공학석사 학위논문

Thermal Conductivity of Glass Microsphere / Epoxy Matrix Syntactic Foam

유리중공입자-에폭시 syntactic foam의

열전도도에 관한 연구

2016 년 2 월

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Abstract

Thermal Conductivity of Glass Microsphere / Epoxy Matrix Syntactic Foam

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To investigate the effect of the surface roughness of glass-microsphere on thermal conductivity of glass microsphere/epoxy-matrix syntactic foam, some experiments and measurements were performed. The surface roughness of the glass-microsphere was reduced by a surface treatment. After making specimen having volume fraction 55 ~ 80% with treated and untreated glass-microspheres, the thermal conductivities of the specimens were measured. As the result, it was confirmed that the thermal conductivity of the syntactic foam with the surface-treated particles slightly decreased compared to the syntactic foam using the untreated particles.

Keyword : syntactic foam, thermal conductivity, glass microsphere, epoxy, surface roughness
Student Number : 2011 - 24064

Table of Contents

List of Nomenclature

k_c	:	thermal conductivity of composite
k_f	:	thermal conductivity of filler
k_m	:	thermal conductivity of matrix
f	:	volume fraction of filler
z	:	surface height on a pixel
$\bar{z}$	:	average height of an AFM image
N	:	the number of the pixels of an AFM image

Chapter 1. Introduction

1.1. Purpose of Research

Syntactic foams are a kind of composite material with a polymeric binder and hollow microspheres. Microspheres are made of various materials like glass, polymers, ceramics, metals, etc[1]. The characteristics of polymeric syntactic foams are low density, high specific strength, low thermal conductivity, low moisture absorption and good chemical stability[2-3]. And unlike other foams, the density of syntactic foam does not change during the curing. Because of this dimensional predictability, syntactic foam can be used in the manufacturing of aerospace and marine structures[4].

Due to low thermal conductivity of glass microsphere, it can be applied to products for insulation. In this study, thermal conductivity of composite using microsphere was investigated. Especially, the effect of the interface between glass microspheres and epoxy matrix is considered as the main subject. By a treatment on the surface of glass microspheres, the interface condition between the glass microsphere and the epoxy matrix could be changed.

1.2. Study Background

1.2.1 Rule of Mixture

In the case where the properties of matrix and filler in a composite material are known, the properties of the composite material can be predicted theoretically by using rule of mixture. There are two models of rule of mixture, "Voigt model" (Eq.(1)) for upper−bound modulus and "Reuss model" (Eq.(2)) for under−bound modulus. Generally, material properties of the composite is located between the two bounded values.

$$k_c = f k_f + (1-f)k_m \tag{1}$$

$$k_c = \left(\frac{f}{k_f} + \frac{1-f}{k_m}\right)^{-1} \tag{2}$$

12.2 Surface Roughness

Surface topography of the glass microsphere was measured using AFM(Atomic Force Microscopy, XE−100, Korea) in order to analyze the roughness information. There are some mathematical parameters to quantify roughness of surfaces, but RMS(Root Mean Square) roughness is generally used. If N pixels exist in an image, the RMS roughness can be calculated as follows[5].

$$(RMS)^2 = \frac{1}{N^2}\sum_{n=1}^{N}(z(n) - \bar{z})^2 \tag{3}$$

Chapter 2. Experiments

2.1. Surface Treatment of Microspheres

It was reported that a treatment with acid / basic solution generates 10 ㎚ scale RMS roughness on surface of glass fibers[6]. In this study, a basic solution treatment was chosen for changing roughness of microspheres. The basic solution of pH 12 was prepared by dissolving lithium hydroxide powder into deionized water. And the mixing ratio of the solute was 40 g/L. Without steady mixing of the solution, the glass microspheres float due to the density difference between the microspheres and the solution. The float of the microspheres can cause insufficient wetting and make the surface treatment ineffective. To prevent that, the solution was placed in a polyethylene cylindrical bottle of 8㎝ diameter, and the reaction was carried out in a roller mixer at 60 rpm, for 200 hours(Fig. 1).

After the reaction, the microspheres are cleaned with deionized water and dried in a vacuum oven at 70℃ for 10 hours(Fig. 2-3). The agglomerated powders generated during the drying are separated by a sieve of 500X500㎛ mesh size(Fig. 4).

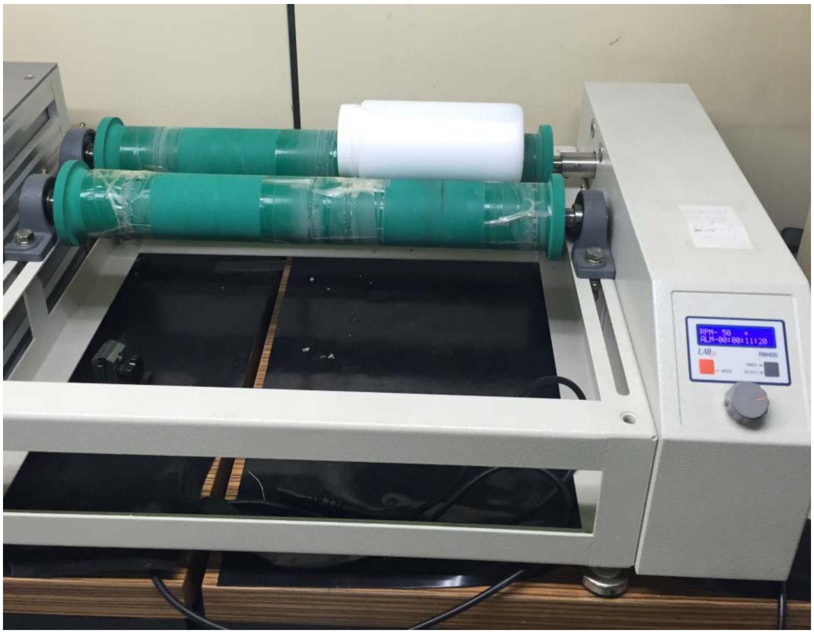


Fig. 1 Surface treatment using roller mixer

Fig. 2 Drying in vacuum oven

Fig. 3 Agglomerated glass microspheres during drying

Fig. 4 Sieving glass microspheres

Fig. 5 Treated glass microspheres

2.2. Making Syntactic Foam Specimens

The syntactic foam specimens were made of epoxy as the matrix and glass microspheres as the filler. The range of the volume fraction of the microspheres is from 60% to 80% at an interval of 5% volume fraction. The microspheres are 3M™ Type K1 Glass Bubbles and its density and size are listed in Table 1.

Untreated glass microspheres and treated glass microspheres were used for syntactic foam specimens respectively. To measure the thermal conductivities of the specimens, 4 specimens were made for one volume fraction.

The making process is as follows: After mixing the epoxy resin with the glass microspheres in a container(Fig. 6), the mixture was placed in a vacuum oven at 70℃, for 30 min in order to remove air bubbles in it. Then the mixture was cooled down to 30 ~ 40℃. Hardener is added into the mixture and the mixture put into a open mold which has 40X40X5㎜ rectangular parallelepiped shape cavities(Fig. 7). Curing was performed in a convection oven at 80℃, for 4 hours(Fig. 8, Fig.9).

	Typical	Minimum	Maximum
True Density(g/cc)	0.125	0.10	0.14
Size Distribution	10th%	50th%	90th%
Particle Size(micron)	30	65	115

Table 1. Properties of 3M™ Type K1 Glass Bubbles

Fig. 6 Mixed glass microspheres and epoxy resin

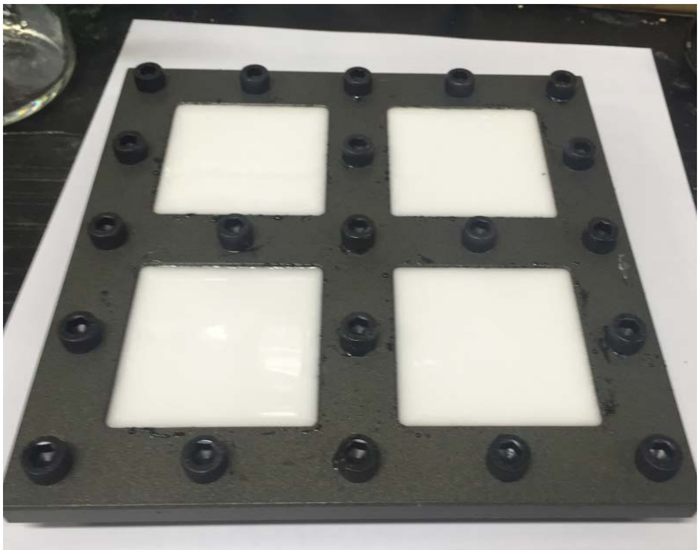


Fig. 7 Mixture in open mold

Fig. 8 Convection oven

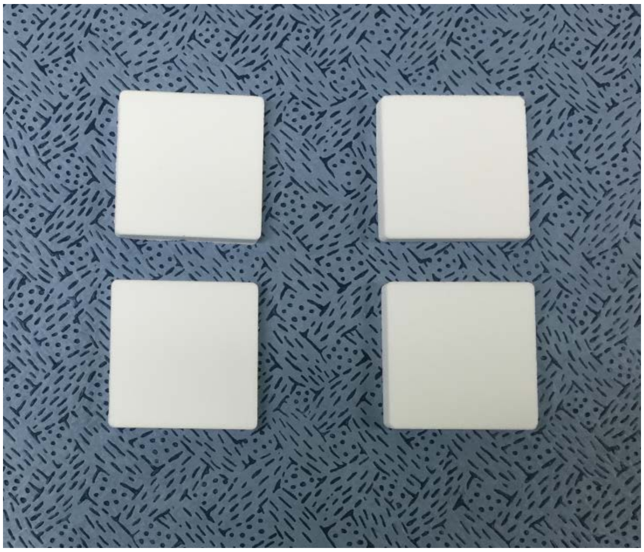


Fig. 9 Syntactic foam specimens

2.3. Measurement of Thermal Conductivity

To measure thermal conductivity C−THERM Tci™ Thermal Conductivity Analyzer was used. This apparatus applies MTPS(Modified Transient Plane Source) method, its sensor contacts only one surface of the syntactic foam specimen during the thermal conductivity measurement. And it can be adjusted not only measurement for solid materials but for liquid or powder material. Using this advantage, the thermal conductivities of untreated and treated glass microspheres were measured by immersing the sensor into the beaker where the glass microspheres are in(Fig. 10).

The thermal conductivities of the syntactic foam specimens in this experiment range from 0.05 to 0.15 W/mK. As being 0.12 W/mK the boundary, different calibrations were applied. In over thermal conductivity of 0.12W/mK, deionized water was used as the contact agent between the specimen and the sensor. In under 0.12 W/mK, contact agent was not used but another calibration mode was applied.

The measurement for one volume fraction was performed with 4 specimens of same volume fraction, and each value was determined by which 4 measured values except the first measurement(Fig. 11).

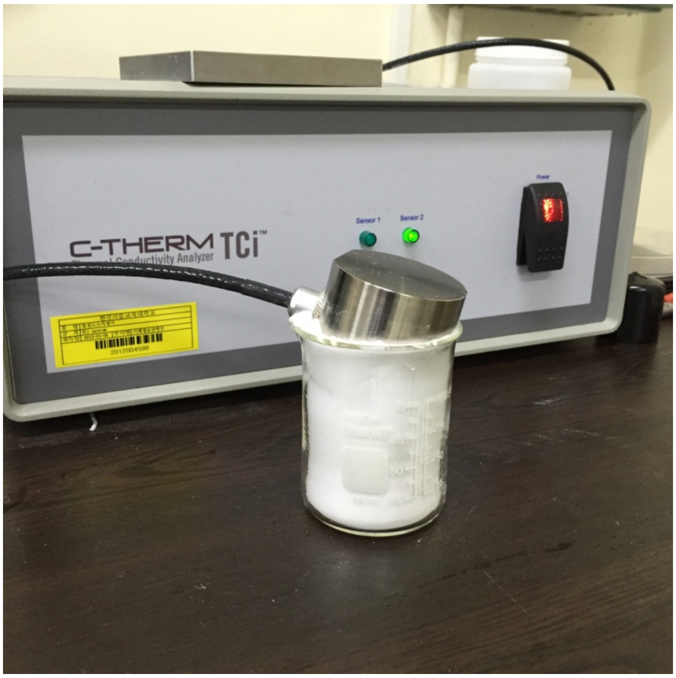


Fig. 10 Measurement of thermal conductivity of glass microsphere

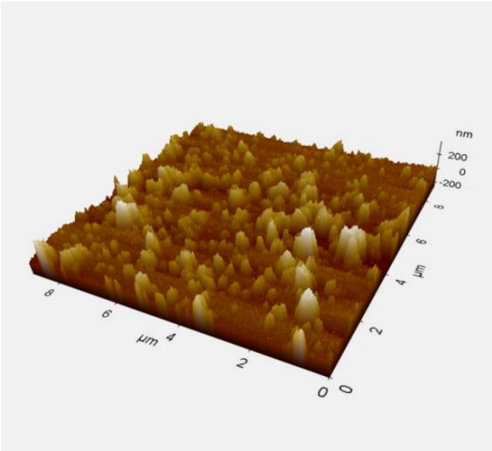
Fig. 11 Measurement of thermal conductivity of syntactic foam

Chapter 3. Results & Discussion

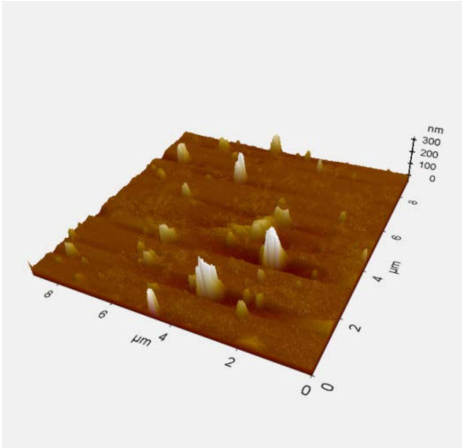
3.1. Surface Roughness of Glass Microspheres

For taking AFM images, the glass microspheres were fixed on glass substrates. And images of 100h–treated, 200h treated, and untreated glass microspheres were obtained. The scan range of the AFM is 9X9㎛ , each image is composed of 256X256 pixels

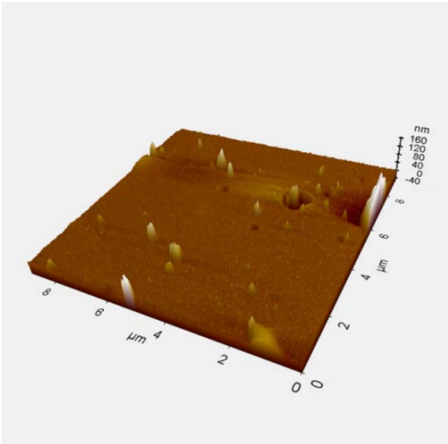
The result of AFM is shown in Fig. 12. The existence of many bumps of under–2㎛–width and under–300㎛–height were determined on the surface of untreated microspheres. The number of bumps decreased sharply after the surface treatment by basic solution and that causes a decrease in RMS roughness of the surfaces. Using Eq.(3) for the calculation, the RMS roughness of the untreated microspheres was 66㎚, and it decreased to 24㎚ for 100h–treated, 10㎚ for 200h–treated. These decrease in the RMS roughness is considered to be caused by the corrosion due to the basic solution and the relative friction between microspheres in the roller mixer.



(a)



(b)



(c)

Fig. 12 AFM image of surface of glass microspheres :
(a) Untreated, (b) 100h-treated, (c) 200h-treated

3.2. Thermal Conductivities of Glass Microspheres

To examine effect of the surface treatment on the thermal conductivity, the thermal conductivities of the treated and untreated glass microspheres were measured respectively. The results were shown in Table 2. The thermal conductivity of untreated glass microspheres was 0.044W/mK. After the surface treatment, it was reduced by about 7% (0.041W/mK). It was thought that the loss of mass on the surface of microspheres and the change of the contact condition among microspheres are the reason for the reduction of the thermal conductivity.

In addition, there was no difference between the thermal conductivity of 100h-treated microspheres and that of the 200h-treated microspheres. The thermal conductivity of the glass microsphere becomes lower as its shell thickness is thinner. But despite of the additional 100h-treatment, the thermal conductivity of microspheres did not decrease. This means that the treatment affected to the surface roughness, but didn't to the shell thickness.

Material	Epoxy resin	Glass microsphere		
		Untreated	100h treated	200h treated
Thermal conductivity (W/mK)	0.23	0.044	0.041	0.041

Table 2. Thermal conductivities of epoxy resin and microspheres (measured)

3.3. Thermal Conductivities of Syntactic Foams

The thermal conductivities of the syntactic foam with the untreated glass microsphere were measured and plotted in Fig. 13. Since the thermal conductivity of the glass microsphere is much smaller than that of the epoxy resin, the thermal conductivity of a syntactic foam becomes lower as the volume fraction of microsphere is higher. The result corresponds well with the value expected by rule of mixture.

And the thermal conductivities of the syntactic foams using the treated microspheres were measured to ascertain the difference caused by the surface treatment. The result of the measurement was shown in Fig. 14. As the result, when the treated microspheres were used, the average of thermal conductivity of the syntactic foam was reduced by about 9% compared to that of the syntactic foam with untreated microspheres.

In consideration of no change in the thickness of the glass microspheres during the surface treatment, the changes in interface of microsphere−microsphere and microsphere−resin may be responsible for the reduction of the thermal conductivity of the syntactic foam.

Although the 100㎚ scale bumps on the surface of microspheres were removed, it was reported that the treatment with basic solution generates 10㎚ scale roughness. To determine whether scale roughness causes the change of the thermal conductivity, additional experiments should be needed.

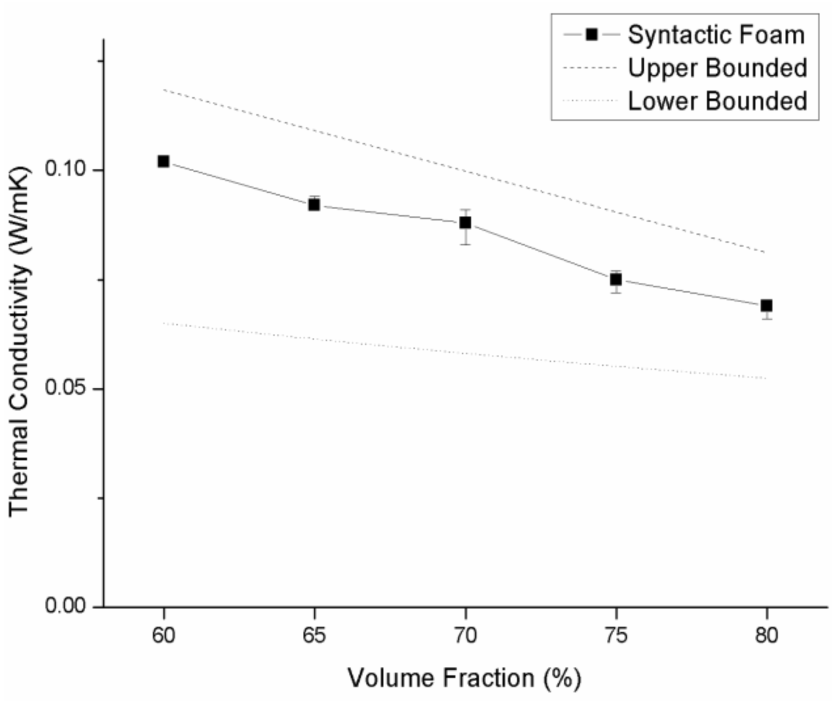


Fig. 13 Thermal conductivity of syntactic foams with untreated glass microspheres

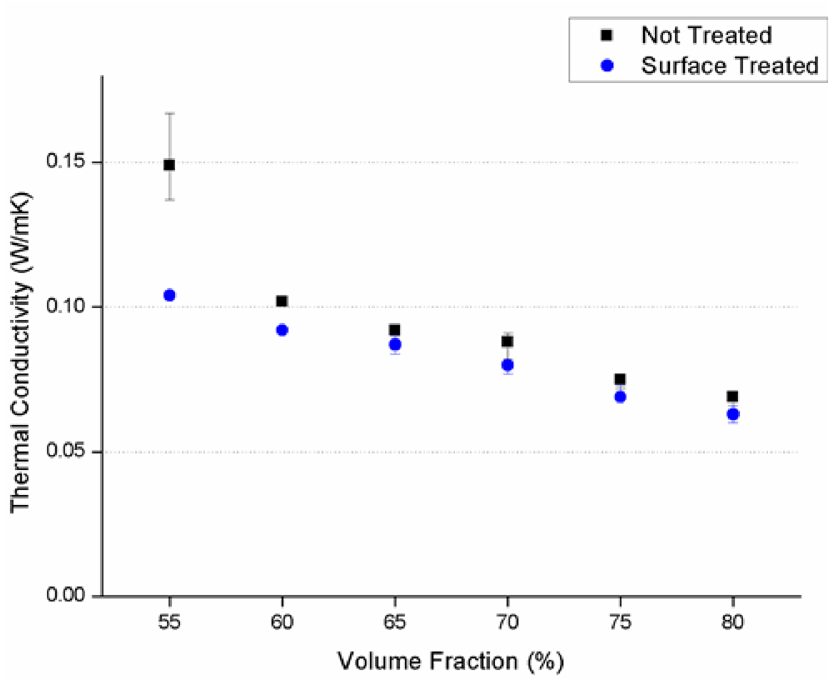


Fig. 14 Change in thermal conductivity of syntactic foams by surface treatement

4. Conclusion

The purpose of this study is to determine the effect of surface roughness of glass microsphere on the thermal conductivity of glass microsphere / epoxy syntactic foam. For that, a surface treatment which changes the roughness of glass microspheres was performed, and syntactic foam specimens were made with the treated microspheres.

As the result of the measurement of syntactic foam specimens, the thermal conductivity of syntactic foam using treated glass microspheres was reduced by 9% in average. The change in the thermal conductivity is considered to be caused by the increase of thermal resistance between glass microsphere and epoxy resin. But additional experiments are needed to determine whether 100㎚ scale bumps are responsible for the reduction of the thermal conductivity of the syntactic foams or 10㎚ scale roughness is.

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국 문 초 록

유리중공입자-에폭시 Syntactic Foam의 열전도도에 관한 연구

서울대학교 대학원

기계항공공학부 멀티스케일 기계전공

이 기 모

본 연구는 유리중공입자 표면의 거칠기가 유리중공입자-에폭시 syntactic foam의 열전도도에 어떠한 영향을 미치는지 알아보기 위하여 진행 되었다. 유리중공입자의 표면 거칠기를 변화시키기 위하여 염기성 용액을 이용한 표면처리를 실시 하였다. 그 후, 표면 처리 완료된 입자를 사용하여 55 ~ 80% 의 유리중공입자 부피비를 가지는 syntactic foam을 제작하였고, 그 열전도도를 측정하여 표면처리가 되지 않은 입자를 사용한 syntactic foam의 열전도도와 비교하였다. 그 결과, 표면처리를 실시 한 유리중공입자를 사용하여 제작 한 syntactic foam의 열전도도가 기존 입자를 사용 한 경우와 비교해 소폭 감소 되는 것이 확인 되었다.