (s(^{-1}))</td>
</tr>
<tr>
<td>(T_{\text{crit}}) (K)</td>
</tr>
</tbody>
</table>

Fig. 3a
Shear modulus vs. T.

Fig. 3b
Young's modulus vs. T.

The shear modulus and the Young's modulus for both alloys illustrate the expected result of a decreasing modulus with increasing temperature. At the critical temperature for order–disorder the decrease is somewhat steeper. This reflects the normal behaviour at the order–disorder transformation.

4. Discussion and conclusions. - The mechanism of the slow and the fast process is diffusion controlled. From the results of Anusavice et al. 1972 who measured the self diffusion of Cu, Ni and Zn on ordered ternary Cu-Ni-Zn alloys, it is possible to ascribe the slow and the fast process to the mean time of stay of the Ni atoms, and the Zn atoms, respectively. In the disordered structures the same two processes are responsible for the difference in relaxation times. A slight enhancement of the relaxation time in the ordered structure compared to the disordered structure might be explained on the basis of tighter binding and smaller lattice parameter. The sharp decrease in relaxation time around the critical temperature can be explained by the increase of the atomic mobility when disorder starts to develop. The atomic mobility is mainly ascribed to the vacancy correlation factor, which is in the ordered state lower than in the disordered state.

As stated in the introduction only the A\textsubscript{3}B–disorder phase transformation is detectable by the internal friction method. Apart from the difference in atomic sizes
this is accomplished by the ordering energies for the three structures. During the phase transformation of $A_2BC$ to $A_3B$ the number of CuZn and NiZn bindings remains the same, while the number of CuNi bindings is decreased in favour of the CuCu and NiNi bindings. The pairpotential of CuNi is about one tenth of that of CuZn and of NiZn, so the difference in ordering energy between $A_2BC$ and $A_3B$ is very small. During the transformation of $A_3B$ to disorder the largest effect is almost entirely due to the decrease of the number of CuZn and NiZn bindings. From this point of view the decrease in ordering energy between $A_3B$ and disorder is about ten times as large as the decrease between $A_2BC$ and $A_3B$. The magnitude of the internal friction and the elastic moduli is for a great deal dependent on the size effect and the ordering energies. Indeed the $A_2BC$-$A_3B$ phase transformation is completely hidden within the experimental error.

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