JOURNAL OF MATERIALS SCIENCE 20 (1985) 654–660

# Phenomenological study of elastic and anelastic properties of dental alloys

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Measurements of Young's modulus, mechanical damping, stress and unit damping energy have been made as a function of temperature on five dental amalgams with the PUCOT (piezoelectric ultrasonic composite oscillator technique). In Dispersalloy amalgam the mechanical damping changed by a factor of 4.3 and Young's modulus was reduced by 0.5% as the temperature increased from 20 to 80° C. The effective activation energy for the change in damping was 0.42 eV. For the five amalgams the curves of unit damping energy against stress, damping against modulus, and (damping/modulus) against modulus had a variety of forms: one, two, three or four straight lines, with or without hysteresis effects. All the curves for Tytin amalgam showed a sharp change near 68° C. The data are examined phenomenologically to show that they lend themselves well to the detection and monitoring of transformations taking place in dental alloys.

## 1. Introduction

The mechanical property that is most often taken into consideration in the design and application of materials is elasticity. This consideration applies particularly in the case of restorations made from dental alloys where in-service plastic deformation is highly undesirable. Elasticity is defined by the conditions that:

1. the strain response for each level of stress (or vice versa) has a unique equilibrium value;
2. the equilibrium response is achieved instantaneously;
3. the response is linear.

Anelasticity can be defined by modifying condition 2 from instantaneous equilibrium response to a time dependent one. This paper focuses on a particular elastic property – Young's modulus – and a particular anelastic property – mechanical damping – as functions of stress, temperature and time.

Recently one of the authors [1] made measurements with the PUCOT (piezoelectric ultrasonic composite oscillator technique) of Young's modulus, $E$, and mechanical damping, $Q^{-1}$, on eight silver dental alloys. During the investigation the time dependence for ageing at 37° C and the temperature dependence for the range 20 to 80° C were studied. With increasing ageing time the $E$ values increased from around 17 GPa at 15 min and saturated at about 70 GPa in times of $10^3$ to $10^4$ min. Over the temperature range 20 to 80° C the $Q^{-1}$ values increased rapidly by factors of 6 to 32 while the $E$ values decreased less than 5%. It was suggested that these changes in mechanical properties were related to the $\gamma_1 \rightarrow \beta_1$ phase transformation in the alloys. Activation energies of 0.34 to 3.1 eV were obtained from Arrhenius plots and some of these values could be related to the reported activation energy values for the diffusion of various elements in the amalgams. The observed time dependence of $Q^{-1}$ followed a relation of the type $Q^{-1} \propto t^n$ where the exponent $n$ was interpreted in terms of various diffusion mechanisms.

The present paper deals with the results obtained from an extension of the research reported earlier [1]. For one amalgam (Dispersalloy) the temperature dependencies of $E$ and $Q^{-1}$ and the time dependence of $E$ are reported and discussed. For five amalgams (Shofu, Dispersalloy, NTD (New True Dentalloy), Tytin and G&C) the following measurements are examined phenomenologically and discussed: $E$, $Q^{-1}$, stress $\sigma$, temperature $T$ and unit damping energy UDE $(= \pi Q^{-1} \sigma^2/E)$. It is


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TABLE I Dental amalgams used in the investigation

| Alloy | Manufacturer |
|---|---|
| New True Dentalloy (NTD) | S. S. White, Philadelphia, PA 19102 |
| G&C (Luna Atomic non-zinc) | G-C Chemical Manufacturing Co, Ltd, Tokyo |
| Shofu (Spherical) | Shofu Dental Manufacturing Co, Ltd, Kyoto |
| Dispersalloy | Johnson & Johnson, East Windsor, NJ 08520 |
| Tytin | S. S. White, Philadelphia, PA 19102 |

shown that for the Dispersalloy amalgam $Q^{-1}$ changes by a factor of 4.3 and $E$ is reduced only slightly (0.5%) as the temperature goes from 20 to 80° C; the effective activation energy for the change in $Q^{-1}$ is 0.42 eV. For the five amalgams this investigation shows that: the UDE–$T$ curves were in the form of one, two or four straight lines with hysteresis for one alloy; the UDE–$\sigma$ curves were two, three or four straight lines, some with hysteresis; the $Q^{-1}$–$E$ and $Q^{-1}/E$–$E$ curves were possibly one straight line or curves with hysteresis; all the curves for one amalgam (Tytin) showed a sharp change (almost a step function) near 68° C. It is shown that the presentation of the data in certain forms lends itself well to the detection and monitoring of transformations occurring in dental alloys.

## 2. Experimental procedure

Table I gives a list of the dental amalgam alloys used in this investigation. Three are low copper (less than 5%) Ag–Sn–Hg alloys and two (Dispersalloy and Tytin) contain between 12 and 15% of copper. The details of the trituration (preparation of the amalgam) and of the PUCOT have been reported elsewhere [1–4]. One of the amalgams (Dispersalloy) was in two varieties – "normal" (or "regular") and "fast set". Briefly, the PUCOT consisted of piezoelectric quartz drive D and gauge G crystals to excite longitudinal ultrasonic (80 kHz) resonant stress waves in a quartz spacer rod Q and in the cylindrical amalgam specimen S of appropriate resonant length. From the measured values of drive and gauge voltage, the resonant periods of D–G–Q and of D–G–Q–S, the masses of the various components, and the specimen length standard calculations [1, 3] gave values of $E$, $Q^{-1}$, $\sigma$ and UDE. Changes in stress during the experiments were effected by changes of temperature and hence of $E$.

## 3. Results

The completion of the studies on Dispersalloy has yielded the new results of Young's modulus, $E$, as a function of ageing time at room temperature for fast set and for regular amalgam (Fig. 1), mechanical damping $Q^{-1}$ and $E$ as a function of temperature (Fig. 2) and an Arrhenius plot of $Q^{-1}$ (Fig. 3). In Figs. 2 to 20 the open data points refer to heating and the filled data points to cooling. For the five amalgams the UDE–$T$ curves are shown in Figs. 4 to 8; the UDE–$\sigma$ curves in Figs. 9 to 13; the $Q^{-1}$–$E$

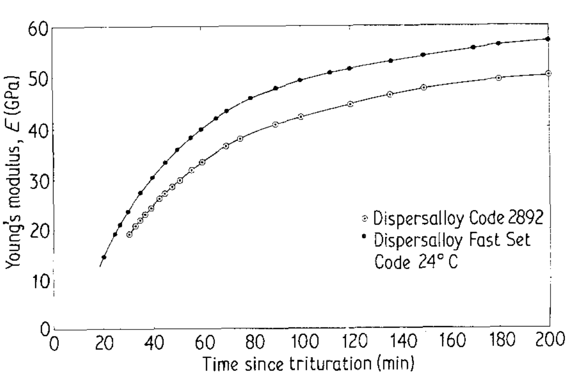


*Figure 1* Young's modulus, $E$, as a function of ageing time for two types of Dispersalloy amalgams.

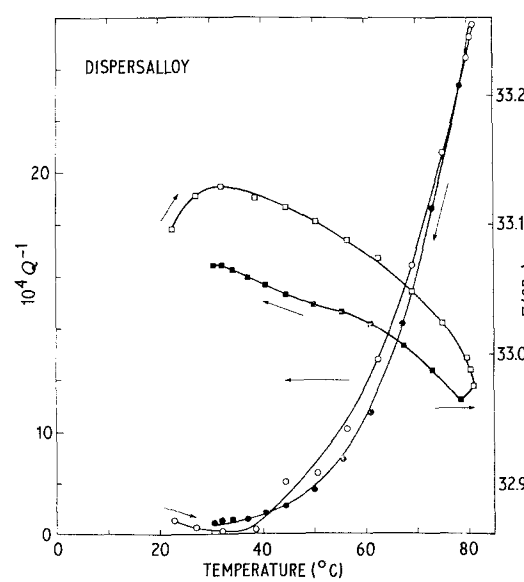


*Figure 2* The temperature dependence of mechanical damping (circles) and Young's modulus (squares) for Dispersalloy.

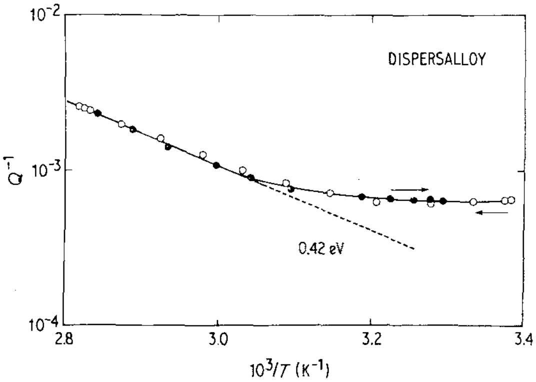


*Figure 3* Arrhenius plot of mechanical damping for Dispersalloy.

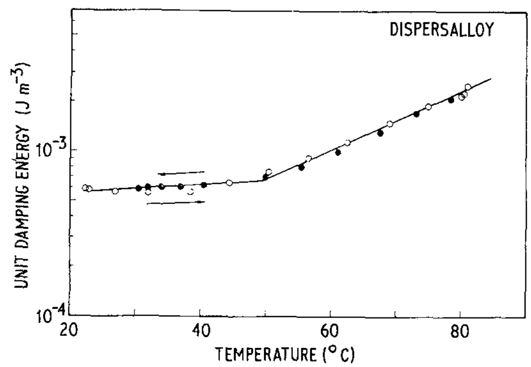


*Figure 5* Temperature dependence of unit damping energy for Dispersalloy.

curves in Figs. 14 to 18; and $Q^{-1}/E$–$E$ curves in Figs. 19 and 20.

## 4. Discussion

One of the advantages of the PUCOT is that the kinetics of the hardening process in amalgams can be monitored easily. Fig. 1 shows that the hardening rate of fast set Dispersalloy exceeds that of the regular set amalgam: the former attains a modulus value of 50 GPa in 100 min after trituration, while the latter requires 200 min to achieve the same value of Young's modulus. These figures compare well with some of those obtained previously [1] for other amalgams.

For Dispersalloy the temperature dependences of mechanical damping and Young's modulus (Fig. 2) indicate that $Q^{-1}$ changes by a factor of 4.3 and $E$ decreases by only 0.5% for a temperature, $T$, change from 20 to 80° C. These changes are smaller than those reported for other amalgams previously [1]. There is a rapid increase in $Q^{-1}$ near 70° C which is probably connected with the $\gamma_1 \rightarrow \beta_1$ phase transformation [5]. For heating and cooling the $Q^{-1}$–$T$ curves are essentially the same. This was not the case for the $E$–$T$ curves which showed that the cooling curve is generally about 0.1% lower than the heating curve. The significance of the slight peak in the $E$–$T$ curve near 32° C is not clear – it has been noticed before only for G&C [1].

The effective activation energy for the change of $Q^{-1}$ with temperature (Fig. 3) is 0.42 eV. This is equal in value to that obtained by Timmons *et al.* [6] for the diffusion of mercury in $\gamma_1(Ag_2Hg_3)$. Thus the diffusion of mercury in the $\gamma_1$ phase appears to be influencing the damping properties of the dental amalgam. The tail on the curve for $10^3/T > 3.2$ may correspond to the thermal inertia of the specimen–furnace system and should be neglected.

The temperature dependence of the UDE shows varying behaviour for the various amalgams. For Shofu the curve for heating and cooling can be

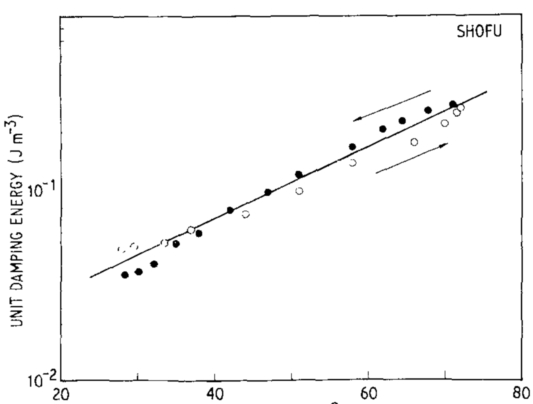


*Figure 4* Temperature dependence of unit damping energy for Shofu.

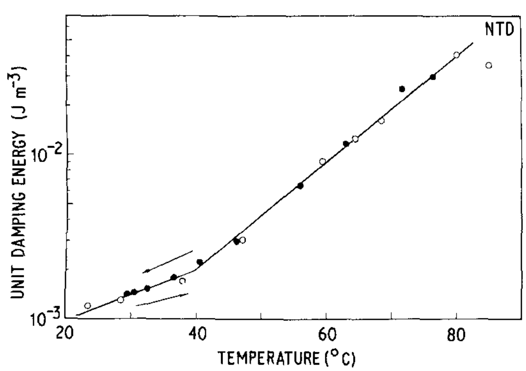


*Figure 6* Temperature dependence of unit damping energy for NTD.

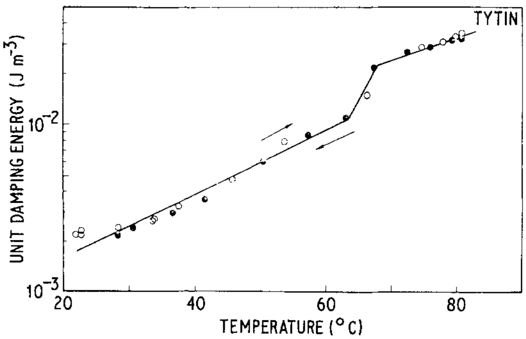


*Figure 7* Temperature dependence of unit damping energy for Tytin.

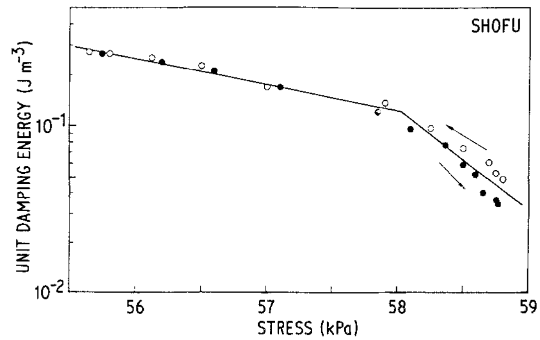


*Figure 9* Stress dependence of unit damping energy for Shofu.

fitted to a single straight line (Fig. 4). However, for Dispersalloy, NTD and Tytin the curves for heating and cooling are fitted to two straight lines, with a short step function for Tytin (Figs. 5 to 7). The curves for G&C are more complicated: here we can fit four straight lines, with a hysteresis loop for heating and cooling (Fig. 8). While the changes in slopes near 68° C (Tytin) and 72° C (G&C) are most likely related to the phase transformation in the alloys, the changes of slopes near 50° C (Dispersalloy) and near 40° C (NTD) are not so easy to interpret. In the case of Dispersalloy the temperature of 50° C is near the point in Fig. 3 ($10^3/T = 3.1\ \mathrm{K}^{-1}$) where the Arrhenius plot is changing slope rapidly. Such a slope change may indicate a change in effective activation energy from a low value near zero to 0.42 eV. This undoubtedly relates to a change in damping mechanism: the value of 0.42 eV has been interpreted above; the other (low) value requires further elucidation.

The values of UDE for NTD, Tytin and G&C shown in Figs. 6 to 8 lie near $10^{-2}\ \mathrm{J\,m^{-3}}$, while those for Dispersalloy (Fig. 5) are lower (near $10^{-3}\ \mathrm{J\,m^{-3}}$) and those for Shofu (Fig. 4) are high (about $10^{-1}\ \mathrm{J\,m^{-3}}$). In fact, the UDE values for Shofu rival those of turbine blade alloys such as 410 stainless steel, 15 Cr–Mo steel, 26 Cr–1 Mo steel and Ti–6 Al–4 V [7].

The stress dependence of unit damping energy for the amalgams has been obtained via the influence of temperature on Young's modulus during the tests. In effect, therefore, stress is not strictly an independent variable and varies only a few percent. The UDE–$\sigma$ curves presented in Figs. 9 to 13 show various types of behaviour. For Shofu (Fig. 9) the curves are two straight lines; for Dispersalloy (Fig. 10) there is hysteresis with a straight line for a decrease in stress (heating) and a different straight line for increasing stress (cooling); NTD has four straight lines (Fig. 11) with hysteresis; for Tytin (Fig. 12) the curves are two straight lines with a jump or step function; and the results for G&C can be fitted to three straight lines with hysteresis (Fig. 13). In experiments at constant temperature UDE normally increases with stress [7] and Figs. 9 to 13 thus seem at first glance to be inconsistent

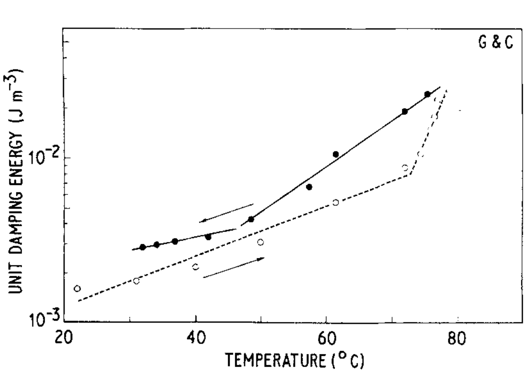


*Figure 8* Temperature dependence of unit damping energy for G&C.

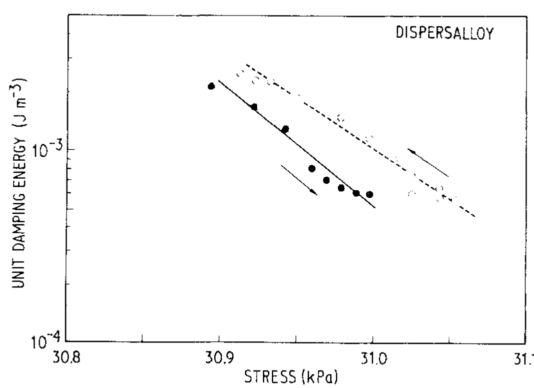


*Figure 10* Stress dependence of unit damping energy for Dispersalloy.

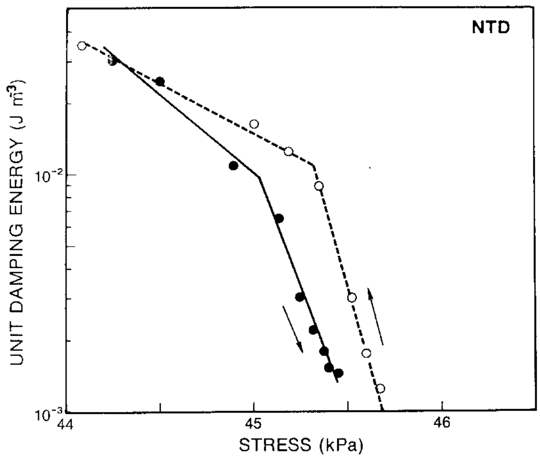


*Figure 11* Stress dependence of unit damping energy for NTD.

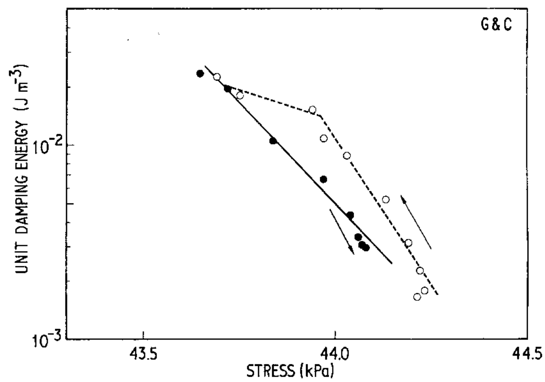


*Figure 13* Stress dependence of unit damping energy for G&C.

with the normal observation. This apparent inconsistency can be rationalized easily by considering the formula for UDE and keeping in mind that $\sigma$ and $T$ are simultaneous variables. Thus UDE $= \pi Q^{-1}\sigma^2/E = \pi Q^{-1}E\epsilon^2 = \pi Q^{-1}\sigma\epsilon$, where $\epsilon$ is maximum strain amplitude ($= \sigma/E$), and $Q^{-1} = f(T)$, $E = g(T)$ and $\sigma = \epsilon g(T)$, where $f$ is a strong function of temperature and $g$ is a weak function of temperature. Therefore, the stress dependence of UDE is essentially the inverse of the temperature dependence of UDE (i.e. as stress increases, temperature decreases), and changes in UDE are dominated by changes in $Q^{-1}$. When some of the UDE–$\sigma$ curves are given a left-to-right inversion they resemble the UDE–$T$ curves. The data for Tytin and G&C work very well in this respect.

Two other ways of examining the modulus and damping results are now discussed. The plots of $Q^{-1}$ as a function of $E$ (Figs. 14 to 18) are all decreasing functions. For Shofu the results could be fitted to one straight line if necessary (Fig. 14) but two curves – one for heating and one for cooling – are shown. In the case of Dispersalloy, NTD and G&C there is a definite hysteresis in the $Q^{-1}$–$E$ curves (Figs. 15, 16 and 18). For Dispersalloy (Fig. 15) there is an interesting effect near $E$ = 33.1 GPa (32° C) which is equivalent to the small peak seen in Fig. 2. Again its significance is not clear but the effect merits further research. The $Q^{-1}$–$E$ curve for G&C (Fig. 18) shows a sharp increase near $E$ = 46.9 GPa on heating only. This increase occurs near 75° C and again probably reflects changes associated with the $\gamma_1 \rightarrow \beta_1$ transformation. A striking example of sharp changes on the $Q^{-1}$–$E$ curves occurs in Fig. 17 for Tytin, again at temperatures near 68° C (heating) and 65° C

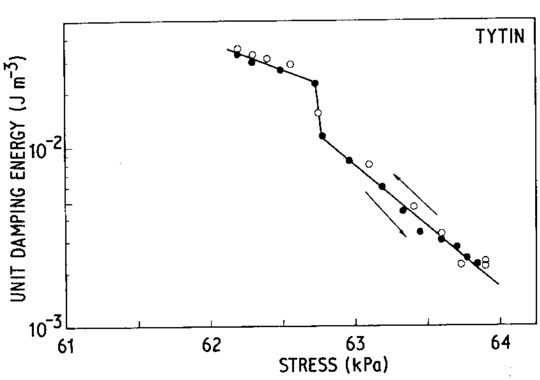


*Figure 12* Stress dependence of unit damping energy for Tytin.

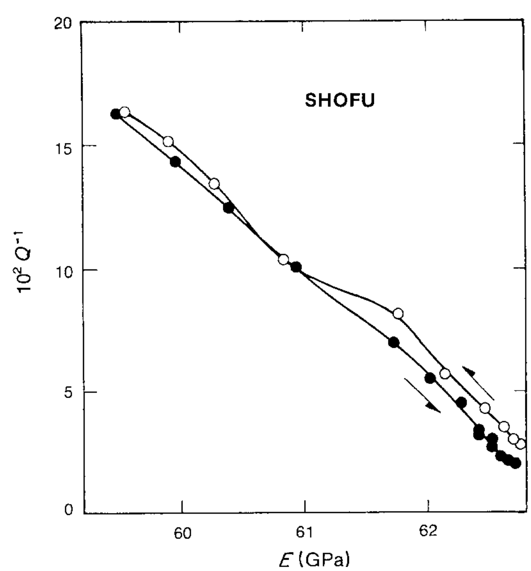


*Figure 14* Mechanical damping as a function of Young's modulus for Shofu.

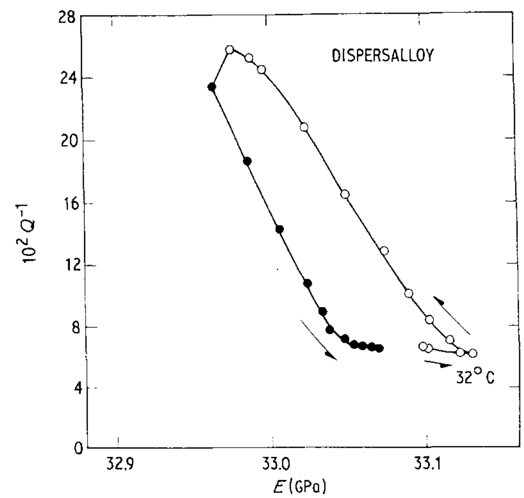


*Figure 15* Mechanical damping as a function of Young's modulus for Dispersalloy.

(cooling), at an $E$ value of 67 GPa, and is also probably related to the phase transformation.

The graphs of $Q^{-1}$ as a function of $E$ have been examined also in terms of $Q^{-1}/E$ against $E$, that is, the mechanical damping has been adjusted for the changes in Young's modulus. As may be deduced from the results in Figs. 14 to 18, the $Q^{-1}/E$–$E$ and $Q^{-1}$–$E$ curves are essentially the same in character because the changes in $E$ with temperature are small ($< 5\%$) for the range of temperatures covered in the experiments. Two representative $Q^{-1}/E$–$E$ curves (for Dispersalloy and Tytin) are shown in Figs. 19 and 20, respectively. The details mentioned

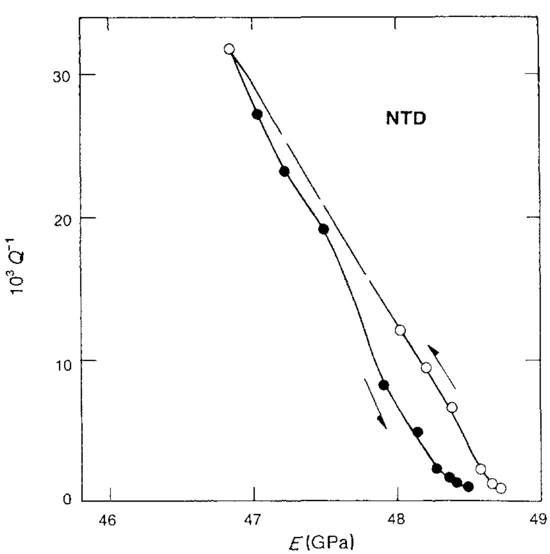


*Figure 16* Mechanical damping as a function of Young's modulus for NTD.

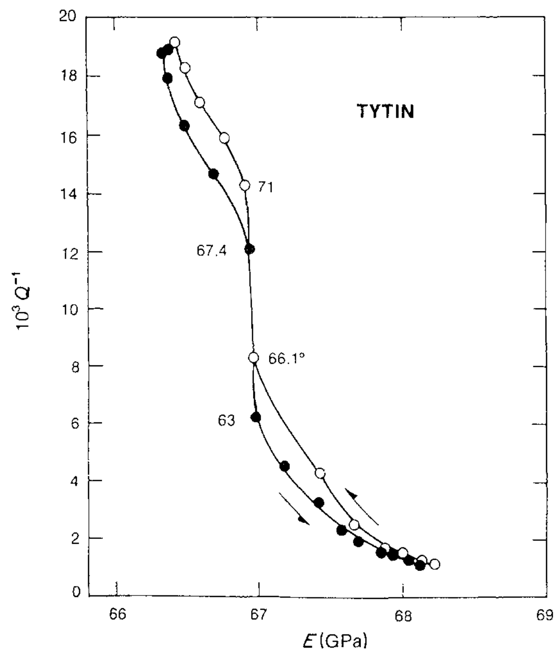


*Figure 17* Mechanical damping as a function of Young's modulus for Tytin.

earlier – the effect near 32° C in Dispersalloy (Fig. 15) and the sharp change in mechanical damping probably related to the phase change in Tytin (Fig. 17) – are also clearly documented in these figures.

## 5. Conclusions

The PUCOT has been used to study the elastic

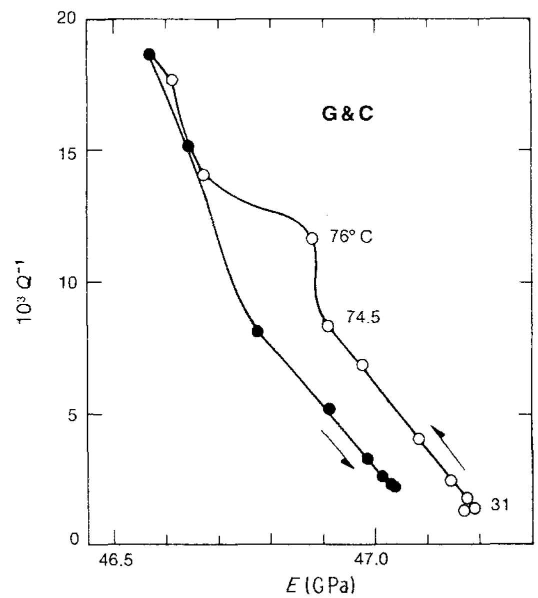


*Figure 18* Mechanical damping as a function of Young's modulus for G&C.

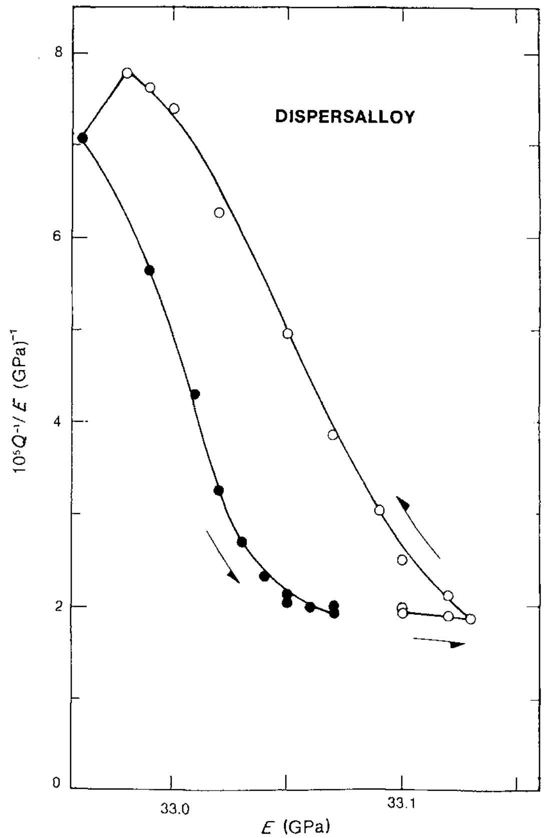


*Figure 19* The ratio of mechanical damping to Young's modulus as a function of Young's modulus for Dispersalloy.

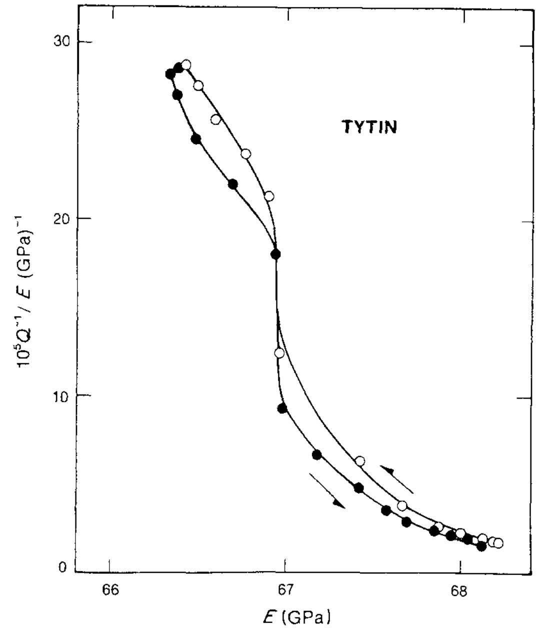


*Figure 20* The ratio of mechanical damping to Young's modulus as a function of Young's modulus for Tytin.

properties (Young's modulus $E$) and anelastic properties (mechanical damping $Q^{-1}$) of five silver dental amalgams. From experiments conducted over the temperature range 20 to 80° C the following measurements were made and then analysed: $E$, $Q^{-1}$, stress $\sigma$, temperature $T$ and unit damping energy UDE. The phenomenological analysis of the UDE–$T$, UDE–$\sigma$, $Q^{-1}$–$E$ and $Q^{-1}/E$–$E$ curves showed that their form was one of the following (depending on the amalgam): one, two, three or four straight lines, with or without hysteresis. All the curves for Tytin showed a sharp change near 68° C. The additional new results for Dispersalloy (the temperature dependencies of $E$ and $Q^{-1}$ and the time dependence of $E$) indicated that $Q^{-1}$ changes by a factor of 4.3 and $E$ is reduced by 0.5% as the temperature changes from 20 to 80° C. The effective activation energy for the change in $Q^{-1}$ of 0.42 eV is equal to the activation energy for diffusion of mercury in the $\gamma_1$ phase. The presentation of the elastic and anelastic data in the forms examined in this study is useful in the detection and monitoring of transformations in the Ag–Hg–Sn amalgams.

## Acknowledgements

Earlier discussions with Dr J. A. A. Hood of the University of Otago Dental School, New Zealand and more recent discussions with Dr Robert Deluca of Johnson and Johnson Dental Products Division, East Windsor, New Jersey are greatly appreciated.

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*Received 30 January and accepted 12 April 1984*