

สมบัติเชิงกล สมบัติทางความร้อน และสมบัติทางไทรโบโลยี ของวัสดุอิพอกซีเรซิน เสริมแรงด้วยอนุภาคอะลูมิเนียม เพื่อช่วยส่งเสริมอายุการใช้งานวัสดุแม่พิมพ์ สำหรับผู้ผลิตแม่พิมพ์รวดเร็ว: การทบทวนวรรณกรรม Mechanical, Thermal and Tribological Properties of Aluminium-filled Epoxy Composites to Enhance Lifetime of Mould Material for Rapid Manufacturer: A Review

Peerawatt Nunthavarawong

Introduction

The fabrication of both injection and compression moulds are normally made by cold-working tool steels, aluminium alloys, etc. These materials are costly, and their fabrication processes are time consuming. In order to reduce a lead time spent in producing the prototype, the composite tooling can be fabricated by one of many commercially available rapid tooling techniques. Rapid tooling or rapid mould is referred to as the indirect tooling method that can quickly produce the mould-inserts. It is considered as a good alternative to producing moulds for short-run production or complicated parts, which are often not worth the construction of a mould from a bulk metal via machining. Several processes can be utilised in fabrication of the rapid tooling, such as rapid prototyping, casting, thermal spraying of zinc-based coating, etc. These processes are commonly used to fabricate a mould for light-usage parts, such as silicone, epoxy resin, which are widely used in plastic and rubber forming [1, 3, 5, 6, 9, 11, 12]. However, the lifetime of the mould material produced by the aluminium-filled epoxy composites (AL-EP) is much shorter than that produced from steel [2, 8, 9]. The tool life of AL-EP depends on several factors, including mechanical strength, thermal property and wear of material itself, etc [2, 6-10]. Therefore, in this review, the general developments of material properties of the AL-EP material for guiding the rapid mould maker are proposed to enhance the lifespan.

Mechanical properties

The mechanical properties of epoxies and their composites are of great interest in a wide range of technological applications, such as in electronic & packaging, aerospace automotive food and industries, including civil construction [2, 3, 5-9]. Most studies to date have focused on the accumulation of data on mechanical properties of AL-EP material, in particular the strength and moduli at ambient well elevated as as temperatures. AL-EP material is one of several types of the particle-reinforced polymer composites [2, 6, 10], as shown in Fig.1 [4]. To achieve the optimum efficiency of this material, the effects of mass mixture ratio on its mechanical strength was investigated. Related studies of the AL-EP properties have, however, been slightly proposed.

Lecturer, Department of Teacher Training in Mechanical Engineering, Faculty of Technical Education, King Mongkut's University of Technology North Bangkok and Industrial Tribo-Systems & Design Centre (TRIBOSYSLab), Tel. 0-2555-2000 Ext. 3279, E-mail: peerawattn@kmutnb.ac.th

In 2002. Cheah et al used mixing ratio of 25% for epoxy and 75% for aluminium powder (Manufactured by MCP). However, the mixing ratio depends upon the size of particulate reinforcement, due to large particles being separated by epoxy resin [5]. Lui and Gong studied mechanical properties of Al-filled polyethylene (AL-PE) and AL-EP under compression load. They found that the compressive modulus of AL-EP is greater than AL-PE material [7]. In 2003, Gonvanes et al proposed a compression test for AL-EP material. They used AI powder with the grain size ranging from 106-125 um to produce compressive specimens with the content of aluminium powder of 20%, 25% and 30 vol.%. They found the highest compressive strength occurs in the workpiece having the aluminium fraction of 30 vol.% [8]. Chung et al studied the dimensional accuracy by comparing the effect of the shrinkage rate of specimens. They found the AL-EP material has better accuracy than the neat resin. Moreover, they also reported a higher content of aluminium powder can provide higher tensile strength and thermal conductivity of the soft tools to improve [2, 9]. By evaluating the content of the filler, 40 vol.% is the optimum value for of soft tooling application [9]. Chungchoo et al use epoxy resin produced by SIKATM, and the aluminium powder prepared by grinding the aluminium ingot in which the grain size varies from sub-microns to 63 um. By

using this method, the shape of aluminium powder can be considered as irregular. In order to obtain the minimum amount of air bubbles formed inside the specimens during curing process, all mixing and casting processes were carried out in a vacuum chamber with a suitable decassing time of 30-45 min. Then, the specimen was cooled in this chamber for 16 to 24 hours, during which a mechanical property study was performed. The tensile and compressive strength are shown in Tables 1 and 2, respectively. Their results obtained by tensile test show that the mass mixture ratio between epoxy and aluminium powder of 55:45 gives the highest tensile strength of around 50 MPa. When the content of aluminium filler is greater than epoxy resin, the tensile strength decreases to about 25 MPa. The compressive strength for all mixture ratios is moderately similar at about 85 MPa. A higher content of aluminium powder aives the higher compressive strength, but tensile property was decreased due to a weakness in interfacial bonding between an epoxy matrix and a reinforced Al particle (cf. Table 1). While the Young's modulus of the AL-EP as a function of tested temperature is shown in Fig. 2 for mixing ratio of 55:45 composition. Fig. 2 shows that the modulus was increased when the temperature was decreased and vice versa [6].



Fig. 1 Composite materials [4]



This is often due to the effects of thermal softening in the AL-EP material [17]. Hence, the thermal properties of this material should be examined.



Fig. 2 Young's modulus vs. Temperature [6]

Thermal properties

All amorphous polymer materials reveal a glass transition temperature (Tg) [14]. As a polymer is thermal, it involves a discontinuous change in a thermodynamic quantity in terms of the heat capacity or the coefficient of thermal expansion, CTE (*cf.* Table 3) [13, 14]. In reference to the effects of temperature, thermal softening occurred in polymers where a polymeric contact approaches a temperature close to the Tg or crystalline melting temperature, there is an illustrated decrease in the elastic modulus [2, 14, 17]. Besides, the thermal conductivity (K) of a polymer and polymer-based composites is an essential property for several applications in industries [2, 6, 15].

Aluminium Contents	Ultimate Tensile	Elongation	Young's Modulus	
(by mass %)	Strength (MPa)	at break (%)	(MPa)	
0	18.44	1.17	2,157.00	
40	13.42	0.72	3,370.23	
45	49.99	3.50	3,334.03	
50	25.07	1.14	3,881.63	
55	24.44	1.55	3,855.29	

Table 1 Tensile strength of AL-EP materials [6]

 Table 2 Compressive strength of AL-EP materials [6]

Aluminium Contents	Ultimate Compressive Shrinkage		Young's Modulus	
(by mass %)	Strength (MPa)	at break (%)	(MPa)	
40	85.13	4.12	3,754.46	
45	83.30	4.29	3,804.94	
50	83.06	4.29	4,498.10	
55	87.72	4.29	4,719.89	

Especially, epoxy resins are widely used as polymer matrices in a production process of the reinforced polymer composites [1-3, 5-9, 17, 18]. Nevertheless, the thermal conductivity of the epoxy resin is about 0.19-0.25 Wm⁻¹K⁻¹ [16], which is very

low when compared to metal material. Thus, thermally conductive materials are filled to these composites in order to disperse heating in hightemperature environments [1-3, 5-9, 17, 18]. Various types of fillers, such as metal oxides, metals, copper,



brass, and aluminium powders, have been used to fabricate thermally conductive epoxy composites [2, 10-12, 16]. In utilisation of metal fillers, aluminium is widely used as the filler in epoxy based composites. This is because it has a high-compatible material to mix with the other materials. Besides, it is a low-cost, non-toxic (green material) and high thermal-conductive material [2, 3, 5-12, 17, 18]. The thermal conductivity of aluminium particles is around 250 Wm⁻¹K⁻¹ [16].

In the injection moulding process, the CTE and the thermal conductivity of the AL-EP mould material are a necessary factor in this application. Thermal conductivity has not only an indirect effect on the accuracy of part dimensions, but also has a direct effect on increasing of the injection cycle and the cooling time [5, 6, 9-12, 17, 18], thus the coefficient of thermal conductivity in each mass mixture ratio was also determined. In previous work from Chungchoo *et al*, they found that the mixing ratio at 60:40 gives the lowest value of the thermal conductivity of about 1.09 $Wm^{-1}K^{-1}$, while the other compositions have similar value of around 1.19 $Wm^{-1}K^{-1}$ (see Table 3). However, the ratio of 55:45 is an optimal strength in their works, and hence, in Fig. 3 shows that the average values of Tg and CTE are 83.73°C and 46.63 ppmK⁻¹, respectively [6].



Fig. 3 Tg and CTE of AL-EP material [6]

Table 3 Therma	conductivity	(K) vs. A	Al contents [6]
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AI contents	0%	40%	45%	50%	55%
K (Wm ⁻¹ K ⁻¹)	0.23	1.09	1.19	1.19	1.21

Tribological properties

The field of tribology was established in 1966 by the JOST committee in UK. The word "Tribology" is derived from Greek words "Tribos" which originally means "rubbing". Also tribology is to study the phenomenon of interacting surfaces in relative motion. Tribological studies consist of friction, wear and its lubricant, and to prevent the premature failures and degradations of material surfaces using heat treatment process or coating process. Friction is



usually classified as a branch of physics or of mechanical engineering. Wear and surface treatment are often studied to be a part of material science & engineering and metallurgical engineering. Lubricants and lubrication may be employed by chemists and mechanical engineers, respectively, Surface coating can be considered as a field of studies in metallurgical and mechanical engineering. Thus, the multidisciplinary studies of tribology are a successful way to improve the lifetime of engineered materials in related industries [19-23]. Recently, particle-filled polymer composites attracted the interest of several industries and related researchers worldwide. Various kinds of particulate fillers are often utilised to increase the friction resistance and the wear resistance of their polymer matrices. In recent years, there have been a number of successful composites made by blending hard particulate fillers in polymer matrix composites. Nevertheless, in particular the tribological property of the AL-EP material in previous works has been less studied. As first mentioned, the tool life of the AL-EP material is much shorter than that of a steel material. This is because the durability of the AL-EP material depends on wear behaviours of the material itself [5. 9, 17]. An investigation of wear behaviours of the AL-EP material, in previous work from Vasconcelos et al, the pin-on-disc wear test was performed at room and elevated temperatures. The specimen was prepared by a volume mixture ratio of 41% for aluminium and 51% for epoxy resin. They found that the wear rate of neat epoxy resin is less than the epoxy resin filled with aluminium powder. This failure cause is because the micro-cracks assist the debonding of aluminium reinforcement (see Fig. 4). At 160°C, it has been found that the wear rate of AL-EP is higher than AL-EP at room temperature, approximately 544%. Moreover, they have found that the wear rate of AL-EP is higher than the epoxy resin without aluminium

addition [17]. Due to the fact that aluminium particles are harder than the epoxy resin, when aluminium particles are pulled out from epoxy matrix, they become a third-body abrasion. This may lead to a rising of wear rate in AL-EP [17, 18].



Fig. 4 Worn surface of AL-EP material [17]

Therefore, it can be summarised that the AL-EP material will lose wear resistance, especially in elevated temperature environments. To improve the friction and wear resistance, the hybrid composite should be employed. Hybrid polymer composites are composed of particle-reinforced, fibre-reinforced and their polymer matrices [17]. In particular fibres are the reinforcing agents and can carry along their longitudinal directions to increase the toughness of the polymer composites. Especially in applications of pair materials in relative motion, the fibre can form to reduce the friction and wear in their composites. Most commonly used fillers include glass fibre, carbon fibre, natural fibre, etc. Similar common polymer matrix materials include phenolic resin, polyurethane, epoxy resin, etc [3, 4, 16, 17]. Larsen et al studied the sliding wear behaviour of fibre composites which compared a glass fibre filled epoxy (G-E) and a



carbon fibre filled epoxy (C-E). They composed the fibre content of 44 vol.% in both materials. They found that wear rate of G-E is higher than C-E material. This is because the carbon fibre in C-E material acts as the lubricant [24]. In 2007, Suresha *et al* also claimed that the three-body abrasive wear of C-E is less than G-E material due to the same reason in the work from Larsen and co-workers, as shown the result in Fig. 5 [25].





While, AL-EP hybrid materials, containing strong fibres of high wear resistance in a brittle matrix, are advanced materials with proper specific mechanical and wear properties. Their resistance to wear by sliding and abrasion has, however, been slightly studied. Lino and co-workers pointed out that the scheme for life extension of the mould involves carbon fibre reinforcement in AL-EP material to decrease the friction and wear. The microstructure of this material shows in Fig. 6.



Fig. 6 Microstructure of AL-EP hybrid composites [17]

They studied the abrasive wear behaviours of a glass fibre filled AL-EP material (G-AL-EP), and compared to a carbon fibre filled AL-EP material (C-AL-EP) as the hybrid composites. The SiC abrasive test under low pressure and water lubrication was performed. They found that wear rate of AL-EP material was reduced when filled their fibres in AL-EP matrix, as shown in Fig 7. This is due to the short fibres that can improve the interfacial strength of AL-EP material [17]. Particularly, carbon fibre can form as the solid lubricant to reduce the friction and wear of AL-EP material [14, 24, 25]. Therefore, it can be concluded that the C-AL-EP is a good alternative to improve the lifetime of the soft tool [3, 17].



Fig. 7 Wear rate of AL-EP hybrid composites [17]

Conclusions and outlook

Aluminium filled epoxy-cast technology, such as the hybrid composites can improve the material properties of the original AL-EP mould. However, some of these aspects enclose the influence of the mechanical and thermal properties as well as tribological properties on the tool wear and life. Hence, a variety of factors in AL-EP hybrid composites material fabrication, such as filler types of the carbon fibre or the graphite fibre, mixing contents, particle sizes, should be investigated to improve their AL-EP material properties, and should be integrated to ensure that it will be feasible to keep



proper mechanical and thermal properties, including the friction and wear as well as structural integrity during the mould life. Furthermore, the AL-EP hybrid composite materials must allow a moulding cycle time to be as short as possible. An emerging issue being addressed in the context of designing injection moulds is their reutilisation. A proposed strategy must be designed for the mould making industry of Thailand or of worldwide to get some advantage from reutilisation in view of further cost reduction and savings in tool development and manufacturing time.

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References

- Chua, C. K., H. Hong and S. L. Ho. 1999.
 Rapid tooling technology. Part 1. A comparative study. Int J Adv Manuf Technol 15: 604–608.
- [2] Brito, Z. and G. Sanchez. 2001. Influence of metallic fillers on the thermal and mechanical behaviour in composite of epoxy matrix. Comp Struct 48: 79-81.
- [3] Vasconcelos, P. V., F. J. Lino, A. Magalhaes and J. L. R. Neto. 2005. Impact fracture study of epoxy-based composites with particle and milled fibres. J Mater Proc Technol 170: 277-283.
- [4] Zhang, Z. and K. Friedrich. 2003. Artificial neural networks applied to polymer composites: a review.
 Compos Sci Technol 63 (14): 2029-2044.
- [5] Cheah, C. M., C. K. Chua and H. S. Ong. 2002. Rapid moulding using epoxy tooling resin. Int J Adv Manuf Technol 20: 368-374.
- [6] Chungchoo, C., S. Kerdnoonwong, A. Wongto and T. Sirithanapipat. 2007. The development of rapid mould. Final Report. Kasetsart University Research and Development Institute.

- [7] Liu, Y. and A. Gong. 2006. Compressive behavior and energy absorbrion of metal porous polymer composite with interpenetrating network structure. Trans Met Soc Chaina 16: 439-443.
- [8] Goyanes, S., G. Rubiolo, A. Marzocca, W. Salgueiro, A. Somoza, G. Consolati and I. Mondragon. 2003. Yield and internal stresses in aluminum filled epoxy resin: A compression test and positron annihilation analysis. **Polymer** 44: 3193-3199.
- [9] Chung, S. I., Y. G. Im, H. D. Jeong and T. Nakagawa. 2003. The effects of metal filler on the characteristics of casting resin for semimetallic soft tools. J Mater Proc Technol 134: 26-34.
- [10] Liu, X-q., L. Ji-yan and C. Shao-jun. 2012. Comparative study of aluminum diethylphosphinate and aluminum methylethylphosphite-filled epoxy flame-retardant composites. **Polym Comp** 33 (6): 918-926.
- [11] Nunwong, P., C. Chungchoo, S. Sirivedin and P. Niranatlumpong. 2011. The effects of thermally sprayed coating thickness on substrate deformation: experimental investigation and finite element analysis. J Solid Mech. Mat Eng 5 (1): 1-13.
- [12] Kuo, C-C. 2013. Development of green rapid tooling technologies. Indian J. Comp 20 : 245-251.
- [13] Holliday, L and J. Robinson. 1973. Review: The thermal expansion of composites based on polymers. J Mater Sci 8 (3): 301-311.
- [14] Robertson, C. G. and C. M. Roland. 2008. Glass transition and interfacial segmental dynamics in polymer-particle composites. Rubber Chem. Technol. 81: 506-52.



- [15] Behrens, E. 1968. Thermal conductivities of composite materials. J Compos Mater 2(1): 2-17.
- [16] Vinson, J. R. and R. L. Sierakowski. 2008. The Behavior of Structures Composed of Composites Materials. 2nded. Springer, Amsterdam.
- [17] Vasconcelos, P. V., F. J. Lino, A. M. Baptista and J. L. R. Neto. 2006. Tribological behaviour of epoxy based composites for rapid tooling. Wear 260: 30-39.
- [18] Ma, S., I. Gibson, G. Balaji and Q. J. Hu. 2007. Development of epoxy matrix composites for rapid tooling applications. J Mater Proc Technol 192-193: 75-82.
- [19] Department of Education and Science, HMSO.1966. Lubrication (tribology) education and research. Jost Report.
- [20] Czichos, H. 1978. Tribology: A Systems Approach to the Science and Technology of Friction, Lubrication, and Wear (Tribology series v. 1). Elsevier Scientific Publishing Company, Amsterdam.
- [21] Dowson, D. 1979. The History of Tribology. Longman Group Limited, London.
- [22] Jost, H. P and J. Schfield. 1981. Energy saving through tribology a techno-economic study. Proc. IMechE 195 (June, 1981): 151-174.
- [23] Briscoe, B.J. 1981. Wear of polymers: an assay on fundamental aspects. Tribo Int 14: 231-243.
- [24] Larsen, Ø. T., L. T. Andersen, B. Thorning, A. Horsewell and E. M. Vigild. 2007. Comparison of friction and wear for an epoxy resin reinforced by a glass or a carbon/aramid hybrid weave. Wear 262: 1013-1020.

[25] Suresha, B., G. Chandramohan, P. Siddaramaiah, P. Samapthkumaran and S. Seetharamu. 2007. Three-body abrasive wear behaviour of carbon and glass reinforced epoxy composites. Mater Sci Eng A 443: 285-291.