THERMAL EXPANSION IN MAGNESIUM COMPOSITES

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The thermal expansion behaviour of Al_2O_3 fibre-reinforced magnesium composites was studied in the temperature range from room temperature to 375 °C. The linear thermal expansion curves of the composites show thermal hysteresis. After the first complete heating and cooling cycle irreversible changes of the specimen length are observed; permanent (residual) strains are measured. The coefficient of thermal expansion of the composites depends on the volume fraction of fibres. The temperature dependence of the thermal expansion coefficient for heating stage is different from that measured in cooling stage of the cycle. A rapid drop in the temperature variation of the coefficient occurs at a certain higher temperature during heating stage of the first cycle. The results are discussed.

Key words: thermal expansion, coefficient of thermal expansion, magnesium composite

TEPLOTNÍ ROZTAŽNOST HOŘČÍKOVÝCH KOMPOZITŮ

Teplotní roztažnost hořčíkových kompozitů zpevněných vlákny Al_2O_3 byla studována při teplotách od pokojové až do 375 °C. Křivky lineární teplotní roztažnosti kompozitu vykazují hysterezi. Po prvním teplotním cyklu (ohřev a chlazení) pozorujeme reziduální (ireverzibilní) změnu délky kompozitních vzorků. Koeficient teplotní roztažnosti závisí na objemovém podílu zpevňujících vláken. Teplotní závislost koeficientu teplotní roztažnosti měřená během ohřevu je jiná než meřená během chlazení. Během ohřevu v prvním cyklu dochází při určité teplotě k prudkému poklesu koeficientu teplotní roztažnosti. Výsledky se diskutují.

1. Introduction

Some industrial applications require lightweight materials. Among them, magnesium based alloys and composites have been developed and used. In recent years, the investigations of properties of magnesium composites have attached increasing

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scientific attention due to their low density and significant improvement in physical and mechanical properties (e.g. high specific strength) compared with unreinforced magnesium alloys [1–5]. It is well known that many properties of metals are influenced by temperature. Thermal behaviour of a material determined by the coefficient of thermal expansion (CTE) and thermal conductivity and good dimensional stability in a certain temperature range are important for components. The coefficient of thermal expansion of monolithic alloys is usually a simple function of temperature and it is not affected by heating or cooling rate. In many cases, composites are often subjected to thermal cycling and changes in temperature. The CTE of the composite may be a complex function of temperature. The temperature dependence of the CTE of a composite depends not only on the composition and the volume fraction of the composite components but also on the thermal treatment, cooling and heating rate, and fabrication processes. The CTE is one of the important properties because it may characterise the thermal stability of a metal matrix composite (MMC) subjected to temperature change. The CTEs of the MCC play a key role in geometrical changes of the composite. The thermal behaviour of a MMC is sensitive to temperature changes due to the large differences between the CTEs of the matrix (metal or alloys) and the reinforcement (ceramics). When the composite temperature is changed, thermal misfit strains at the interface and therefore internal thermal stresses can be created. If temperature variations are large enough, the internal thermal stresses can reach a considerable magnitude and may lead to the matrix yielding. Then a permanent deformation of the MMC may result [6]. The thermal stresses can influence considerably the mechanical and damping composite properties as reported by Trojanová et al. [7] for the QE22-SiC composites under stress-free thermal cycling. Measurements of the CTE of a MCC can be used as an indirect test of the residual thermal stresses formation. The temperature variation of the CTE of a composite may indicate the permanent deformation of the composite.

The composite can be deformed under an applied stress and/or due to a temperature change. In the following analysis we assume that no interface sliding occurs. For simplicity we consider that the specimen deformation is essentially one-dimensional. For a small temperature change ΔT, the thermal strain of an unconstrained solid is $\alpha\Delta T$, where α is the coefficient of linear thermal expansion. In a composite the thermal strain of the matrix and the reinforcement is $\alpha_{\mathrm{m}}\Delta T$ and $\alpha_{\mathrm{r}}\Delta T$, respectively, where the subscripts m and r refer to matrix and the reinforcement. These strains are unequal because $\alpha_{\mathrm{r}} < \alpha_{\mathrm{m}}$. At the reinforcement-matrix interface the strain continuity is required and, thus, a mechanical strain ε due to the presence of the other component must be superimposed on the stress-free thermal strain. The total strain (mechanical plus thermal) of either component occurring in one direction can be expressed as

$$\varepsilon_{\mathrm{m}} = \sigma_{\mathrm{m}}/E_{\mathrm{m}} + \alpha_{\mathrm{m}}\Delta T, \tag{1a}$$

$$\varepsilon_{\mathrm{r}} = \sigma_{\mathrm{r}}/E_{\mathrm{r}} + \alpha_{\mathrm{r}}\Delta T, \tag{1b}$$

where σ_{m} and σ_{r} are the stresses generated in the matrix and reinforcement, and E_{m} and E_{r} are the Young's moduli of the corresponding components. From the condition of strain compatibility it follows

$$\varepsilon_{\mathrm{m}} = \varepsilon_{\mathrm{r}} = \varepsilon_{\mathrm{c}} = \alpha_{\mathrm{c}}\Delta T, \tag{2}$$

where α_{c} is the linear coefficient of thermal expansion of the MMC. The change of the total strain ε_{c} with temperature is the in-situ coefficient of thermal expansion of the composite. The relative elongation of the MMC can be measured easily.

The stress equilibrium equation for a composite is simply given by the rule of mixtures

$$\sigma_{\mathrm{m}} f_{\mathrm{m}} + \sigma_{\mathrm{r}} f_{\mathrm{r}} = \sigma_{\mathrm{a}}, \tag{3}$$

where σ_{a} is the externally applied stress and f_{r} and $f_{\mathrm{m}} = 1 - f_{\mathrm{r}}$ are the volume fractions of the composite components.

In the absence of external loading the stress equilibrium equation for two components is simply

$$\sigma_{\mathrm{m}} f_{\mathrm{m}} + \sigma_{\mathrm{r}} f_{\mathrm{r}} = 0. \tag{4}$$

Using Eqs. (1–4) one gets

$$\alpha_{\mathrm{c}} = \alpha_{\mathrm{m}} - (\alpha_{\mathrm{m}} - \alpha_{\mathrm{r}})/[1 + (f_{\mathrm{m}}/f_{\mathrm{r}})(E_{\mathrm{m}}/E_{\mathrm{r}})]. \tag{5}$$

If the CTE, Young's modulus and the volume fraction for each component are known, the CTE for the composite could be calculated [8].

The aim of the present paper is to measure the linear thermal expansion of two magnesium composites with different volume fraction of Al_2O_3 fibres and to estimate the coefficient of thermal expansion of the composites. The values of the CTEs are also important for applications.

2. Experimental

The experiments were carried out on Mg – 9 volume fraction of Al_2O_3 (Saffil) fibres supplied by the Institute of Materials and Machine Mechanics, Slovak Academy of Sciences, Bratislava, and on Mg – 30 volume fraction of Al_2O_3 supplied by the Institute of Materials Engineering, Technical University, Clausthal. The $9Al_2O_3/Mg$ composites were manufactured by the gas pressure infiltration of evacuated fibrous preform with magnesium melt in an autoclave. The preform consisting of discontinuous δ-alumina fibres was prepared by the ultrasonic stimulated dispersion of commercial Saffil® product in a water medium, subsequent sedimentation and drying at 150 ℃ to obtain planar random distribution of fibres. A mean fibre length and a fibre diameter were about of 100 μm and 3 μm, respectively. The preform was embedded without any binder into evacuated low-carbon steel container (8 × 8 × 70 mm) and infiltrated under argon gas pressure at 690 ℃/6 MPa/30 s with molten magnesium.

The $30Al_2O_3/Mg$ composites were prepared by squeeze casting method. The preform consisted of planar randomly distributed δ-alumina short fibres with a mean length of about 87 μm and a mean diameter of 3 μm and a binder system (containing Al_2O_3 and starch). Preforms were preheated to a temperature higher than the melting point of magnesium and then inserted into preheated die (290–360 ℃). Preforms were infiltrated using two-stage application of the pressure (50 and 130 MPa, respectively).

The linear thermal expansion was measured in argon atmosphere using the Netzsch 402E dilatometer over a temperature range from room temperature (RT) to 375 ℃ at heating and cooling rates of 5 K/min. Specimens with a mean length of 50 mm and a mean diameter of 6 mm were used. Planes with planar randomly distributed fibres were parallel with the specimen axis. The accuracy of the apparatus was determined that the measured values of CTE of pure Mg were compared with those published in literature. A perfect agreement was obtained. At least three thermal (heating and cooling) cycles (runs) were performed to estimate the thermal expansion curves for each specimen. The composites were studied in the as-cast state. The volume concentration of the fibres was determined using the density measurements.

3. Results

The temperature dependences of the relative elongation and the CTE for $9Al_2O_3/Mg$ composite obtained in the first cycle (run) are shown in Fig. 1. It can be seen that the irreversible reduction of the specimen length occurs after the thermal cycle. On the other hand, no change of the specimen length is observed in the second and following thermal cycles as shown for the second cycle in Fig. 2 (the third cycle had the same course as the second one). From the temperature dependences of the CTE measured in both runs, a perfect agreement between the cooling

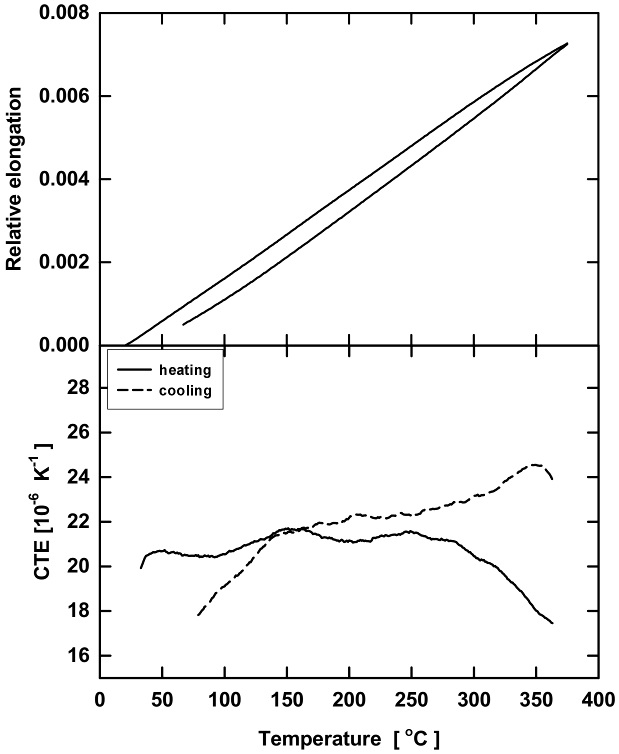

Fig. 1. Thermal linear expansion curve and the temperature dependence of the coefficient of thermal expansion in the first heating and cooling cycle for $9Al_2O_3/Mg$ composite.

branches is obvious, whereas different variations of the CTE with temperature are observed during heating stages of the cycles. The values of the CTE measured during heating in the second cycle are higher than those measured in the first cycle. It is important to note that the values of the CTE measured during heating decrease with increasing temperature if the test temperature is above about 250 ℃. The values of the CTE (for heating and cooling) are between 18×10^{-6} K^{-1} and 24×10^{-6} K^{-1}.

The values of the CTE for the composite are lower than those for pure magnesium. Baker [9, 10] determined that at temperatures between 0 and 550 ℃, the coefficient of linear thermal expansion of polycrystalline magnesium can be expressed as

$$\alpha = (25.0 + 0.0188t) \times 10^{-6}\ \mathrm{K}^{-1}, \qquad (6)$$

where t is the value of the temperature in ℃. The mean CTE for pure Mg in the

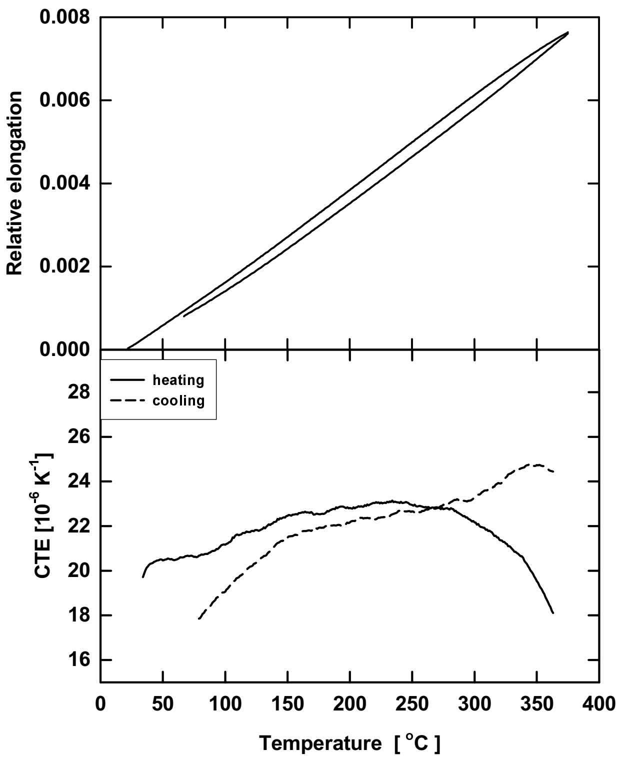

Fig. 2. Thermal linear expansion curve and the temperature dependence of the coefficient of thermal expansion in the second heating and cooling cycle for $9Al_2O_3$/Mg composite.

temperature range from 20 to 400 ℃ is 29×10^{-6} K^{-1}. It is obvious that the values of the CTE of Mg are higher than those for the $9Al_2O_3$/Mg composite. The value of CTE for pure Mg at room temperature is 25.2×10^{-6} K^{-1}.

The temperature dependences of the relative elongation and of the CTE for $30Al_2O_3$/Mg composite were estimated both in the temperature range from room temperature to 375 ℃ and at temperatures ranging from room temperature to 150 ℃ which are presented in Figs. 3 to 6. Figures 3 and 4 show the temperature dependences of the relative elongation and of the CTE for $30Al_20_3$/Mg obtained between room temperature and 375 ℃ in the first cycle and in the second cycle, respectively. It can be seen that whereas the permanent change of the specimen length occurs after the first cycle, only the reversible change of the specimen length is observed after the second and subsequent cycles. The irreversible shrinkage of the $30Al_2O_3$/Mg composite is higher than that of the $9Al_2O_3$/Mg composite.

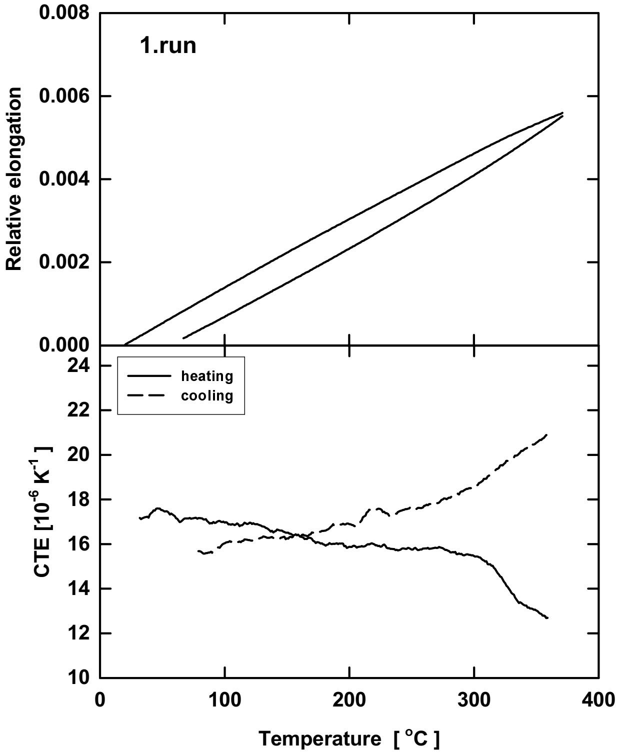

Fig. 3. Thermal linear expansion curve and the temperature dependence of the coefficient of thermal expansion in the first heating and cooling cycle for $30Al_2O_3/Mg$ composite.

The CTE during heating in the first cycle is slowly decreasing with increasing temperature from the value of about 18×10^{-6} K^{-1} to about 12.4×10^{-6} K^{-1} whereas the CTE during heating in the second cycle is constant below 250 ℃ and above this temperature the CTE decreases with increasing temperature. The values of the CTE in the second cycle are higher than in the first one. The temperature variations of the CTE during cooling stages are the same. The CTE values increase with increasing temperature. The range of values is 16 to 21×10^{-6} K^{-1}.

Figures 5 and 6 show the temperature dependences of the relative elongation and of the CTE for $30Al_2O_3/Mg$ estimated between room temperature and 150 ℃ in the first cycle and in the second one. The irreversible change of the specimen length after the first cycle is much lower than that obtained after the thermal cycle between room and 375 ℃. No hysteresis is observed during the second cycle. The values of the CTE in cooling stages are the same for both cycles (runs) and they

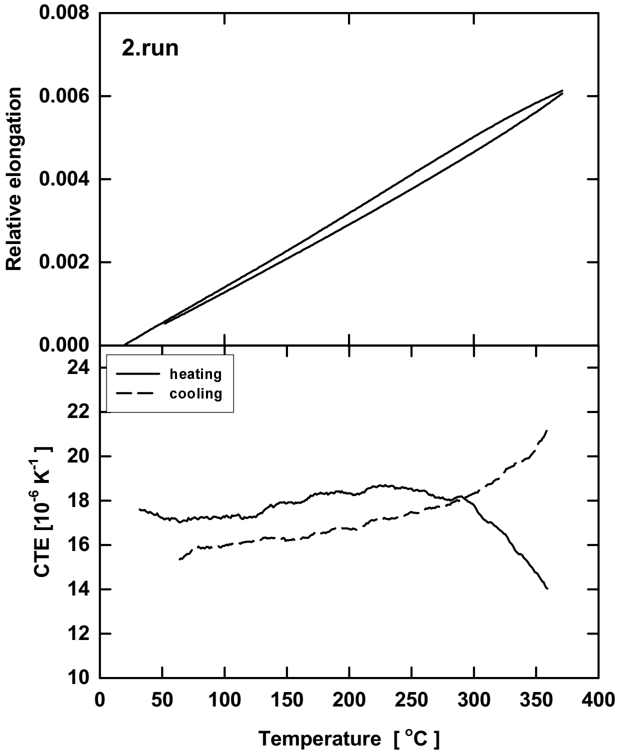

Fig. 4. Thermal linear expansion curve and the temperature dependence of the coefficient of thermal expansion in the second heating and cooling cycle for 30Al_2O_3/Mg composite.

Table 1. The residual changes of the specimen length in Mg composites

Composite	Max. temperature [°C]	Residual length change [μm]
Mg – 9 vol.% Al_2O_3 fibres	370	−9
Mg – 30 vol.% Al_2O_3 fibres	370	−27
Mg – 30 vol.% Al_2O_3 fibres	150	−9

are ranging from 16.4 to 20 × 10^{-6} K^{-1}. It is interesting to note that the CTE during heating decreases slowly during the first cycle, whereas it exhibits a very small increase with increasing temperature in the second cycle.

It can be seen from the figures presented that the length reductions of the composite specimens occur after the first cycle of measurements. The values of the irreversible changes of the specimen length are given in Table 1.

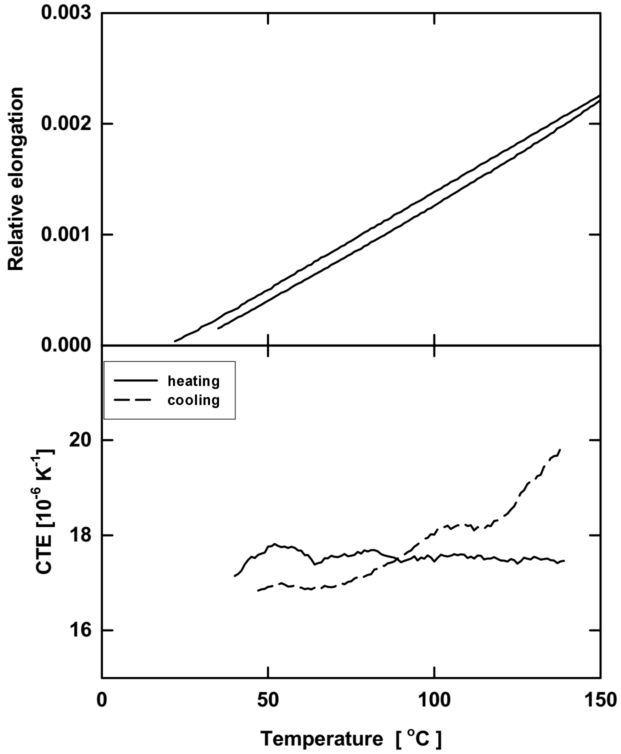

Fig. 5. Thermal linear expansion curve and the temperature dependence of the coefficient of thermal expansion in the first heating and cooling cycle for $30Al_2O_3/Mg$ composite. The peak temperature is 150 °C.

4. Discussion

The curves of relative elongation and the CTE versus temperature for Mg composites depend on the volume fraction of fibres, on heating and cooling stages, on the maximum temperature of the thermal cycle of measurements, and the thermal cycle number. It means that the thermal expansion behaviour is dependent on the thermal history.

As shown in figures and Table 1, the measured thermal linear expansion curves show similar characteristics. It can be seen that after a complete heating and cooling cycle, the composites exhibit residual strain. The value of the residual strain increases with increasing Al_2O_3 fibre volume fraction. Residual strain in the $30Al_2O_3/Mg$ composite increases also with the maximum temperature of the thermal cycle. Similar behaviour was observed by Chmelík et al. [6] who measured the influence of thermal cycling on the relative elongation of Mg-based composites.

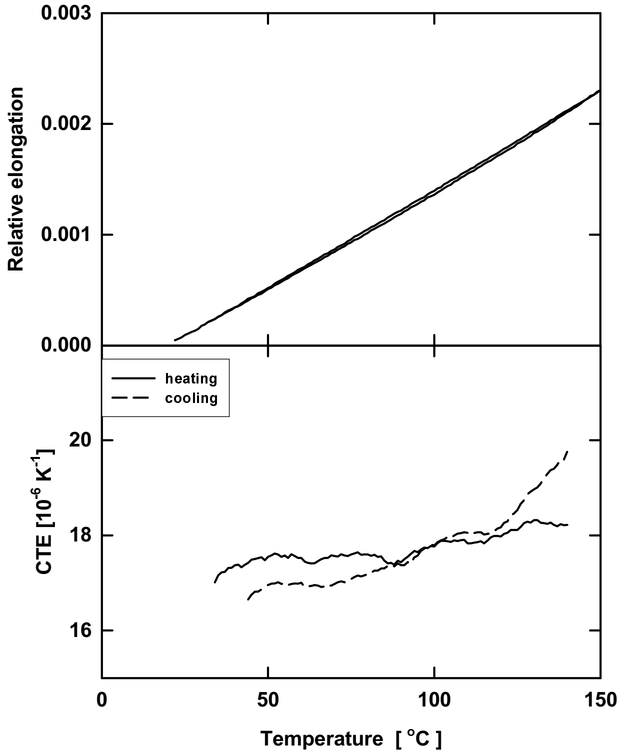

Fig. 6. Thermal linear expansion curve and the temperature dependence of the coefficient of thermal expansion in the second heating and cooling cycle for $30Al_2O_3$/Mg composite. The peak temperature is 150 °C.

The thermal hysteresis behaviour of a composite may be caused by the internal stress release in the composite. The thermal stresses arising in the matrix due to the difference in the CTE values between the reinforcement and the matrix, close to the matrix-fibre interface may be calculated in the simple approximation [11–13] as

$$\sigma_{\mathrm{TS}} = \frac{E_{\mathrm{f}} E_{\mathrm{M}}}{(E_{\mathrm{f}} f + E_{\mathrm{M}}(1-f))} f \Delta\alpha\Delta T, \tag{7}$$

where E_{f} and E_{M} are the Young's moduli of the fibres and the matrix, respectively, f is the volume fraction of fibres and $\Delta\alpha\Delta T$ is the thermal strain. With increasing temperature, if the specimen is heated, the internal thermal stresses at the interface may achieve the yield stress of magnesium. Thus, plastic deformation of the matrix follows at a certain temperature. Above this temperature the CTE should decrease, which is observed. Once the composite had undergone plastic deformation during heating cycle, the produced thermal stress during cooling cannot cause deformation of the matrix back to its original size. Thus, residual strains arise after a complete heating and cooling cycle, which is observed experimentally. The value of the thermal stress in the composite with a higher volume fraction of fibres is higher than that in the composite with lower volume fraction of fibres. Thus, the residual strain in the composite with the higher volume fraction of fibres should be higher, which is observed. The influence of the weak interfacial bonding between reinforcements and matrix on the thermal hysteresis behaviour cannot be excluded. After the first thermal cycle the internal thermal stress generated during preparation of specimens is released. This influences the thermal hysteresis in the second and following complete heating and cooling cycles. The residual strains should be smaller or no residual strain should be observed, which is in agreement with experimental results.

A decrease in the CTE with increasing temperature starting at a certain temperature indicates that plastic deformation occurred when the thermal stresses exceed the yield stress of the matrix. It is known that the yield stress of magnesium (matrix) decreases very rapidly with increasing temperature. A gradual decrease in the CTE for the $30Al_2O_3/Mg$ composite could be caused by small plastic deformation due to creep; the minimum creep rate increases with increasing temperature. The time at which the specimen is held at a certain temperature of measurements is very short and the minimum creep rate at temperatures below 150 ℃ is very slow. It means that the plastic deformation induced is very low, so that its influence on reduction of the CTE at temperatures below 150 ℃ is insignificant.

5. Conclusions

The thermal characteristics of magnesium reinforced with 9 and 30 volume fraction of Al_2O_3 fibres were studied in the temperature range from room temperature to 375 ℃. The thermal linear expansion curves for the Mg composites exhibit the thermal hysteresis behaviour. After the first complete heating and cooling cycle, residual strains are observed. The residual strain increases with increasing volume fraction of the fibres. The residual strain decreases with a reduction of the peak temperature of the thermal cycle. This may be explained assuming the influence of the thermal stresses induced by the difference in coefficients of thermal expansion between the matrix and the fibres.

The CTE values of Mg composites depend on temperature. The temperature variation of the CTE is different for the heating and cooling stages of the thermal cycle. The CTE decreases with increasing volume fraction of Al_2O_3 fibres. A reduction of the CTE with temperature at a certain temperature indicates plastic deformation in the matrix.

The thermal characteristics are dependent on the thermal history.

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REFERENCES

[1] CLYNE, T.—WHITHERS, P. J.: An Introduction to Metal Matrix Composites. Cambridge, Cambridge Press 1993.
[2] DROZD, Z.: Kovove Mater., *40,* 2002, p. 413.
[3] KIEHN, J.—KAINER, K. U.—MORDIKE, B. L.—CHMELÍK, F.— LUKÁČ, P.: In: Magnesium and Their Alloys. Eds.: Mordike, B. L., Kainer, K. U. Frankfurt, Werskstoff-Iformationsgesellschaft 1998, p. 671.
[4] TROJANOVÁ, Z.—KÚDELA, S.—LUKÁČ, P.—DROZD, Z.—MÁTHIS, K.—KOLENČIAK, V.—SCHWEIGHOFER, G.: Kovove Mater., *39,* 2001, p. 1.
[5] TROJANOVÁ, Z.—DROZD, Z.—LUKÁČ, P.—KÚDELA, S.: Mater. Sci. Forum, *419–422,* 2003, p. 817.
[6] CHMELÍK, F.—MOLL, F.—KIEHN, J.—MÁTHIS, K.—LUKÁČ, P.—KAINER, U. K.—LANGDON, T. G.: J. Acoustic Emission, *20,* 2002, p. 108.
[7] TROJANOVÁ, Z.—CHMELÍK, F.—LUKÁČ, P.—RUDAJEVOVÁ, A.: J. Alloy Comp. *339,* 2002, p. 327.
[8] SMAGORINSKI, M. E.—TSANTRIZOS, P. G.: Mater. Sci. Tech., *16,* 2000, p. 853.
[9] BAKER, H.: Physical Properties of Magnesium and Magnesium Alloys. Midland, ML, The Dow Chemical Company 1967.
[10] ASM Specialty Handbook Magnesium and Magnesium Alloys. Eds.: Avedesian, M. M., Baker, H. Materials Park, OH, The Materials Information Society 1999.
[11] CHAWLA, K. K.: In: Materials Science and Technology. Vol. 13. Eds.: Cahn, R. W., Haasen, P., Kramer, E. J. Weinheim, VCH 1993, p. 121.
[12] URRETA, S. E.—SCHALLER, R.—CARENO-MORELLI, E.—GABELLA, E., J. Physique IV, *6,* 1996, p. C8-774.
[13] TAYA, M.: In: Mechanical Properties of Metallic Composites. Ed.: Ochiai, S. New York, Marcel Deker, Inc. 1994, p. 553.

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