FABRICATION OF HIGH THERMAL CONDUCTIVITY CERAMIC HYBRID MATERIALS FOR POWER ELECTRONICS AND INTEGRATED PACKAGES

Lee S.T.[1], Kim C.H.[1,2], Nahm S.[2], Lee S. H.[3], Lee S.G.[3], and Kim H.T.[1]*
*Author for correspondence
[1]Korea Institute of Ceramic Engineering and Technology, Jinju-si, Gyeongsangnam-do 660-031
[2]Korea University, Department of Materials Science and Technology, Seoul 136-701
[3]Gyeongsang National University, Department of Materials Science and Technology, Jinju-si 660-701
Republic of Korea
E-mail: hytek@kicet.re.kr

ABSTRACT

High thermal conductivity with electrically insulating substrates and thermal interface materials are in a great demand for high density and high power electronic packaging applications. The choice of ceramic-polymer hybrid films provide many advantage in electrical and thermal behaviour due its intrinsic nature of high thermal conductivity and high breakdown voltage, environmental and thermal stability. This work present the preparation hybrid films with mechanically exfoliated h-BN ceramic sheets and PVA polymer, and the effect of exfoliation, solid loading, particle sizes, compression pressures on the composite films were investigated. And the results are compared with the theoretical models like Arithmetic and Wiener equations that frequently used in the two phase mixture composites.

INTRODUCTION

The demand for high thermal conductivity substrates with insulating materials are increasing with the emerging markets in power electronic packages for EV/HEV, in application processor chips so called AP for mobile applications such as smart phones and tablet PCs, and in high power LED packages. High thermally conductive as well as electrically insulating dielectric films on rigid substrate can be fabricated by using conventional DBC (direct bonded copper) or DBA(direct bonded aluminium) process that uses a thin (about 0.2mm) sintered alumina (Al_2O_3) or aluminium nitride (AlN) substrate bonded with copper or aluminium heat spreader by poster heat process. Recently authors demonstrated a room temperature direct deposition of AlN thin film on aluminium heat spread (heat sink) and fabricated an LED array module, which resulted in great enhancement in the heat transfer properties so that the thermal resistance of the module was decreased almost a half of the conventionally fabricated FR-4 based PCB [1]. More recently we have succeeded in the fabrication of ceramic/metal hybrid films that uses AlN and aluminium (Al) metal particles with high thermal conductivity and electrical insulation by using the same aerosol deposition method (ADM). These techniques have great potential to fabricate a chip on board (CoB) or a chip on heat sink (CoH) type highly efficient heat transfer package modules [2].

On the other hand, ceramic/polymer hybrids (or composites) that can be used as insulating dielectric films or thermal interface materials (TIM) such as thermal pads, pastes, films etc. are also draws a great attention to both rigid and flexible electronics application. There were several works on polymer/ceramic hybrid films using highly thermal conductive ceramics such as AlN, boron nitride (BN), and alumina powders mixed with PVA, PMMA and epoxy materials [3-13]. Among them BN ceramics is one of the most attractive candidate filler materials as the BN have excellent electrical, environmental and thermal properties. Moreover, h-BN powder exhibits an un-isothermal property due to its platelet morphology of powders.

In this paper, we present BN/polymer composite films prepared by using mechanical exfoliation of h-BN particles and PVA. The effects of an exfoliation, BN particle size, and uniaxial compression on the microstructure and thermal conductivity of the composite film are investigated. Also, theoretically calculated thermal conductivities of the films are compared with those of measured samples.

NOMENCLATURE

c		proportion of component 1
1-c		proportion of component 2

Special characters

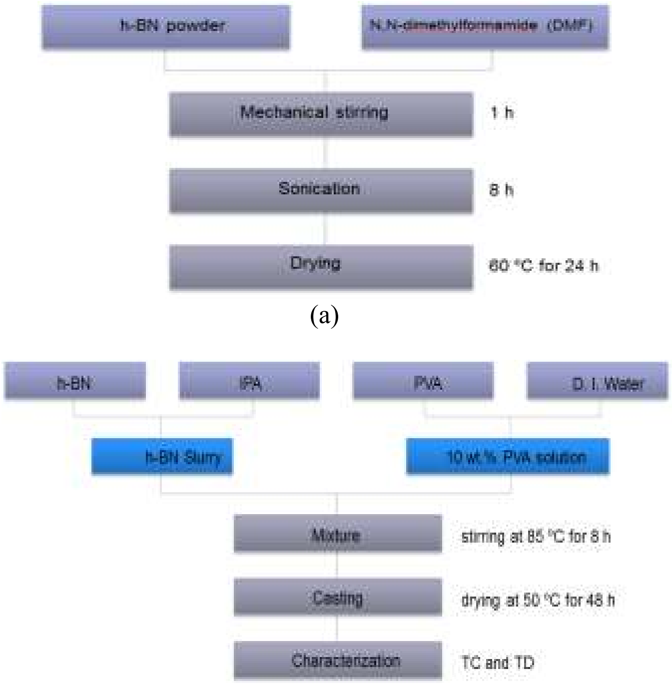
α		Constant of Wiener model (dispersion model α=0.5)
λ	[W/m·K]	Overall thermal conductivity
λ_1	[W/m·K]	Thermal conductivity of component 1 (=0.2)
λ_2	[Wm·K]	Thermal conductivity of component 2 (=33)

EXPERIMENTAL

An experimental procedure of the preparation of mechanical exfoliation of h-BN particles are represented in Figure 1 (a) and in the previous works [7-9, 14]. As a result an as-received h-BN particles with particles sizing 5um (D_{50}) in diameter and 5~10nm in thickness was thinned into 0.5~3nm with similar diameters.

The h-BN/PVA hybrid films are prepared in three steps: i) preparation of BN/solvent mixture slurry, ii) dissolving PVA powders in a de-ionized water, and mechanical stirring followed by sonication to homogenized the h-BN/PVA mixtures as presented in Fig. 1 (b). Then the mixtures are casted on petri dish or releasing agent coated PET film and dried for 48 hours at 50°C.

The microstructures of fabricated films are examined by FE-SEM. The compression temperature of the h-BN/PVA films was determined by measuring glass transition temperature (Tg) using differential scanning calorimeter (DSC). The compression was done at 90°C for 30 minutes under 100MPa using uniaxial press with heating module.



(a)

(b)

Figure 1 Experimental procedure of (a) mechanical exfoliation process and (b) fabrication of h-BN/PVA composites

The evaluation of thermal conductivities are measured in a two manners, a transvers mode (z-direction, i.e. thickness direction) thermal conductivity that is mostly used method and an in-plane mode (x-y direction of the film) thermal conductivity as illustrated in Fig. 2, as a single h-BN particle has an un-isothermal conducting behaver due to platelet like morphology.

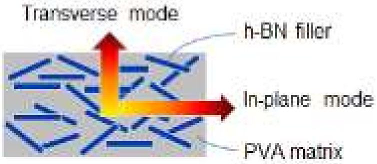


Figure 2 Illustration of transvers mode and in-plane mode thermal conductivities measurement scheme in h-BN/PVA composites film

RESULTS AND DISCUSSION

The transverse mode thermal conductivities of h-BN/PVA composites with and without exfoliation of h-Bn particles are plotted in Fig. 3 with the amount of h-BN loading. The result indicates an improvement in the thermal transport by using exfoliated h-BN powders in the composite film. The increase in the thermal conductivity is almost doubled when the h-BNs are loaded over 40 vol%. The effect of h-BN particle size was also investigated using two mean particle sizes, D_{50} = 1.5um and 5um. A noticeable increase at the composite films with 1.5um h-BN particles was observed in 20~40 vol% h-BN contents however a little difference was observed in low contents under 20vol% and higher contents over 40 vol%.

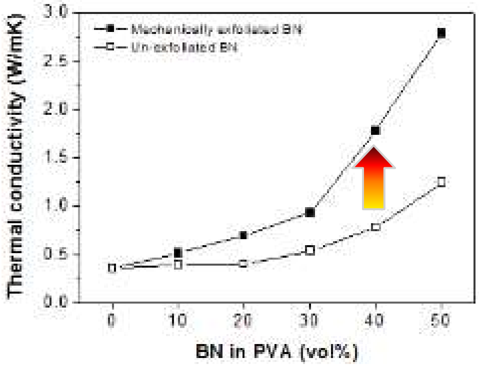


Figure 3 Effect of exfoliation in the h-BN/PVA composites

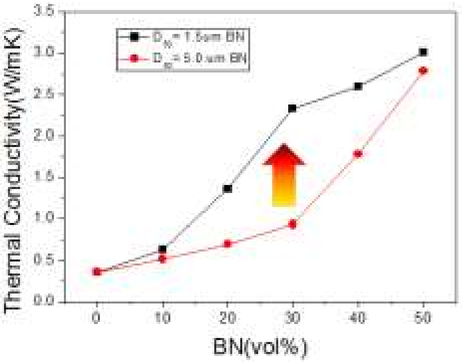


Figure 4 Effect of particle sizes on the thermal conductivity of h-BN/PVA composites

The previous other authors work [8] gave a lesson that the mechanical stretching enhanced the thermal conductivities of in-plane mode but did not demonstrated transverse mode values for the same sample, so we tried to apply an uniaxial compression on the film to get an improvement in thermal property and see how the transverse mode as well as the in-plane mode thermal conductivities are changed with thermal compression in uniaxial direction. The thermal compression condition was adjusted based on the DSC result that the compression temperature was set at 90°C which is above the Tg of PVA polymer, where the PVA is a rubbery state to aid a flexibility for h-BN particles rearrangement in the film. The following FE-SEM photos provide the microstructural difference between the un-compressed and compressed h-BN/PVA composite films in a cross-sectional view. Quite lots of h-BN platelet particles are settled in an insufficient alignment state to the in-plane direction at the un-compressed samples, while those in the compressed samples are well aligned with respect to the in-plane direction.

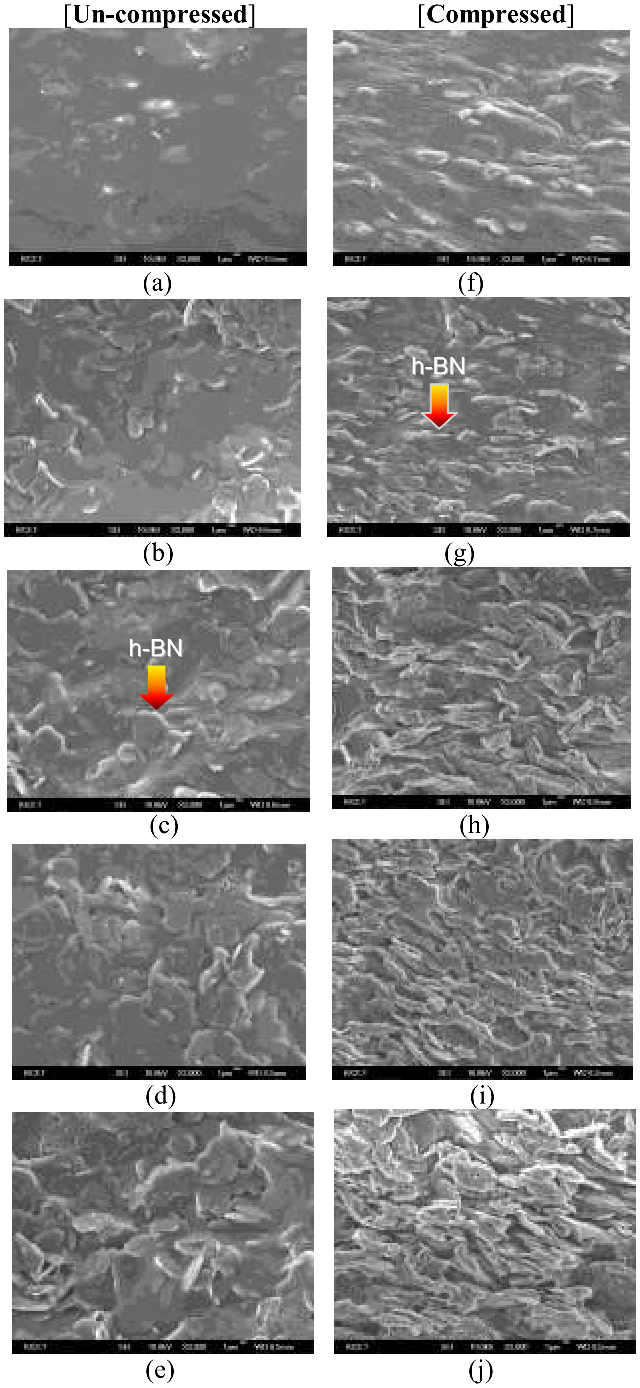


Figure 4 Cross-sectional views of h-BN/PVA composite films observed by FE-SEM: (a) and (f) 10vol% BN; (b) and (g) 20vol% BN; (c) and (h) 30 vol% BN; (d) and (i) 40 vol% BN; (e) and (j) 50 vol% BN

The transvers and in-plane mode thermal conductivities for un-compressed and compressed films are measured and the results are plotted in Fig. 5. Also, we calculated the theoretical values of the thermal conductivities by using arithmetic and Wiener model which is frequently used in a two phase composite system as described in the following equations:

- Arithmetic model:

$$\lambda = c\lambda_1 + (1-c)\lambda_2 \quad \text{--------------------------------} \ (1)$$

where, c = proportion of component 1
1-c = proportion of component 2
λ_1, λ_2 = thermal conductivity of component 1, 2

- Wiener model:

$$\lambda/\lambda_2 = (1-c((1-\lambda_1/\lambda_2) / (1+\alpha\ \lambda_1/\lambda_2))) / (1+\alpha c((1-\lambda_1/\lambda_2) / (1+\alpha\ \lambda_1/\lambda_2))) \quad \text{-------------------------------} \ (2)$$

where, $\alpha = 0.5$ for dispersion model, $\lambda_1 < \lambda_2$

- Raw materials data:

$\lambda_1 = 0.2$ W/m·K (PVA)

$\lambda_2 = 33$ W/m·K (BN, directional average)

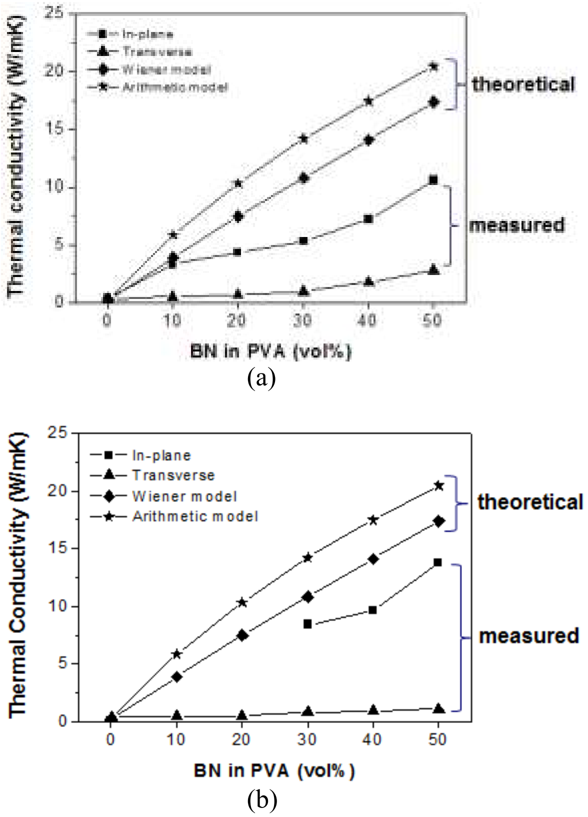


Figure 5 Effect of uniaxial compression on the transverse and in-plane mode thermal conductivities of BN/PVA composite films: (a) un-compressed and (b) compressed sample

The in-plane mode thermal conductivities exhibited an almost 5 times in un-compressed and 10 times higher values in compressed films compared to the transvers mode thermal conductivities. The maximum in-plane thermal conductivity obtained at 50 vol% h-BN was reached to over 13 W/m·K. However, the transverse thermal conductivities of compressed films were decreased after thermal compression in uniaxial direction. The reason is explained in elsewhere [14] that is caused by an increased disconnectivity of h-BN platelet particles to the compressed direction due to h-BN particles re-arrangement (alignment) to the in-plane direction. In case of compressed films, the in-plane mode thermal conductivities are pretty much close to the Wiener model calculated values. It was also found that the theoretically calculated thermal conductivities are far above the measured values that imply a further enhancement of thermal conductivity may accomplish if an additional treatments such as filler particle coating or modification of polymers are optimized.

CONCLUSION

Ceramic/polymer hybrid films containing mechanically exfoliated h-BN nanosheets and PVA polymer are fabricated, and the effect of exfoliation, solid loading, particle sizes, thermal compression with uniaxial press on the composite films were investigated. The application of exfoliated h-BN particles enhanced the thermal conductivity of the film, and the values increased in accordance to the loaded h-BN contents. The SEM microstructures of compressed films exhibited an improved alignment of h-BN platelet particles along with the in-plane direction of film, which helped the thermal conduction path through the h-BN particles longitudinal direction. However, the compressed samples resulted in decreased transverse mode thermal conductivities. Though the measured thermal conductivities are quite different form the calculated ones via arithmetic and wiener model, the in-plane mode thermal conductivities are a little bit close to the Wiener model calculated values in the compressed films.

REFERENCES

[1] Heo Y.J., Kim H.T., Kim K.J., Nahm S., Yoon Y.J., and Kim J.H., Enhanced heat transfer by room temperature deposition of AlN film on aluminum for a light emitting diode package, Applied Thermal Engineering, 50 (2013), pp. 799-804

[2] Heo Y.J., Nahm, and Kim H.T., Room temperature direct deposition of high thermal diffusivity ceramic-metal hybrid films for high power electronic packages, to be submitted to the Advanced Materials (2015)

[3] H. E-Dehaghani and M. Nazempour, Thermal conductivity of nanoparticles filled polymers, Smart Nanoparticles Technology, edited by Abbass Hashim, ISBN: 978-953-51-0050-8 (In Tech, 2012), pp.520-540.

[4] S. Kemaloglu, G. Ozkoc, and A. Aytac, Thermally conductive boron nitride/SEBS/EVA ternary composites: “Processing and Characterization”, Polymer Composites, 1398 (2010).

[5] J. P. Hong, S. W. Yoon, T. S. Hwang, Y. K. Lee. S. H. Won, and J. D. Nam, Interface control of boron nitride/epoxy composites for high thermal conductivity, Korea-Australia Rheology Journal, 22 (4) 259 (2010).

[6] Y. Xu and D. D. L. Chung, Increasing the thermal conductivity of boron nitride and aluminum nitride particle epoxy-matrix composites by particle surface treatment, Composites Interfaces, 7, 243 (2000).

[7] X. Wang, A. Pakdel, J. Zhang, Q. Weng, T. Zhai, C. Zhi, D. Golberg, and Y. Bando, Large-surface area BN nanosheets and their utilization in polymeric composites with improved thermal and dielectric properties, Nanoscale Research Letters, **7**, 662 (2012).

[8] W. L. Song. P. Wang, L. Cao, A. Anderson, M. J. Mezani, A. J. Farr, and Y. P. Sun, Polymer/boron nitride nanocomposite materials for superior thermal transport performance, Angew. Chem. Int. ed., 51, 6498 (2012).

[9] Z. Lin, A. Mcnamara, Y. Liu, K. S. Moon, and C. P. Wong, Exfoliated hexagonal boron nitride polymer nanocomposite with enhanced thermal conductivity for electronic encapsulation, Composites Science and technology, **90**, 123 (2014).

[10] Nam K.M., Lee Y.J., Kim B.Y., Kwon W.T., Kim S.R., Shin D.G., and Kim Y.H., Biomimetic preparation of boron nitride/PMMA composite, Journal of the Korean Ceramic Society, vol.51, No. 2, 2014, pp.103-106

[11] Liu F., Mo X., Gan H., Guo T., Wang X., Chen B., Deng S., Xu N., Sekiguchi T., and Bando Y., Cheap, gram-scale fabrication of BN nanosheets via substitution reaction of graphite powders and their use for mechanical reinforcement of polymers, Scientific Reports, 4: 4211 (2014)

[12] Hong J., Yoon S., Hwang T., Oh J., Lee Y., Nam J., Effect of filler size and its bimodal distribution for highly thermal-conductive epoxy composites, Proceedings of 18th International conference on composite materials,

[13] Egan E. and Amon C.H, The effect of filler type and volume fraction on the thermal conductivity of several polymer composites, Carnegie Mellon University Research Showcase @ CMU, 1996

[14] Lee S.T., Kim H.T., Kim C.H., and Nahm S., High thermal conductivity ceramic/polymer nano-composite films using boron nitride, submitted to the Journal of Vacuum Science and Technology B, March 2015