MECHANICAL AND THERMAL PROPERTIES OF EPOXY-INORGANIC FILLER SYSTEM – EFFECT OF FAR INFRARED CURING

D. Shin, S. Kim, H. Lee, H. Jung, J. Cho, S. Lim, C. Lee*
AMD Lab, Samsung Electro-Mechanics Co., Ltd., Suwon, Korea
* Corresponding author (**ck1107.lee@samsung.com**)

Keywords: *F-IR, Curing, Epoxy, Filler*

1 Introduction

In addition to the difficulties in PCB industry for the past 10 years, the severe competition and sourcing problem of raw material is getting serious. It has resulted in serious contradiction to meet the needs of benefit-creation and high-end technology at a same time. Such problem is not restricted to a certain business field. So, it is necessary to approach simultaneously to the increase of benefit creation and satisfaction the future needs on technological roadmap based on the combination of new technology, new process and new material innovation. Furthermore, those challenges should be performed timely without any delay [1-4].

Based on the current situation above, it is right time for high end PCB manufacturer like us to develop new innovative processes which can achieve two major goals; profit making and technology leading. In other words, it is crucial factors to develop new technology which can attain cost saving and high density integration process to solve the internal problem such as the decrease of profit contrary to the numerical increase of gross sales. However, under the urgent situation requiring technological capability concerning basic and core material which is fundamental feature for new technology and new process, it is true that our business systems are so much dependent on foreign material suppliers and set-makers' strategical requirements. To meet the needs of upcoming high technology, we are systematically pursuing core technology development for electronic materials based on organic material capability and extension to other technical area.

There are many types of insulating materials for industrial printed circuit board (PCB) manufacturing; copper clad lami-nate(CCL), resin coated copper foil (RCC), prepreg (PPG), and build-up film. Each substrate can be selected depending on the type and function of PCB. In general, most of PCB dielectrics are in the form of organic-inorganic hybrid consisting of epoxy and glass fabric or silica filler [1]. In those systems, epoxy based material is one of the core components to meet the final requirement of electronic products.

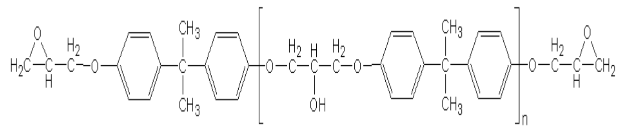
In addition to the conventional curing process to get the optimum material properties of epoxy, it is continuously needed to increase the efficiency and speed of curing cycle. It is generally understood that crosslinking mechanism in network formation of epoxy is very complicated due to many factors such as curing method, temperature, viscosity, reactor size, and curing agent, etc. In addition to that, the final molecular structure of epoxy network system cannot be defined in terms of low or high molecular weight concept. To get the well defined internal structure and efficient process condition, we need to employ alternative curing methods to thermal curing process which is requiring long time process and resulting inefficient curing structure. The incentive for such investigation is typically to reduce the time of cure, or to find more energy-efficient method for curing.

So far, many curing systems such as UV light, e-beam, microwave and far infrared have been studied to make cross-linked epoxy. In case of UV light, it has limited application due to poor penetration ability. Although, e-beam curing has been proved to an accelerated and efficient curing method, it often requires unacceptably high capital outlay [5-8]. In this study, the far infrared curing of epoxy and inorganic filler system has been investigated to achieve improved mechanical, thermal properties and microstructures compared to previous thermal systems. Far-infrared rays allow epoxy resin to form more efficient networking system through the molecule resonance during the curing [9-10] process.

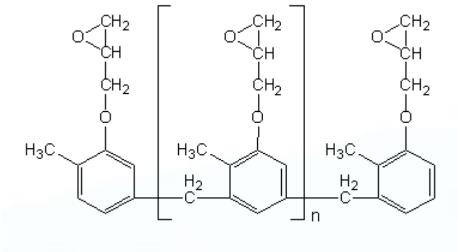
2 Experimental procedures

Main epoxy resins used in this work are Bisphenol A and Novolac types produced by Kukdo chemical Co., Ltd. As an inorganic counterpart for hybrid structure, we choose $CaCO_3$ with 99% purity (Kyoungkipowder Co., Ltd.)

[Bisphenol A type epoxy resin]



[Novolac type epoxy resin]



Epoxy film with inorganic filler was processed with slot die coating machine, after a series of well defined blending processes including homogeneous dispersion of $CaCO_3$ powder. Then the film was cured using tunnel type far-infrared drying heater. Finally, the cured film was treated with permanganate chemical etching for morphological structure on film surface. Measurement of mechanical properties was performed using a UTM (Instron 5543). Sample shape for measurement is typical dog-bone type. Thermal analysis was also conducted with TA-Q400 in the temperature range from ambient temperature to 300℃.

3 Result and discussion

3.1 Mechanical properties

Generally, it is not easy to cure epoxy system in relatively short period via conventional heating process such as box-type convection oven or conveyer-type inline dryer. In the case of far-infrared system, we could get high mechanical properties of final cured epoxy products, in spite of short curing time, as shown in Table 1. It is a direct evidence for more high curing density of epoxy via far-infrared process. The curing time for such results is ranging from 3min to 18min at 210℃. It is assumed that homogeneous crosslinking was formed by the resonance of epoxy molecules with far-infrared rays. Especially, epoxy film which is cured at 210℃ for 6min shows the best properties. On the other hand, Table 2 shows low mechanical properties cured at 230℃ and 250℃. Such as peel strength, tensile strength and so on. This is because low or incomplete curing level of epoxy obtained over 230℃, which is relatively high or severe condition for optimum curing.

3.2 Thermal properties

Figure 1 (a) shows the results of thermal property as a function of far-infrared curing time at 210℃. There is a dramatic decrease of CTE over 6 minutes exposure for curing. However, epoxy system which is cured over 230℃ by far-infrared process shows increase of CTE in Figure 1 (b).

Table 1. Mechanical properties of epoxy-$CaCO_3$ film for different far-infrared curing times at 210℃.

Time (min)	Peel strength (kN/m)	Tensile strength (Mpa)	Modulus (GPa)	Elongation (%)
3	0.916	54.34	4.7	1.37
6	0.985	76.6	5.2	2.12
12	0.976	72.41	5	1.95
18	0.975	72.16	5	2.11

Table 2. Mechanical properties of epoxy-$CaCO_3$ film for different far-infrared curing temperature.

Temp. (℃)	Peel strength (kN/m)	Tensile strength (Mpa)	Modulus (GPa)	Elongation (%)
210	0.985	76.6	5.2	2.12
230	0.893	56.65	4.7	1.34
250	0.881	64.56	4.8	N/A

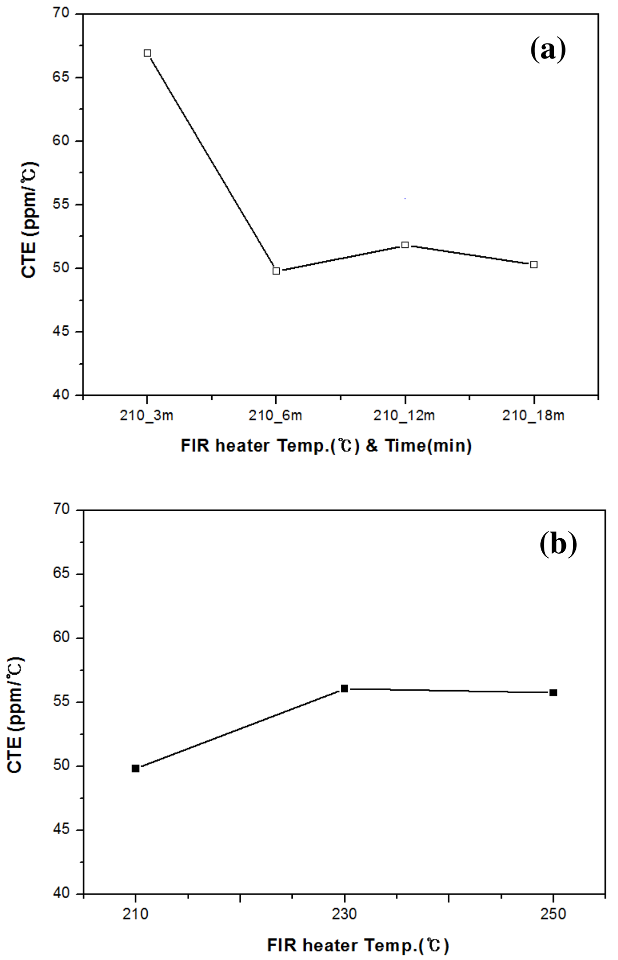


Fig. 1. Variation of thermal properties of epoxy-$CaCO_3$ system for different far-infrared curing condition ; (a) plot of CTE and Tg versus curing time at 210℃ (b) plot of CTE and Tg versus cure temperature for 6min.

3.3 Microstructures

Figure 2, 3 show microstructures of epoxy film after curing and chemical etching. It demonstrates the microstructual effect of curing density between temperature and time. It is also found that there is optimal curing time for densification having thick wall and deep hole. Specifically, epoxy film curing at 210℃ for 6min showed a well-densified film.

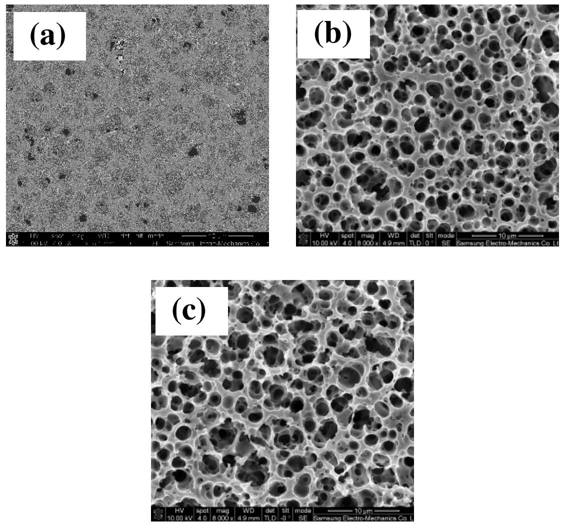


Fig. 2. SEM microstructure of epoxy films with far-infrared curing for (a) 3min (b) 6min (c) 12min at 210℃

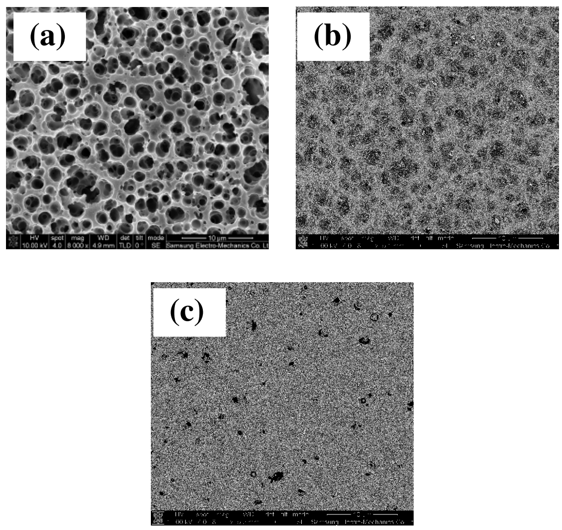


Fig. 3. SEM microstructure of epoxy films with far-infrared curing for 6min at (a) 210℃ (b) 230℃ (c) 250℃

4. Conclusion

In this study, we demonstrated that far-IR system for efficient curing of epoxy composite system is very useful and can be applied to real PCB manufacturing line in comparison with previous conventional heat curing process. Contrary to the convectional heat flow system, far-infrared energy could cause more rapid and defect free intermediate internal structure during cure process. So far, researchers and engineers have avoid such system can cause abrupt boiling of solvent system, and resulted in defects of epoxy matrix. But, we could conclude that there is a certain optimum process condition, and can get perfect state.

The primary objective of this work was to study the effect of far-infrared curing in epoxy and inorganic filler system. Mechanical and thermal properties of epoxy were found to depend on the time of far-infrared curing but it did not follow unlimited rule.

The optimal effect between the temperature and time of far-infrared curing might be the major point when high productivity of epoxy is required in epoxy curing process.

Current work is limited to the effectiveness of far-infrared system to epoxy curing system. To utilize such curing method more generally, we need systematic study on the core factors affecting most efficient epoxy curing such as solvent type, solvent contents, epoxy type, curing speed, mass volume, film thickness, level of film drying, and inorganic filler contents. We will further investigate those fields in near future.

References

[1] US patent 1995/5,428,499

[2] http://www.hitachi-chem.co.jp/english/products/bm/index.html

[3] http://www.taiyoink.co.jp/english/index.html

[4] http://www.dse.co.kr/index.php

[5] M.A. Stuchly, S.S. Stuchly, *IEEE Proc., Part A* 130 (8) pp467-503, 1983

[6] K.L. Carr, *Microwave J.* 26 (1) pp26-36 1991

[7] A.K. Bose, et al., *J. Org. Chem.* 56, pp6968-6970, 1991

[8] D.E. Clark, W.H. Sutton, *Annu. Rev. Mater. Sci.* 26, pp299-331, 1996

[9] M. Younes, et al., *Polymer,* Vol. 35 (24), pp5269-5278, 1994

[10] G. Kortaberria, et al., *Eu. polymer J.* Vol 40 (1), pp129-136, 2004