Epoxy composites filled with boron nitride and aluminum nitride for improved thermal conductivity*)

John M. Hutchinson[1], **), Frida Román[1], Pilar Cortés[1], Yolanda Calventus[1]

DOI: dx.doi.org/10.14314/polimery.2017.560

Abstract: Epoxy composites containing boron nitride (BN) or aluminum nitride (AlN or Al_2N_3) particles have been studied with a view to obtaining increased thermal conductivity. The effect of these fillers on the cure reaction has been investigated by differential scanning calorimetry (DSC) for two systems, epoxy-diamine and epoxy-thiol, and for volume fractions up to about 35 % of these filler particles. For the epoxy-diamine system, the glass transition temperature of the fully cured system, the heat of reaction, and the temperature at which the peak heat flow occurs were all independent of the cure conditions, filler type and content. In contrast, the epoxy-thiol system shows a systematic effect of filler on the peak temperature for both fillers: there is an initial acceleration of the reaction, which diminishes with increasing content, and the reaction is even significantly retarded at high contents for BN. This is interpreted in terms of an improved interface between epoxy matrix and particles, with a consequent enhancement of the thermal conductivity of the epoxy-thiol composites.

Keywords: epoxy composites, boron nitride, aluminum nitride, thermal conductivity, differential scanning calorimetry.

Kompozyty epoksydowe napełnione azotkiem boru lub azotkiem aluminium o ulepszonej przewodności cieplnej

Streszczenie: Otrzymywano kompozyty epoksydowe napełnione cząstkami azotku boru (BN) lub azotku aluminium (Al_2N_3, AlN) o polepszonej przewodności cieplnej. Metodą różnicowej kalorymetrii skaningowej (DSC) oceniano wpływ dodatku (do 35 % obj.) napełniacza na przebieg reakcji sieciowania za pomocą systemu zawierającego epoksy-diaminę lub epoksy-tiol. W wypadku sieciowania systemem z udziałem epoksy-diaminy temperatura zeszklenia w pełni utwardzonego układu, ciepło reakcji i temperatura odpowiadająca maksimum przepływu ciepła były niezależne od warunków utwardzania, rodzaju i zawartości napełniacza. W wypadku użycia systemu z udziałem epoksy-tiolu zaobserwowano systematyczny wpływ dodatku obu rodzajów napełniacza na temperaturę maksimum przepływu ciepła. Początkowo następowało przyspieszenie reakcji sieciowania, której szybkość zmniejszała się wraz ze wzrostem zawartości napełniacza, a w wypadku dużego udziału cząstek BN reakcja spektakularnie spowalniała. Zjawisko to można interpretować zmniejszoną odległością (poprawą oddziaływań) między cząstkami matrycy epoksydowej i napełniacza, a w konsekwencji polepszoną przewodnością cieplną kompozytów epoksy-tiolowych.

Słowa kluczowe: kompozyty epoksydowe, azotek boru, azotek aluminium, przewodność cieplna, różnicowa kalorymetria skaningowa.

Microelectronic devices are nowadays becoming increasingly integrated and used at higher powers and frequencies, for example in light emitting diodes (LEDs), in highly integrated memory chips and for power conversion applications. Under these conditions, the amount of heat generated is becoming increasingly difficult to dissipate in conventional systems based upon printed circuit board technology, and the resulting temperature increase can induce thermal fatigue and chemical reactions which substantially reduce the service life of the device. There are various rules of thumb to estimate the effect of such an increase in operating temperature, for example that the failure rate of an electronic device doubles with every 10 °C increase [1] or that every increase in operating temperature of 10 % reduces the service life by 50 % [2], but the message is clear: the heat must be removed from these devices. One solution to this problem is to use insulated metal substrates (IMS), in which the printed circuit layer is bonded to a metal substrate by means of a dielec-

---

[1] Escola Superior d'Enginyeries Industrial, Aeroespacial i Audiovisual de Terrassa C/ Colom 11, 08222 Terrassa, Spain.

*) This material was presented at 9th International Conference MoDeSt 2016, 4–8 September 2016, Cracow, Poland.

**) Author for correspondence; e-mail: hutchinson@mmt.upc.edu

tric layer, this last being the key to IMS performance. The most important properties required of the material used for this layer are electrical insulation and high thermal conductivity, which must be combined with processability, affordable cost and substrate compatibility.

The prime candidate material is an epoxy composite with a high content of filler or nanofiller, the high loading being required in order to provide pathways for phonon propagation. The connectivity of these pathways depends on many factors: the shape and size of the filler particles is important, bimodal distributions or similar hybrids appearing to be advantageous in this respect [3–8], and the interface between filler and matrix is crucial, often implying the need for some surface modification, such as the use of silane coupling agents [3, 4, 9–11]. Boron nitride (BN) and aluminum nitride (AlN or Al_2N_3) are the most widely used fillers and, in comparison with conventional printed circuit boards with a thermal conductivity of about 0.2 W/(m · K), commercial epoxy-based composites with these fillers (*e.g.*, Tlam, Denka, Bergquist, AIT) can reach values of 3 to 4 W/(m · K). Nevertheless, much higher values have been reported in the literature {for example, 8 W/(m · K) [7], 11 W/(m · K) [3], 19 W/(m · K) [5], 30 W/(m · K) [12]}, but these often require procedures, such as sonication, the use of solvent dispersion, and surface treatment of the particles, which do not represent ease of processability.

We present here an approach which appears promising, in which the epoxy is cured with a thiol. The procedure used in this work for curing of the epoxy, in which a thermal latent initiator is used, falls within the realm of so-called click chemistry, involving efficient, versatile and selective reactions [13]. The epoxy-thiol system is especially interesting due to the formation of hydroxyl and thioether groups in a single step, which can be further transformed into other polymeric structures [14, 15]. This system is the basis of a broader research project investigating the use of dual-cure procedures, which we believe could be of benefit in enhancing the thermal conductivity by permitting the modification of the particle distribution and particle-matrix interactions in a partially cured first stage, with the full cure being effected subsequently in a second stage of the cure. Furthermore, the Lewis acid-base coordination with the BN or AlN/Al_2N_3 particles is expected to provide an improved matrix-filler interface, and hence to enhance the thermal conductivity. The dual-cure process will be investigated in later work; for the present, the effect of the filler particles in a single cure step is investigated here with respect to both the curing reaction kinetics and the final thermal properties of the cured composite.

EXPERIMENTAL PART

Materials

The epoxy resin was diglycidyl ether of bisphenol-A, DGEBA (Araldite GY 240, Huntsman Advanced Materials, equivalent weight 182 g/eq., density 1.17 g/cm^3), with either a thiol, pentaerythritol tetrakis(3-mercaptopropionate) (Sigma-Aldrich, 488.66 g/mol, density 1.28 g/cm^3), or a polyoxypropylenediamine (Jeffamine D-230, Huntsman), as the cross-linking agent. For cross-linking with the thiol, a latent initiator was used, encapsulated imidazole LC-80 (Technicure). The BN filler, in the form of hexagonal plate-like particles with a thickness of the order of 100 nm, and the Al_2N_3, with a more spherical shape, were provided by Benmayor S.A. as white powders with an average particle size of 6 μm, and were used as received, without any surface treatment. The AlN particles, with an average size of 1.4 μm (obtained from Goodfellow) were also used without any surface treatment. Scanning electron microscopy (SEM, Jeol 5610) pictures of the as-received particles of BN and Al_2N_3 are shown in Fig. 1.

Preparation procedure

The preparation procedure for the composite materials was as follows. The fillers were first added to the epoxy resin, in the proportion *x* required; this ranged from 30 % to 70 %, as a percentage of the total mass of filler plus resin. For the epoxy-thiol system, the thiol was then added in a stoichiometric ratio (approximately 60 : 40 epoxy : thiol by mass), and the initiator was added in a proportion of 2.0 phr. For the epoxy-amine system, the diamine was added in a stoichiometric ratio (approximately 75.2 : 24.8 ep-

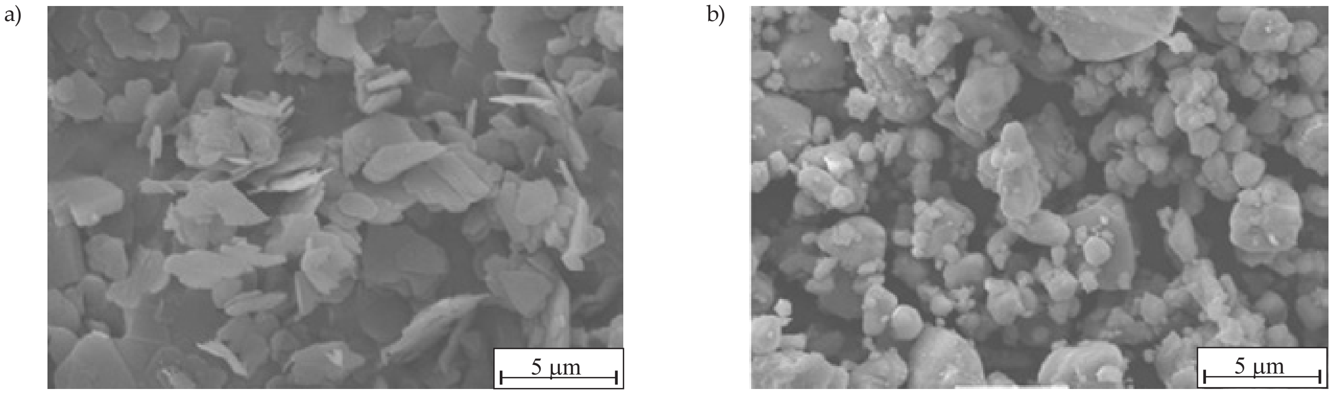

Fig. 1. SEM micrographs of the filler particles: a) BN , b) Al_2N_3, at 5000× magnification; scale bar is 5 μm

oxy : diamine by mass). For both preparations, the whole system was manually mixed for 10 minutes and then degassed under vacuum to less than 26.66 hPa at room temperature for a further 8 minutes. The absence of voids, due to air bubbles, in the cured samples was verified by subsequently examining the fracture surfaces by SEM. As an illustration, an example of a typical fracture surface is shown in Fig. 2 for a sample with 34.2 vol % BN (sample ETLBN60, defined immediately below).

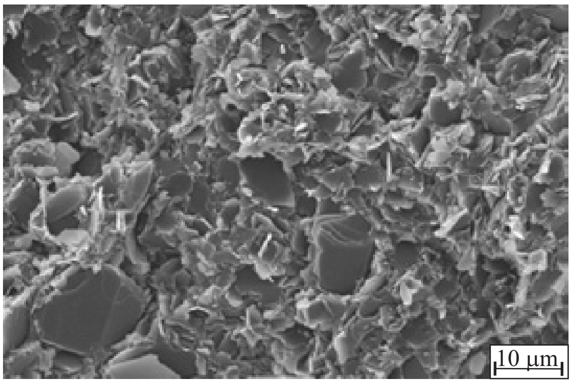

Fig. 2. SEM micrograph of the fracture surface of ETLBN60, with 34.2 vol % BN, at 1500× magnification; scale bar is 10 μm

The content in wt % and vol % of filler in the different epoxy composites is summarized in Table 1, where the following nomenclature is used: ETLBN*x* – epoxy-thiol-LC80-BN; EJBN*x* – epoxy-Jeffamine-BN; ETLAlN*x* – epoxy-thiol-LC80-AlN; EJAlN*x* – epoxy-Jeffamine-AlN; *x* – represents the proportion filler : epoxy + filler.

Methods of testing

Small samples of about 10 mg were taken from this mixture and weighed into aluminum capsules for the calorimetric experiments, while larger quantities were used to fill molds, 10 mm × 40 mm × 4 mm, for curing the samples for the measurement of the thermal conductivity. The differential scanning calorimetry (DSC) experiments were carried out using a conventional DSC (DSC821e, Mettler-Toledo) with a flow of dry nitrogen gas at 50 cm^3/min. Isothermal cure with the LC-80 initiator was made at 60, 70 and 80 °C, and at 50, 60 and 70 °C for the system with Jeffamine, while non-isothermal cure was made at rates of 2, 5 and 10 °C/min for both systems. The thermal conductivity samples for the system with the LC-80 initiator were cured for 1 hour isothermally in an air-circulating oven at 70 °C, whereas those for the system with Jeffamine were first cured isothermally at 70 °C for 6.5 hours and then post-cured at 120 °C for 2 hours.

Table 1. Weight and volume percentages of fillers in the various samples

Sample	wt %	vol %
ETLBN30	20.5	12.9
ETLBN50	37.5	25.7
ETLBN60	47.4	34.2
ETLBN70	58.3	44.7
EJBN30	24.4	14.5
EJBN50	42.9	28.4
EJBN60	53.0	37.3
ETLAlN30	20.5	8.7
ETLAlN50	37.5	18.2
ETLAlN60	47.4	25.1
ETLAlN70	58.3	34.2
EJAlN30	24.4	9.9
EJAlN50	42.9	20.3
EJAlN60	53.0	27.7
EJAlN70	63.7	37.3

The measurements of the thermal conductivity were made using the Transient Hot-Bridge method (Linseis GmbH, THB-100) and a Kapton Hot Point sensor calibrated with poly(methyl methacrylate) (PMMA), borosilicate crown glass, marble, a Ti-Al alloy, and titanium. For each preparation, at least two molded samples were made. The surfaces of the molded samples were polished manually using emery paper (120, 400 and 600 grit size, sequentially), and the sensor was clamped between two flat surfaces of samples from the same preparation. The average of five measurements was taken for each sample.

RESULTS AND DISCUSSION

Cure reaction kinetics

Typical non-isothermal cure curves are shown in Fig. 3 for samples containing BN particles with x = 30, 50, 60 and 70 scanned at 10 °C/min. It is clear that the higher BN contents result in a significant shift of the exothermic peak temperature, T_p, to higher values. There is also a small decrease in T_p for sample ETLBN30, which implies that the reaction is first accelerated and then retarded as the BN content increases; this is more clearly shown in Fig. 4 where, for each filler content, the peak temperature, T_p, relative to that for zero filler content, T_{p0}, is plotted as a function of the vol % filler for three different heating rates.

The cure kinetics are therefore influenced, and in a seemingly complex but systematic way, by the presence of the BN filler. On the other hand, though, the heat of reaction (converted to epoxy equivalent, ee) $\Delta H = 129 \pm 1$ kJ/ee, and the glass transition temperature of the fully cured samples, $T_{g\infty} = 53 \pm 1$ °C, obtained by a second scan at 10 °C/min, are independent of the BN content. Furthermore, this behavior is observed at all the heating rates used, as shown in Fig. 4, while the heat of reaction ΔH and $T_{g\infty}$ are also independent of the heating rate. Thus the reaction kinetics are influenced by the filler content, but the final network structure is not. We conclude that this must be a physical rather than a chemical effect.

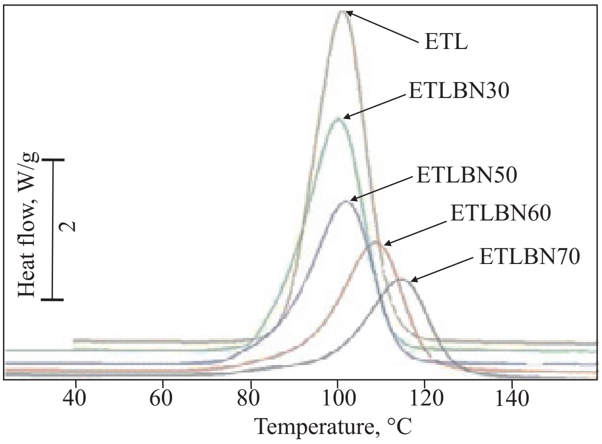

Fig. 3. Non-isothermal DSC scans of samples ETLBN*x* at 10 °C/min; highest peak is for sample ETL without any filler, followed in decreasing height order by samples with 12.9, 25.7, 34.2 and 44.7 vol % BN; exothermic direction is upwards, the heat flow scale is relative, and the curves have been shifted vertically for clarity

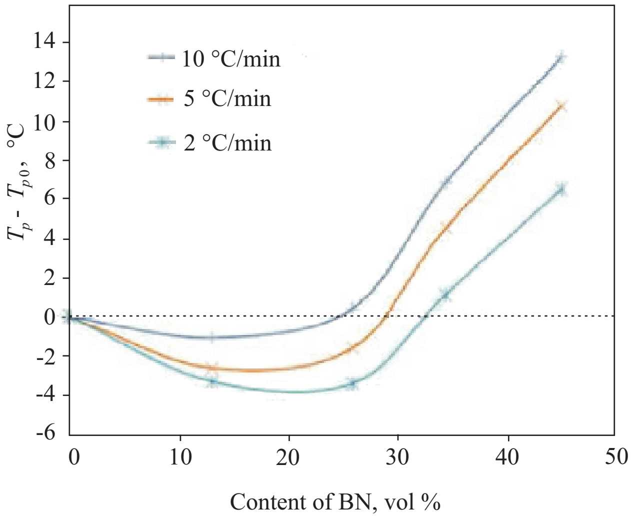

Fig. 4. Exothermic peak temperature, relative to that for zero filler content, during non-isothermal cure of ETLBN*x* samples at the rates indicated; the curves are drawn to guide the eye

A very similar behavior is observed also in the composites containing aluminum nitride, as shown in Fig. 5, where it can be seen that for these AlN composites the initial acceleration of the reaction at low filler contents is more pronounced than in the BN composites.

This acceleration and subsequent retardation of the reaction kinetics is also seen in the isothermal cure of these epoxy composites with both BN and AlN fillers. For example, Fig. 6 shows the time, t_p, to reach the peak exothermic heat flow for the ETLBN*x* system, for which, particularly at the lowest isothermal cure temperature T_c of 60 °C, there is an initial reduction in t_p followed by a gradual increase with increasing BN content. Very similar behavior occurs for the ETLAlN*x* system, the initial reduction again being more pronounced than that for the ETLBN*x* system.

There remains the question of whether these systematic effects of the filler content on the cure kinetics are peculiar to the epoxy-thiol system or whether they would be observed if the epoxy is cross-linked in a different way, for example with a diamine. Accordingly, non-isothermal cure experiments for the epoxy-diamine system with BN (EJBN*x*) and with AlN (EJAlN*x*) were conducted, and the resulting dependence on vol % filler of the peak exotherm temperature relative to that for zero filler content is shown in Fig. 7.

It can be seen that, in striking contrast to the results for the epoxy system cured with thiol, there is no systematic dependence of the peak exotherm temperature on the filler content for either BN or AlN; for each filler in the epoxy system cured with Jeffamine, the cure kinetics are essentially independent of the filler content. There must therefore be some physical interaction between the fillers and the thiol in the matrix that influences the kinetics in this system, and we believe that it is an effect of the

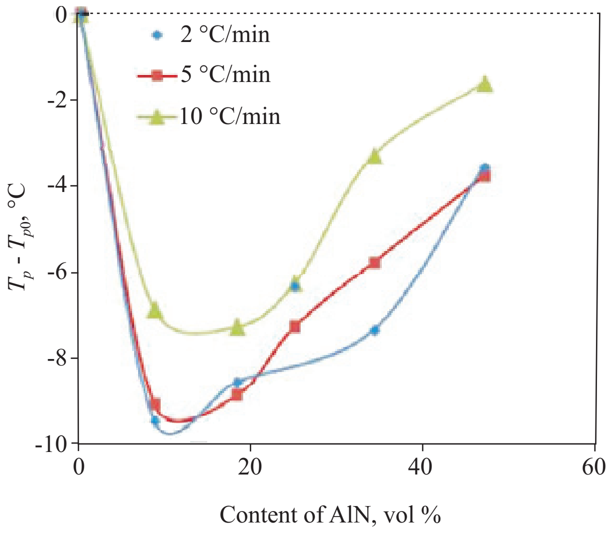

Fig. 5. Exothermic peak temperature, relative to that for zero filler content, during non-isothermal cure of ETLAlN*x* samples at the rates indicated; the curves are drawn to guide the eye

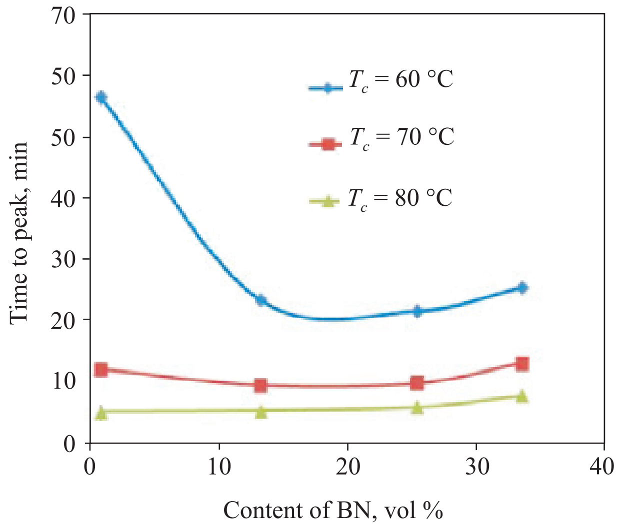

Fig. 6. Time to peak exothermic heat flow during isothermal cure of ETLBN*x* samples at the temperatures indicated; the curves are drawn to guide the eye

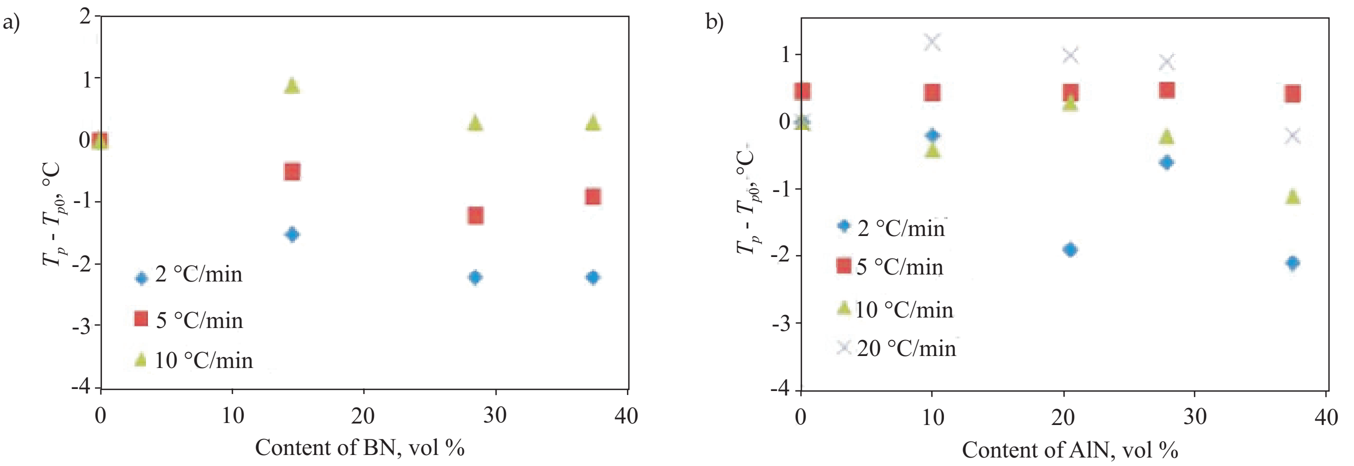

Fig. 7. Exothermic peak temperature, relative to that for zero filler content, during non-isothermal cure of: a) EJBNx samples at the rates indicated, b) EJAlNx samples at the rates indicated

enhanced thermal conductivity resulting from a Lewis acid-base interaction between the thiol and the filler particles, which improves the interface. The argument goes as follows. When the sample is placed into the DSC to monitor the cure kinetics, in an isothermal experiment for example, the sample is initially at a lower temperature (ambient) than the DSC furnace (cure temperature), and requires a certain time to reach the cure temperature. This time is shorter the higher is the thermal conductivity of the sample, so that samples with a certain filler content will reach the cure temperature more quickly than a sample with no filler, and hence will experience an acceleration of the cure with respect to the unfilled sample. On the other hand, when the exothermic cure reaction begins, the heat of reaction must be dissipated in order that the sample should remain at the isothermal cure temperature. This will more readily be achieved the higher is the thermal conductivity of the sample, and hence the unfilled samples, with a lower thermal conductivity, will effectively cure at a higher temperature than the filled samples, and the latter will appear to have a delayed cure kinetics. It is these two opposing effects which give rise to the dependence of the cure kinetics on the filler content in epoxy-thiol samples in Figs. 3 to 6. According to this argument, therefore, we would expect that the epoxy-thiol samples should have a higher thermal conductivity than the epoxy-diamine samples.

Thermal conductivity measurements

The thermal conductivity of many of these samples has been measured by the Transient Hot-Bridge method [16], and the results for the BN-filled samples are shown in Fig. 8.

First, it can be seen that, as anticipated, the thermal conductivities of the epoxy-thiol system are superior to those of the epoxy cured with Jeffamine. This confirms the conclusion, interpreted from the analysis of the cure kinetics by DSC, that there is a better interface between the epoxy matrix and the filler particles in the epoxy-thiol system. Second, the thermal conductivity increases rather rapidly as the BN content increases. The limiting BN content in this study was determined by the workability of the epoxy-filler mixture, which was already a stiff paste at the highest filler contents used here, but extrapolation of the present results suggests that contents higher than 40 vol % would give a significant increase in the thermal conductivity.

In fact, comparing the present results with other values found in the literature for a variety of epoxy-BN composite systems, and included in Fig. 8, it can be seen that the epoxy-thiol system has great potential, comparing favorably with many others. It is noticeable, though, that thermal conductivities achieved by Gaska *et al.* [23] are higher than those obtained here for the same filler content. This may be related to the larger particle sizes used by Gaska

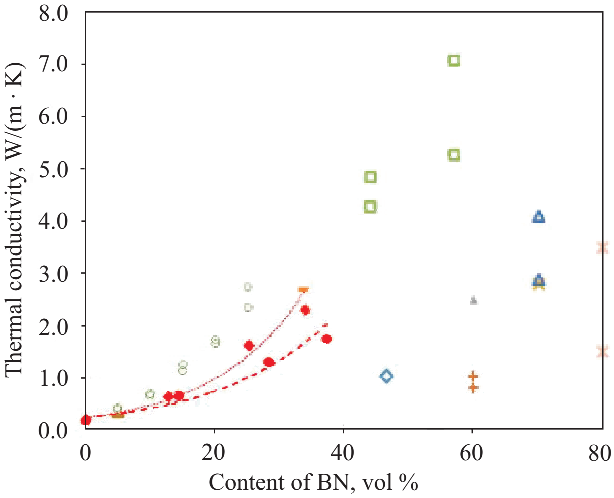

Fig. 8. Thermal conductivity of epoxy composites as a function of vol % BN filler; filled symbols correspond to different systems used in this work: diamonds – LC-80, circles – Jeffamine; the curves are drawn to guide the eye, other symbols from literature: long dash [17], short dash [10], open diamond [18], open squares [19], plus signs [9], open triangles [20], cross [21], asterisks [7], filled triangle [22], open circles [23]

et al., namely 13 μm and 25 μm, the latter being in the form of spherical agglomerates. On the other hand, it is likely that a further improvement in the thermal conductivity could be achieved by appropriate surface treatment of the particles, such as that used by Hong *et al.* [19] to obtain the results indicated by the open squares in Fig. 8. The effects of particle agglomeration and surface treatment are both currently being investigated by ourselves.

For the system filled with aluminum nitride, thermal conductivity results are available only for the samples prepared with the LC-80 initiator, and are shown in Fig. 9.

For comparison, a number of values are included from the literature, for which several different sample preparations are given. For example, Choi and Kim [8] prepared hybrids of AlN and Al_2O_3 particles of different sizes and in different proportions; we include here their maximum values, found for a 7 : 3 volume ratio of large : small particles, with either 10 μm AlN and 0.5 μm Al_2O_3 (squares) or 0.1 μm AlN and 10 μm Al_2O_3 (triangles). On the other hand, Xu *et al.* [3] used a range of AlN particle sizes in their work: 1.5 μm (asterisks), 2 μm (plus sign), 4 μm (short dash), 7 μm (circles), 56 μm (long dash) and 115 μm (diamond). There was no systematic dependence of the thermal conductivity on the particle size, the systems with 7 and 115 μm displaying the highest conductivities. The results for the present work, indicated in Fig. 9 by red diamonds and with a trend shown by the dashed curve, are very similar to those for the BN-filled composites in Fig. 8 in respect of the effect of the volume percentage of filler; the filler content is low, but the trend suggests that the system cured with LC-80 would present high thermal conductivities at higher volume fractions of filler, and in this respect the use of the epoxy-thiol system appears to be advantageous.

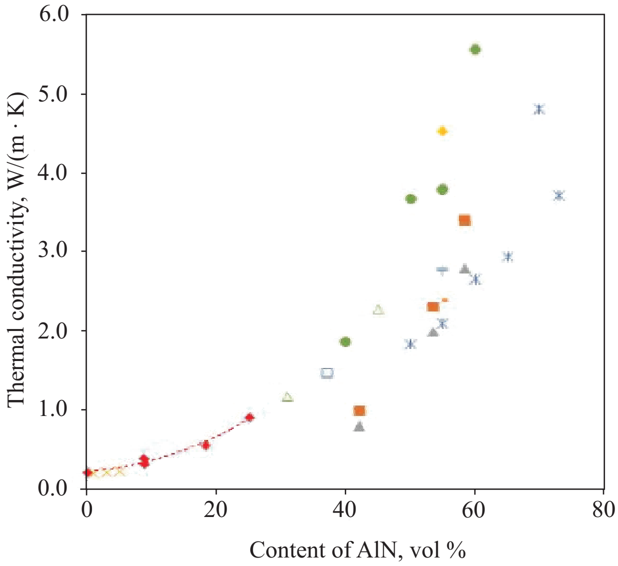

Fig. 9. Thermal conductivity of epoxy composites as a function of vol % AlN filler; symbols with dashed curve, drawn to guide the eye, correspond to ETLAlN*x* system used in this work, other symbols from literature: filled squares, triangles [8], crosses [24], asterisks, circles, plus sign, short dash, long dash, diamond [3], open square [25], open triangles [26]

The preparation of samples with higher volume fractions of filler than those used here, which were up to a maximum of about 40 vol %, presents some difficulties in respect of the workability of the mixtures. The possibility of a dual-cure procedure with these epoxy-thiol systems, however, as indicated in the earlier text, could provide the means for overcoming these difficulties by allowing the mechanical manipulation of the partially cured sample before curing it completely in a second stage. This is currently work in progress, and will be reported later.

CONCLUSIONS

Epoxy composites with boron nitride and aluminum nitride fillers for improved thermal conductivity have been investigated. Two different epoxy systems have been studied: epoxy-thiol with an initiator, and epoxy-diamine. The effect of the fillers on the cure kinetics is observed to be very different for these two systems. In particular, for the epoxy-thiol system the cross-linking reaction is first advanced and then retarded as the filler content increases, whereas there is no noticeable effect of the filler content on the cross-linking reaction kinetics in the epoxy-diamine system. The same behavior is found for both boron nitride and aluminum nitride fillers. Furthermore, the different reaction kinetics for the various filler contents does not have any effect on the cured epoxy network structure, as identified by constant values for the heat of reaction and the glass transition temperature of the fully cured system. This difference in the reaction kinetics is accordingly attributed to the enhanced thermal conductivity in the epoxy-thiol system, which is considered to result from an improved interface between filler particles and matrix as a consequence of a Lewis acid-base interaction. This interpretation is confirmed by measurements of the thermal conductivity, which increases with increasing boron nitride filler content, the increase being significantly greater for the epoxy-thiol system in comparison with the epoxy-diamine system. In fact, the epoxy-thiol system with the boron nitride filler shows great potential for enhanced thermal conductivity when compared with many other results in the literature.

ACKNOWLEDGMENTS

The authors are grateful to ACCI Specialty Materials (Linden, NJ, USA) for the provision of the Technicure initiator, LC-80, and to Benmayor S.A. (Barcelona, Spain) for the BN and Al_2N_3 *particles. This work was financed by the Spanish Ministerio de Economia y Competitividad, project MAT2014-53706-C3-3-R.*

REFERENCES

[1] http://www.aitechnology.com/products/insulated-metal-substrates/thermclads/ (access date 06.10.2016).

[2] http://www.mos-electronic.com/en/downloads/english/Technology_en_07_2016.pdf (access date 06.10.2016).

[3] Xu Y.S., Chung D.D.L., Mroz C.: *Composites Part A: Applied Science and Manufacturing* **2001**, *32*, 1749. http://dx.doi.org/10.1016/S1359-835X(01)00023-9

[4] Lee G-W., Park M., Kim J. *et al.*: *Composites Part A: Applied Science and Manufacturing* **2006**, *37*, 727. http://dx.doi.org/10.1016/j.compositesa.2005.07.006

[5] Yung K.C., Liem H.: *Journal of Applied Polymer Science* **2007**, *106*, 3587. http://dx.doi.org/10.1002/app.27027

[6] Shimazaki Y., Hojo F., Takezawa Y.: *Applied Physics Letters* **2008**, *92*, 133 309. http://dx.doi.org/10.1063/1.2907315

[7] Hong J-P., Yoon S-W., Hwang T. *et al.*: *Thermochimica Acta* **2012**, *537*, 70. http://dx.doi.org/10.1016/j.tca.2012.03.002

[8] Choi S., Kim J.: *Composites Part B: Engineering* **2013**, *51*, 140. http://dx.doi.org/10.1016/j.compositesb.2013.03.002

[9] Xu Y., Chung D.D.L.: *Composite Interfaces* **2000**, *7*, 243. http://dx.doi.org/10.1163/156855400750244969

[10] Wattanakul K., Manuspiya H., Yanumet N.: *Journal of Applied Polymer Science* **2011**, *119*, 3234. http://dx.doi.org/10.1002/app.32889

[11] Ji T., Zhang L.Q., Wang W.C. *et al.*: *Polymer Composites* **2012**, *33*, 1473. http://dx.doi.org/10.1002/pc.22277

[12] Song W-L., Wang P., Cao L. *et al.*: *Angewandte Chemie International Edition* **2012**, *51*, 6498. http://dx.doi.org/10.1002/anie.201201689

[13] Moses J.E., Moorhouse A.D.: *Chemical Society Reviews* **2007**, *36*, 1249. http://dx.doi.org/10.1039/B613014N

[14] Flores M., Tomuta A.M., Fernández-Francos X. *et al.*: *Polymer* **2013**, *54*, 5473. http://dx.doi.org/10.1016/j.polymer.2013.07.056

[15] Carlborg C.F., Vastesson A., Liu Y. *et al.*: *Journal of Polymer Science Part A: Polymer Chemistry* **2014**, *52*, 2604. http://dx.doi.org/10.1002/pola.27276

[16] Hammerschmidt U., Meier V.: *International Journal of Thermophysics* **2006**, *27*, 840. http://dx.doi.org/10.1007/s10765-006-0061-2

[17] Yu J., Huang X., Wu C. *et al.*: *Polymer* **2012**, *53*, 471. http://dx.doi.org/10.1016/j.polymer.2011.12.040

[18] Gu J., Zhang Q., Dang J., Xie C.: *Polymers for Advanced Technologies* **2012**, *23*, 1025. http://dx.doi.org/10.1002/pat.2063

[19] Hong J-P., Yoon, S-W., Hwang T-S. *et al.*: *Korea-Australia Rheology Journal* **2010**, *22*, 259.

[20] Kim K., Kim M., Hwang Y., Kim J.: *Ceramics International* **2014**, *40*, 2047. http://dx.doi.org/10.1016/j.ceramint.2013.07.117

[21] Kim K., Kim J.: *Ceramics International* **2014**, *40*, 5181. http://dx.doi.org/10.1016/j.ceramint.2013.10.076

[22] Wang Z., Iizuka T., Kozako M. *et al.*: *IEEE Transactions on Dielectrics and Electrical Insulation* **2011**, *18*, 1963. http://dx.doi.org/10.1109/TDEI.2011.6118634

[23] Gaska K., Rybak A., Kapusta C. *et al.*: *Polymers for Advanced Technologies* **2015**, *26*, 26. http://dx.doi.org/10.1002/pat.3414

[24] Duwe S., Arlt C., Aranda S. *et al.*: *Composites Science and Technology* **2012**, *72*, 1324. http://dx.doi.org/10.1016/j.compscitech.2012.04.015

[25] Zhu B.L., Wang J., Ma J. *et al.*: *Journal of Applied Polymer Science* **2012**, *127*, 3456. http://dx.doi.org/10.1002/app.37599

[26] Rybak A., Gaska K.: *Journal of Materials Science* **2015**, *50*, 7779. http://dx.doi.org/10.1007/s10853-015-9349-6

Received 12 XII 2016.