INFLUENCE OF THE INTERFACE ON THE THERMAL EXPANSION AND THERMAL CONDUCTIVITY OF QE22 BASED COMPOSITES

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We report on the influence of the interface between the metal matrix and ceramic reinforcement on the thermal expansion and thermal conductivity of three composites – QE22 with 25 vol.% of SiC particles, QE22 with 20 vol.% of Saffil fibres and QE22 with 20 vol.% of SiC and 5 vol.% of Saffil fibres – which was investigated in the temperature range of 20–380 °C. The composites become macroscopically deformed at room temperature due to different thermal expansion coefficients of two components. The thermoelastic stress field in the matrix is influenced by the matrix plastic deformation near the interface. The strongest changes in the thermal expansion coefficient and thermal conductivity occur at temperatures at which the thermoelastic stress releases with the reduction of the plastic deformation.

K e y $\,$ w o r d s: magnesium composites, thermal expansion, thermal conductivity, thermal strain

1. Introduction

Thermoelastic stresses occur in composites due to different contraction of the metal matrix and ceramic reinforcement during cooling within the technological process and yield a deformation of a composite at room temperature. Thermoelastic stresses and accompanying strains can be reduced by heating the composite to reach a stress free state usually at about 0.8 $T_{\rm m}$, where $T_{\rm m}$ is the melting point. This change of the strain with temperature influences various physical properties. Thermal strains in the WC-11vol.%Co composite during two thermal cycles have been determined by Mari et al., using neutron diffraction methods [1]. Initially the lattice parameter increases linearly with increasing temperature up to certain temperature from which gradual reduction of this parameter begins. Warwick and

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Clyne [2] measured the thermal expansion as a function of temperature during the first 3 thermal cycles for the composite of Mg with 11 wt.% of Li and 20 vol.% of SiC_w. An analysis of the release of thermoelastic stresses is presented in [3]. The composite is plastically deformed in matrix near the interface at room temperature. The release of thermoelastic stresses with increasing temperature has initially elastic character. Above the certain temperature the plastic deformation in the interface becomes reduced, the elastic stress field in the composite is further modulated by this process and on the temperature dependence of the lattice constant [1] or CTE [2, 3] a bend occurs.

The relative elongation of a composite is composed of two components; one is due to thermal vibrations and the other due to changes of the thermal strain with temperature. The dilatation thermal strain $\Delta d/l_o$ can be expressed by the following relation:

$$\frac{\Delta d}{l_{\rm o}} = \left(\frac{\Delta l}{l_{\rm o}}\right)_{\rm meas.} - \left(\frac{\Delta l}{l_{\rm o}}\right)_{\rm mix.rule},\tag{1}$$

where $(\Delta l/l_{\rm o})_{\rm meas.}$ is the measured relative elongation and $(\Delta l/l_{\rm o})_{\rm mix.rule}$ is the relative elongation of the composite calculated by the rule of mixtures. The dilatation thermal strain $\Delta d/l_{\rm o}$ represents the departure of the composite behaviour from the state where no thermal stresses are present.

The thermal conductivity is influenced by the changes in the interface as well. Hasselman and Johnson [4], considering the thermal conductance of the interface h, derived relations for the thermal conductivity of the composites with particular and fibre reinforcements. The first relation (2) was derived for composites with spherical particles, the second relation (3) for the short fibre composites and the heat flow perpendicular to the fibres:

$$K_{\rm c} = K_{\rm m} \frac{\left[2f\left(\frac{K_{\rm p}}{K_{\rm m}} - \frac{K_{\rm p}}{r \cdot h} - 1\right) + \frac{K_{\rm p}}{K_{\rm m}} + 2\frac{K_{\rm p}}{r \cdot h} + 2\right]}{\left[f\left(1 - \frac{K_{\rm p}}{K_{\rm m}} + \frac{K_{\rm p}}{r \cdot h}\right) + \frac{K_{\rm p}}{K_{\rm m}} + 2\frac{K_{\rm p}}{r \cdot h} + 2\right]},$$
(2)

$$K_{\rm c} = K_{\rm m} \frac{\left[f\left(\frac{K_{\rm f}}{K_{\rm m}} - \frac{K_{\rm f}}{r \cdot h} - 1\right) + \frac{K_{\rm f}}{K_{\rm m}} + \frac{K_{\rm f}}{r \cdot h} + 1 \right]}{\left[f\left(1 - \frac{K_{\rm f}}{K_{\rm m}} + \frac{K_{\rm f}}{r \cdot h}\right) + \frac{K_{\rm f}}{K_{\rm m}} + \frac{K_{\rm f}}{r \cdot h} + 1 \right]},\tag{3}$$

where $K_{\rm c}$ and $K_{\rm m}$ is thermal conductivity of the composite and matrix, resp., $K_{\rm p}$ and $K_{\rm f}$ is thermal conductivity of the particles or fibres, resp., f is the volume fraction of the reinforcement and r is the radius of particles.

The study of the dilatation strain and the thermal conductance of the interface can contribute to the knowledge of the interface behaviour. The aim of this work is to compare the influence of various kinds of reinforcements on the dilatation strain and interfacial conductance. The thermal expansion and thermal conductivity of the QE22 alloy reinforced by 25 vol.% of SiC particles, 20 vol.% of Saffil short fibres, and 20 vol.% of SiC particles with 5 vol.% of Saffil fibres (hybride) was determined and analysed in the temperature range of 20-380 °C.

2. Experimental

The QE22/SiC composite was prepared by the powder metallurgical method (composition of the QE22 alloy: Mg-2.5Ag-2Nd-0.2Zr). The QE22/Saffil fibre and QE22/hybride composite were produced using the squeeze cast technique. All the composites were produced in laboratories of the Technical University of Clausthal, Germany. The specimens were investigated in the as-extruded state.

The linear thermal expansion was measured in argon atmosphere using the Netzsch 402E dilatometer in the temperature range from 20° C to 380° C with the heating and cooling rate of 5 K/min. The samples had the diameter of 6 mm and length of 50 mm. In the QE22/Saffil composite the planes of 2D-random fibre array were parallel to the longitudinal sample axis. The thermal conductivity was determined using the relation

$$K = ac\rho,\tag{4}$$

where a is the thermal diffusivity, ρ is the density and c is the specific heat capacity. 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 [5]. The fibres in the QE22/Saffil and QE22/hybride composites were arranged parallel to the planes of randomly oriented fibres. The heat capacity of the QE22 alloy was calculated using the Neumann-Kopp rule and the heat capacity c of the composite was calculated from the following relation:

$$c = \frac{\left(f_{\rm m}\rho_{\rm m}c_{\rm m} + f_{\rm r}\rho_{\rm r}c_{\rm r}\right)}{\rho_{\rm c}},\tag{5}$$

where f is the volume fraction of particles and subscripts m and r and c refer to matrix, reinforcement and composite, respectively. The temperature dependence of the density was calculated from the density measured at 21 °C by weighing the sample in toluene and from respective values of the volume thermal expansion calculated from expansion data assuming isotropic dilatation. Three thermal cycles were conducted on each sample. We present results of the second or third thermal cycle, which show the same results.

3. Results

Figure 1 shows the temperature dependence of the CTE for the QE22 alloy and all the composites studied during heating. The temperature dependence for the QE22 alloy is monotonous in the whole temperature range studied whereas for all the composites it bends above 200 °C. The temperature of the bend on the temperature dependence of the CTE is named the transition temperature. The values of observed transition temperature are presented in the Table 1.

In Fig. 2 the temperature dependence of the dilatation strain, determined from the relation (1), is displayed for all the studied composites. All the observed values are negative which means that the "composite effect" yields the sample contraction being smallest for the QE22/hybride composite.

Temperature dependence of the thermal conductivity for the three composites and the QE22 alloy is shown in Fig. 3. The QE22 alloy reinforced by SiC particles exhibits different character of the dependence than the QE22 alloy and the QE22/Saffil fibre composite. Figure 4 shows the temperature dependence of the thermal conductance of the interface for the QE22/SiC and QE22/Saffil composites calculated using relations (2) and (3). The thermal conductance of the





Fig. 1. Temperature dependence of the CTE for QE22 alloy and QE22/SiC, QE22/Saffil and QE22/hybride composites.

Fig. 2. Temperature dependence of the dilatation strain for QE22/SiC, QE22/Saffil and QE22/hybride composites.

Table 1. Transition temperatures for the QE22/SiC, QE22/Saffil and QE22/hybride composites

Composite	QE22/SiC	QE22/Saffil	QE22/hybride
Temperature [°C]	240	270	290





Fig. 3. Temperature dependence of the thermal conductivity for QE22 alloy and QE22/SiC, QE22/Saffil and QE22/hybride composites.

Fig. 4. Temperature dependence of the thermal conductance for QE22/SiC and QE22/Saffil composites.

interface for the QE22/Saffil fibre composite is nearly constant up to $270 \,^{\circ}$ C and then it increases. The thermal conductance of the QE22/SiC composite was found to decrease with increasing temperature above $70 \,^{\circ}$ C.

4. Discussion

In our previous paper [6] it was shown that the residual strain occurs in alloys or composites due to volume changes during their preparation, after rapid cooling or mechanical loading. The residual strain, which can influence various physical properties, is released during the first heating. It is therefore important to study the influence of the interface on physical properties in the second thermal cycle after the residual thermal strain has been removed.

Note that the thermal strain exists in the composite even after removing the residual strain. It is highest at room temperature and decreases with increasing temperature to reach the stress-free state at high temperatures (> $0.8 T_{\rm M}$). During cooling in the thermal cycle the thermal strain increases to reach the initial value of the thermal strain at room temperature. It was shown in our previous work [3] that this deformation has elastic-plastic character. The plastic deformation exists in the matrix near the interface at room temperature and modulates the elastic stress field in the matrix.

Figure 5 shows the scheme [7] of the release of the shear stress during the

thermal cycle. The shear stress decreases first elastically (linear dependence of the shear stress on the temperature) up to reaching the yield stress limit. The shear

stress decreases dramatically above this temperature because the plastic deformation in the interface is removed. The decrease in the plastic deformation in the matrix near the interface is connected with the bends in the temperature dependence of the CTE and dilatation strain (Figs. 1 and 2). The transition temperature is the temperature at which the yield stress is reached. It can be seen in Fig. 5 that the higher initial shear stress the lower transition temperature. The highest transition temperature was found for the QE22/hybride composite (290 °C), the lowest for the QE22/SiC (240° C) one and hence we conclude that the highest thermal strain at room temperature is



Fig. 5. Scheme of the temperature dependence of the representative shear stress.

in the QE22/SiC composite and the lowest in the QE22/hybride one.

Temperature dependences of the thermal conductivity for the QE22 alloy and composites studied are in Fig. 3. The thermal conductivity of the composites with SiC particles has a different character of the temperature dependence than the QE22 alloy and the QE22/Saffil fibre composite.

Using the relations (3) and (4) we obtained the temperature dependences of the thermal conductance of the interface for the composites QE22/SiC and QE22/Saffil. The values of $K_{\rm m}$ were measured (Fig. 3), the thermal conductivity values of SiC (decreasing from 90 $\rm W\cdot m^{-1}\cdot K^{-1}$ at room temperature to 80 $\rm W\cdot m^{-1}$ $\cdot K^{-1}$ at 300 °C) and Saffil (decreasing from 30 W $\cdot m^{-1} \cdot K^{-1}$ at room temperature to 17 W \cdot m⁻¹ \cdot K⁻¹ at 300 °C) were taken from literature [8, 9]. The radius of SiC particles and Saffil fibres was 4.5 and 1.5 μ m, respectively. Figure 4 shows that the thermal conductance of the interface for the QE22/SiC composite is far higher than that for the QE22/Saffil composite. The thermal conductance of the interface for the QE22/SiC composite decreases with increasing temperature already from 80 °C. This expressive decrease cannot be elucidated by the change of the interface. The interface is defined as a plane and the thermal conductance of the interface depends on the contact plane between the two components. The contact plane increases with increasing temperature due to thermal expansion. The increasing thermal conductance of the interface is therefore expected during heating. The observed decrease in the thermal conductance of the interface with increasing temperature indicates other mechanisms overcompensating the effect due to the interface expansion. Flom and Arsenault [10] reported that the resultant dislocation densities at the Al-SiC interfaces could be very high, about 10^9-10^{10} cm⁻². Similar dislocation densities may be expected at the QE22/SiC interfaces. We assume that these dislocations are localized in the immediate vicinity of the reinforcement at low temperatures [11, 12]. With increasing temperature they can expand into the matrix and the thermal conductivity of the matrix decreases. Thermal conductivity of the matrix is therefore different from the thermal conductivity of the pure alloy, which we used in our calculations.

The increase of the interface thermal conductance above 270 °C for the QE22/ Saffil fibre composite can be attributed to the decreasing plastic deformation in the matrix near the interface as a consequence of the decrease of the thermal strain. It was mentioned that the matrix is plastically deformed near the interface at room temperature. The plastic deformation may be the source of the defects scattering the electrons and phonons. When the yield stress is reached during the release of the thermal strain at certain temperature (Fig. 5) the plastic deformation in matrix near the interface vanishes and the thermal conductivity increases. The increase of the thermal conductivity at high temperatures (above 250 °C) was observed even for the QE22/hybride composite (Fig. 3). Our results show that the thermal conductivity of the metal matrix composite is lowered by the plastic deformation of the matrix near the interface at low temperatures.

5. Conclusions

In all the composites studied the plastic deformation of the matrix near the interface influences the thermal expansion characteristics and thermal conductivity. The large departure of the CTE from linearity occurs in all composites at the temperature at which the plastic deformation in the matrix near the interface decreases. This elastic-plastic transition temperature was the lowest for the QE22/SiCcomposite, the highest for the QE22/hybride composite. The thermal conductivity of the QE22/SiC and the QE22/hybride composite are strongly influenced by the propagation of dislocations from the interface into matrix. The reduction of the plastic deformation in the matrix near the interface yields an increase of the thermal conductivity. The use of various relations for the calculation of the composite thermal conductivity combining the thermal conductivity values of components leads to wrong results because the values of the thermal conductivity of the matrix and of the pure alloy, respectively, are rather different. The matrix in the composite contains various lattice defects, which considerably reduce the thermal conductivity. The thermal conductivity of the alloy in a composite is always lower than for the entire alloy.

Acknowledgements

The paper is dedicated to Prof. RNDr. Pavel Lukáč, DrSc., Dr.h.c. at the occasion of his $70^{\rm th}$ birthday.

This work is a part of the research program MSM 0021620834 that is financed by the Ministry of Education of the Czech Republic. The authors acknowledge financial support from the Grant Agency of the Academy of Sciences under the grant no. A2112302.

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