Thermal Properties of a Magnesium Hybrid Composite: QE22 Alloy Reinforced with 20 Vol% SiC Particles and 5 Vol% Al_2O_3 Fibres

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ABSTRACT

Thermal expansion of a hybrid composite QE22 + 20 vol% SiC_p + 5 vol% Al_2O_3 (Saffil) fibres was measured in the temperature range from 20 to 375°C. The thermal expansion coefficient (CTE) of this hybrid composite is compared with CTE of three composites: QE22/ 15 vol% SiC_p, QE22/ 25 vol% SiC_p and QE22/ 20 vol% Saffil. A decrease of CTEs at certain temperatures above 240°C indicates the thermally-induced elastic-plastic transition. The transition temperature is the highest for the hybrid composite. The lowest transition temperature has been estimated for QE22 reinforced with 25 vol% SiC. The experimental results indicate that the kind and volume fraction of reinforcement may very effectively influence the elastic-plastic transition temperature.

The effect of temperature on the thermal diffusivity and thermal conductivity of the hybrid composite QE22 + 20 vol% SiC_p + 5 vol% Saffil fibres was investigated in the temperature range from 20 to 300°C. The addition of SiC particles and Saffil fibres into magnesium alloy QE22 causes a reduction in both the thermal diffusivity and thermal conductivity at any temperature. The thermal diffusivity of this hybrid composite will be compared with those of QE22/ 25 vol% SiC_p and of QE22/ 20 vol% Saffil estimated in our prior study.

KEYWORDS

Magnesium alloys, magnesium composites, thermal properties.

INTRODUCTION

The widely used Mg-Ag-Nd-Zr alloy, commercially denoted as QE22 (Mg-2.5 wt% Ag – 2 wt% Nd rich rare earths – 0.7 wt% Zr) is well known as an age hardenable, high strength and creep resistant material. Considerable improvement in mechanical properties of Mg- based alloys is achieved by their reinforcement by ceramic particulates and fibres.

Kiehn *et al.* /1/ investigated precipitation effects in QE22 alloy reinforced with 22 vol% Al_2O_3 fibres. Precipitation in this composite prepared by squeeze casting was studied after a preceding solution heat treatment by isochronal annealing up to 300°C by means of electrical resistivity, hardness and reversible stress relaxation measurements. In the composite the fibres act as nucleation centres in the precipitation process promoting e.g. precipitation of Al_2Nd or Ag compounds (the Al content in the matrix is enhanced due to the decomposition of the preform binder). At 300°C, the transformation of all new particles occurs to semicoherent $Mg_{12}Nd$ particles. The precipitation process in unreinforced alloy is different involving only change of the morphological features of tetragonal semicoherent $Mg_{12}Nd$ particles already existing in the alloy in the initial state after solution heat treatment /2/. It was found that the thermal stability of the composite is extremely dependent on the microstructure.

Moll *et al.* /3/ and Mordike *et al.* /4/ described the creep behaviour of a QE22 alloy matrix composite reinforced with 15 vol% of SiC particles and fabricated by powder metallurgy procedure (further designated QE22/SiCp where p denotes particulates). It was found that the presence of SiCp in the creep resistant QE22 led to a decrease in the creep resistance. The thermal conductivity of the QE22/ SiC_p composites was studied as a function of temperature and concentration of reinforcement (10, 15 and 25 vol% SiC) in /5/. It was found that the lattice defects localized in the vicinity of reinforcements at room temperature expand in matrix during heating. The presence of SiC particles causes a decrease in the thermal conductivity. The thermal expansion and the thermal diffusivity of QE22/ 20 vol% Al_2O_3 fibres as a function of temperature are reported elsewhere /6/.

The objective of this paper is to determine the thermal properties such as thermal expansion, thermal diffusivity and thermal conductivity of a hybrid composite based on QE22 as a matrix with 20 vol% SiC particles and 5 vol% short Al_2O_3 fibres. The results will be compared with those obtained for the QE22 alloy reinforced only by either SiC_p or Al_2O_3 fibres.

EXPERIMENTAL DETAILS

The investigations were conducted using a QE22 alloy reinforced by 5 vol% of short Al_2O_3 fibres (the diameter of Al_2O_3 fibers was 3μm, the mean length was about 87μm) and 20 vol% SiC particles (the average diameter was 9μm). The hybrid composite was produced using squeeze cast technique. The thermal expansion was measured before (in the as prepared state) and after T6 heat treatment, the thermal diffusivity was measured in the as prepared state. The measured thermal properties will be compared with the thermal properties of QE22/ 15 vol% SiC_p and QE22/25 vol% SiC_p prepared by powder metallurgy /5/ and of QE22/20 vol% Al_2O_3 fabricated by squeeze casting /6/.

The linear thermal expansion was measured in argon atmosphere using the Netzsch 402E dilatometer in the temperature range from 20 to 375°C. The samples of QE22/20 vol% Al_2O_3 and hybrid composite were taken parallel to the planes of randomly oriented fibres.

The measurement of the thermal diffusivity was performed in the temperature range from 20 to 300°C in argon atmosphere using the flash method described elsewhere /7/. For the composite QE22/20 vol% Al_2O_3 and hybrid composite two orientations of fibres were studied, the random planar orientation of fibres parallel to the flat faces (marked $\perp$) and orientation perpendicular to it (marked $\|$). The specific heat capacity of QE22 alloy was calculated using the Neumann-Kopp rule, the specific heat of composites was calculated by the mixture rule. The temperature dependence of the density was calculated from the density measured at 20°C by weighing the sample in water and from respective values of volume thermal expansion data. The thermal conductivity was calculated using the relation $\lambda = a\rho c$, where a is the thermal diffusivity, ρ is the density and c is the specific heat capacity.

RESULTS AND DISCUSSION

It is known that the physical properties of magnesium alloys and composites depend on their thermal history. In our previous work /8/ dealing with the AZ91 alloy and AZ91/20 vol% Al_2O_3 fibres composite, we have shown that the thermal diffusivity and the thermal conductivity (in the temperature range from 20 to 300°C) for the first run of measurement are lower than for further runs even if the samples were exposed to the thermal treatment (18 hours at 413°C and then cooled by air).

In this work, the thermal expansion measurements were performed on the hybrid composite as prepared and after T6-heat treatment. In Fig.1 the temperature dependence of CTE of the as-prepared composite is shown for the first and second runs. The significant difference between both runs appears. On the contrary, only a small variance in the first and second runs was detected for the T6-treated composite, see Fig. 2. As to the temperature of the downward bend on the heating branches for second runs, the T6-treated composite exhibits a slightly lower value in comparison to the as prepared material. The hysteresis in the heating-cooling cycle after the T6-treatment is smaller than that for the as-prepared state.

In Fig. 3, the temperature dependencies of CTE of the hybrid composite, pure QE22 alloy, composite with 15 vol% SiC_p and 25 vol% SiC_p and QE22/20 vol%

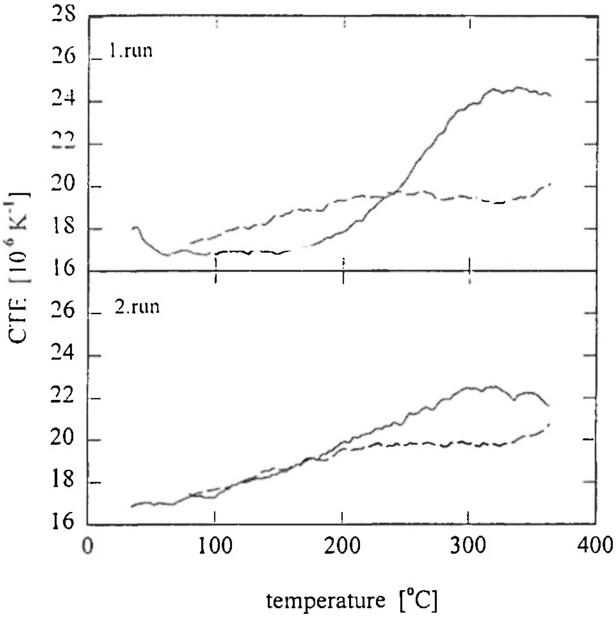

Fig. 1: Temperature dependence of CTE for as prepared hybrid composite, full line - heating, dashed line – cooling

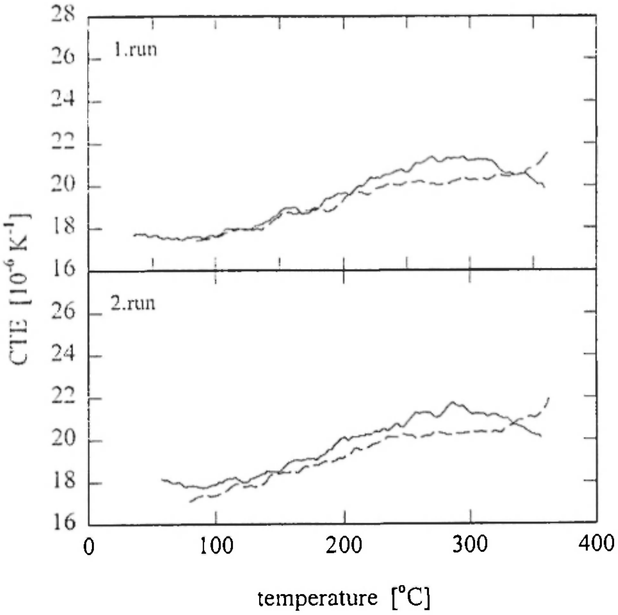

Fig. 2: Temperature dependence of CTE for composite after T6 heat treatment, full line – heating, dashed line – cooling

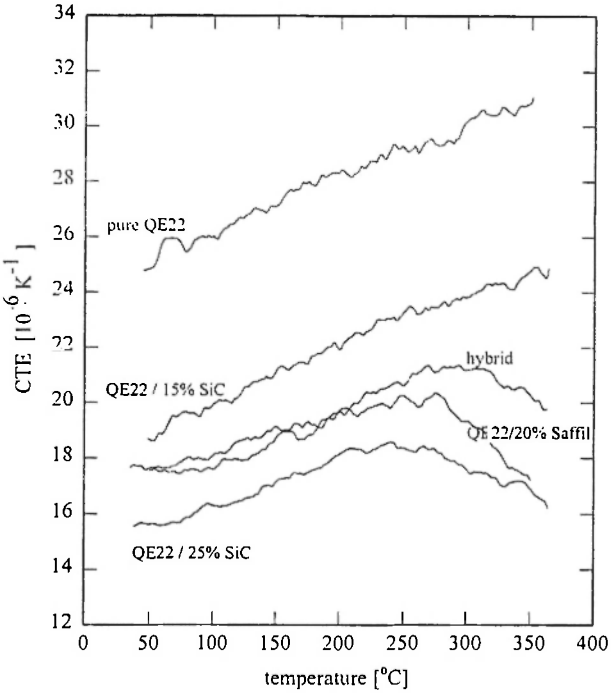

Fig. 3: Temperature dependence of CTE

Table 1
Temperature of the elastic-plastic transition

Composite	Temperature [°C]
QE22/15 vol% SiC	255
QE22/25 vol% SiC	240
QE22/20 vol% saffil	275
QE22/20 vol% SiC + 5 vol% saffil	286

Al_2O_3 fibres are presented for comparison. The curves for composites exhibit a striking downward bend in general. The temperatures of these bends, as dependent on the composite type, are summarised in Table 1.

To discuss this effect most simply but without loss of relevance, we will use some relations assuming the composite as elastically and thermally rigid spheroids embedded in the matrix. In fact, the reinforcements exhibit very low CTE (in $10^{-6}K^{-1}$): $\alpha_{SiC} = 4.5$, $\alpha_{Saffil} = 7$, and very high compression moduli (in GPa): $K_{SiC} = 227$, $K_{Saffil} = 278$, in comparison to the matrix: $\alpha_{QE22} = 25\text{-}31$, $K_{Mg\text{-}alloys} \cong 31$.

If only the elastic response to a temperature change takes place, the increment of elastic volume dilatation of the matrix is

$$\Delta\omega = -\frac{6(1-2\nu)f}{1+\nu+2f(1-2\nu)}\Delta\vartheta \qquad (1)$$

where ϑ is the linear thermal (stress free) dilatation of the matrix ($\alpha \equiv d\vartheta/dT$), f is the volume fraction of particulates, ν is Poisson's ratio. The maximum increment in the shear stress, defined as the difference between angular and radial stress increments near the phase interface, follows the relation

$$\Delta\tau = -\frac{3E}{1+\nu+2f(1-2\nu)}\Delta\vartheta \qquad (2)$$

where E is Young's modulus. The linear total dilatation $\Delta\varepsilon$ of the composite is given by

$$3\Delta\varepsilon = (\Delta\omega + 3\Delta\vartheta)(1-f)\,. \qquad (3)$$

In a pure thermo-elastic case, the uniform value (1) is here to be inserted. If the internal stresses are changing, a volume average of the local elastic dilatations $\Delta\omega$ should be used.

During technological cooling from the fabrication temperature it holds $\Delta\vartheta < 0$ and both the elastic dilatation as well as the shear stress increase to positive values ω_0 and τ_0, respectively.

Heating the composite slowly, the elastic dilatation of the matrix and the representative shear stress decrease firstly elastically according to eqs. (1) and (2), the shear stress $\tau_0 + \Delta\tau$ remaining below the relevant plasticity limit k. But owing to the thermal activation, one can expect a pronounced temperature drop of k(T) and the meeting of the condition

$$\tau_0 + \Delta\tau(T_h) = k(T_h) \qquad (4)$$

at a certain temperature. Then creep starts near the interface and the still positive elastic dilatation $\omega_0 + \Delta\omega(T_h)$ is non-elastically released in general. Consequently, the composite dilatation (3) is dropped with respect to its elastic extrapolation. We identify the downward bend on the heating CTE - curves with the condition (4) for the onset of local plasticity. Because the elastic slope $d\tau/d\vartheta$ according to eq.(2) is "less negative" for larger f, a decrease of T_h with increasing particle content is to be expected, which is experimentally confirmed in Fig.3.

To simply treat the spreading of plastic zones, we can only make some cautious estimates. According to familiar creep experience, the internal stresses σ_i of the dislocation origin are built up at the actually active plastic sites and the plasticity limit is modified to $\tau = \sigma_i + k(T)$. The plastic zones tend to displace into the fresh material with zero σ_i. under simultaneous increase of temperature. Thus, the internal stress in consequent plastic zones decreases according to the increasing temperature of its production.

Switching the heating to the cooling during the creep regime, a short elastic segment appears first when the representative shear stress is raised up as it follows from eq. (2). If the limit of reverse creep is reached, the shear stress in actually re-creeping zones follows the trajectory $\tau = \sigma_i - k(T)$ and increases initially as the zones with higher σ_i become active. But a maximum in the function $\sigma_i - k(T)$ via the pronounced kinetic drop of $-k(T)$ at low temperatures is to be expected. Hence, at a certain temperature T_c, the negative slope $\delta[\sigma_i - k(T)]/\delta T$ reaches the elastic value $\delta\Delta\tau/\delta T$ after eq. (2) and complete elasticity occurs. Similarly as in the case of heating, the region of reverse creep during cooling may be identified with the drop of CTE below the elastically interpolated values. Indeed, the reversion of creep rate clearly results in the time reversion of the corresponding effect on the elastic volume dilatation.

The final value τ_1 of the representative shear stress after cooling will serve as a new initial value τ_0 for further heating . In such a case (or if τ_1 and τ_0 do not differ extremely) the above sketched construction gives the result $T_c < T_h$, in full agreement with Figs. 1, 2.

The temperature dependence of the thermal diffusivity of the investigated hybrid composite is given in Fig.4. The samples were investigated as prepared. The temperature dependencies of QE22 alloy, QE22/15 vol% SiC, QE22/25 vol% SiC and QE22/20 vol% are also shown in Fig. 4. The values of thermal conductivity for the hybrid composite are summarised in Table 2.

Rudajevová and Lukáč /5/ compared the measured thermal conductivity of three composites QE22 with 10, 15 and 25 vol% SiC_p with the calculated thermal conductivity by Hasselman-Johnson relation /9/. It may be seen by inspection of the data given in /5/ that the

Table 2

Temperature dependence of the thermal conductivity for the hybrid composite

Temperature [°C]	20	100	200	300
Thermal conductivity [W/mK]	91.7	95.1	95.6	94.2

highest difference between measured and calculated values is for the composite with 25 vol% SiC_p. It was assumed that departure of measured and calculated values is due to the lattice defects which are localized at interfaces between particles and matrix at room temperature. As temperature increases the lattice defects may expand into the matrix and the thermal diffusivity and thermal conductivity decrease. The higher the temperature, the more dislocations are moving in the matrix (the more scattering places) and hence the larger is the decrease in the thermal conductivity and thermal diffusivity.

It is apparent from Fig.4 that the decrease in the thermal diffusivity of the hybrid composite with increasing temperature is lower than for QE22/25 vol% SiC_p composite. We assume that a combination of the particles and fibres as reinforcements of QE22 alloy may lead to a lower influence of interfacial defects on the thermal diffusivity and thermal conductivity. This is in agreement with the expansion results where the elastic-plastic transition temperature for hybrid composite is the highest of all those obtained for the composites in the present investigation.

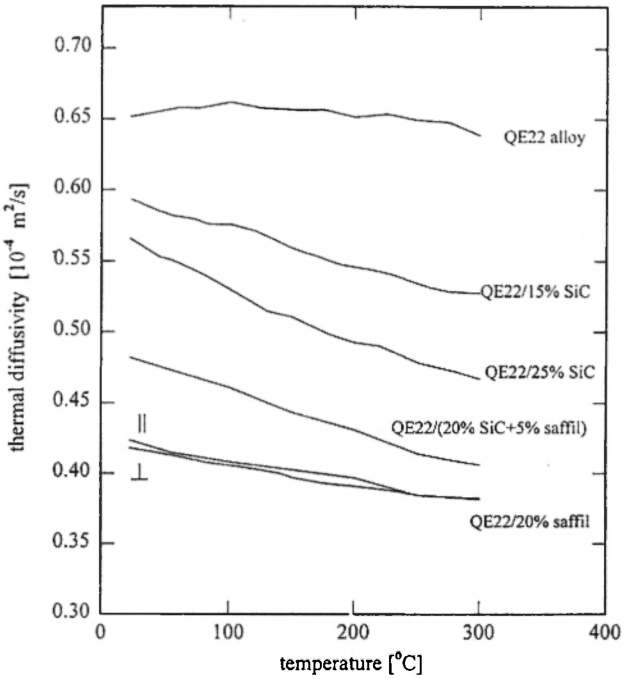

Fig. 4: Temperature dependence of thermal diffusivity

CONCLUSIONS

The CTE and the thermal conductivity of QE22 magnesium alloy reinforced with 20 vol% of SiC particles and 5 vol% of Al_2O_3 short fibres were investigated in a wide temperature range. The CTE of the hybrid composite increases with increasing temperature up to 286°C and then it decreases. The downward bent on the heating CTE curve was identified with local plasticity. The addition of SiC particles and Al_2O_3 short fibres into QE22 alloy reduces the thermal diffusivity and thermal conductivity at any temperature. The thermal diffusivity and thermal conductivity are influenced by the kind and content of the reinforcement in matrix.

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REFERENCES

1. J. Kiehn, B. Smola, P. Vostrý, I. Stuliková and K.U. Kainer, *phys. stat. sol. (a)*, **164**, 709 (1997).
2. J. Kiehn, W. Riehemann and K.U. Kainer, *Mat. Sci. Forum* **210-213**, 503 (1996).
3. F. Moll, K.U. Kainer. and B.L. Mordike, In: *Magnesium Alloys and Their Application*, B.L. Mordike and K.U. Kainer (Eds)., Werkstoff-Informationsgesellschaft, Frankfurt, 1998; p.647.
4. B.L. Mordike, K.U. Kainer, F.Moll. and B. Sommer, In: *Proceedings of the First Israeli International Conference on Magnesium Science & Technology*, E Aghion, D.E. Eliezer (Eds)., Dead Sea, Israel, 1997; p.178

5. A. Rudajevová and P. Lukáč, *J. Mat. Sci. Letts.* in press.
6. A. Rudajevová and P. Lukáč, *phys. stat . sol. (a)*, **175**, 457 (1999) .
7. A. Rudajevová, *Mat. Res. Bull.* **26,** 1363 (1991).
8. A. Rudajevová, J. Kiehn, K.U. Kainer, B.L. Mordike and P. Lukáč, *Scripta Mat.* **40**, 57 (1999).
9. D.P.H. Hasselman and L.F. Johnson, *J. Comp. Mater.* **21**, 508 (1987).