

## Microencapsulation of Imidazole Curing Agent by Solvent Evaporation Method Using W/O/W Emulsion

Min Jae Shin,<sup>1</sup> Young Jae Shin,<sup>2</sup> Seung Won Hwang,<sup>3</sup> Jae Sup Shin<sup>3</sup>

<sup>1</sup>Department of Chemical and Biomolecular Engineering, KAIST, Daejeon 305-701, Korea

<sup>2</sup>Macromolecular Science and Engineering, University of Michigan, Ann Arbor, Michigan 48109, USA

<sup>3</sup>Department of Chemistry, Chungbuk National University, Cheongju, Chungbuk 361-763, Korea

Correspondence to: J. S. Shin (E-mail: jsshin@chungbuk.ac.kr)

**ABSTRACT:** The epoxy–imidazole resin system is used to form the anisotropic conducting film. The latent character of the system is very significant. In this study, imidazole (Im) or 2-methylimidazole (2MI) was encapsulated for the latent curing system to use in the reaction of epoxy resin. Polycaprolactone was used as a wall material, and the solvent evaporation method was used to form the microcapsule using W/O/W emulsion. The shelf life of the microcapsules was studied for the epoxy resin, and the curing behavior of the microcapsules for epoxy resin was examined using a differential scanning calorimeter. The curing times at 150 and 180°C were estimated using an indentation method. The microcapsules of Im or 2MI exhibited a long shelf life for epoxy resin. When comparing the results of the previous methods with the results of this study using the W/O/W emulsion, finer microcapsules were formed and the microcapsule has longer shelf life. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 129: 1036–1044, 2013

**KEYWORDS:** microencapsulation; solvent evaporation method; curing agent; imidazole; epoxy resin

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### INTRODUCTION

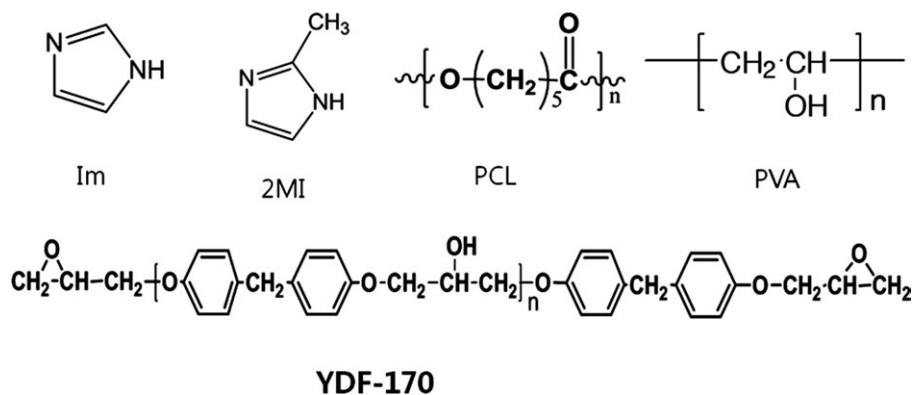
Epoxy resins are widely utilized in a number of industrial applications, including adhesives, coatings, and electronics because of their excellent mechanical and chemical properties, such as their high tensile and compressive strengths, good solvent and chemical resistance, and high heat distortion temperatures. The curing process of the epoxy resins can be carried out using a wide range of curing agents, such as amines, anhydrides, polyamides, phenol formaldehyde resins, and polysulfides.<sup>1–4</sup>

Although epoxy resins are cured with primary and secondary amines through a step growth polymerization, tertiary amines undergo a chain growth polymerization. Imidazoles are tertiary amines that are often used as hardeners in a variety of epoxy resin systems to initiate the homopolymerization of the epoxy compounds.<sup>5–12</sup>

Recently, an epoxy–imidazole resin system was used to form an anisotropic conducting film (ACF) for use in electronic equipment, such as LCDs.<sup>13,14</sup> The production speed of these LCDs is dependent on the curing rate of the ACF. Therefore, the development of ACFs with fast reactivities and manageable properties is very important. The epoxy system must be a one-pot system for electronic equipment applications, such as LCDs. Therefore, the storage stability is very significant at room temperature. In

the one-pot system, the epoxy resin and the curing agent do not have to react with each other at the storage temperature and the preparation temperature for setting the equipment.

Unfortunately, imidazole is not a latent curing agent for epoxy resin systems. In the epoxy–imidazole system, imidazoles react with the epoxy resin at room temperature, and the epoxy resin changes into a hard polymer after it has been mixed with the imidazole curing agent at room temperature for a period of time ranging from 1 h to 1 day. Imidazoles must be converted to an unreactable form to create a one-pot system for the epoxy–imidazole system. Among the methods of forming unreactable imidazoles, the encapsulation of the imidazole is an easy and economic method.<sup>15,16</sup> In the previous results, we reported the encapsulation of imidazole curing agents for epoxy resin. In the first our result, we reported the encapsulation of imidazole curing agents using solvent evaporation method.<sup>17</sup> In that study, 2-phenylimidazole (2PhI) was used as a core material, but imidazole (Im) and 2-methylimidazole (2MI) could not be used although Im and 2MI have a more superior curing property to epoxy resins than 2PhI. In the solvent evaporation method, the organic solution of the core material with the polymer was added dropwise into the large amount of water. During this process, the organic solvent was evaporated and microsphere was formed. Hence, if the core material is well soluble in water,



**Figure 1.** The structures of the materials that were used in this study.

it is difficult for the core material to stay in a microsphere. But as 2PhI is not much soluble in water, 2PhI was selected as a core material. In our next study, the spray-drying method was used to encapsulate the imidazole curing agents.<sup>18</sup> Im or 2MI could be used as the core material in that method. But the obtained microspheres have battered round shape and showed fast initial release rate in ethanol. This result means that a lot of imidazoles stayed at the surface of the microsphere or the vicinity of the surface area of the microsphere. Hence, we tried the spray-drying using w/o emulsion instead of the solution of imidazoles and polymeric wall material.<sup>19</sup> Using this method, battered round shape and fast initial release rate were somewhat improved. But to accomplish more improved property, we tried the encapsulation by solvent evaporation method using W/O/W emulsion in this study.

In this study, the imidazole curing agents were encapsulated with polycaprolactone (PCL) by the solvent evaporation method using the W/O/W emulsion to create the one-pot system for the epoxy–imidazole system, and the encapsulated curing agent was also characterized.

## EXPERIMENTAL

### Materials

Figure 1 shows the structures of the materials that were used in this study. Diglycidyl ether of bisphenol F (YDF-170) was obtained from Kukdo Chemical. Imidazole (Im), 2MI, PCL ( $M_w = 80,000$ ,  $M_w = 65,000$ , and  $M_w = 14,000$ ), poly(vinyl alcohol) (PVA) (88% hydrolyzed,  $M_w = 22,000$ ), and dichloromethane (DCM) were obtained from Aldrich.

### Instruments

The differential scanning calorimetry (DSC) studies of the curing behavior were performed using a Scinco DSC N-650 differential scanning calorimeter under a nitrogen atmosphere. High-purity indium was used to calibrate the calorimeter. All of the samples ( $\sim 10$  mg) were stored within sealed aluminum DSC pans. The DSC studies of the YDF-170 cure were performed from 15 to 200° at a heating rate of 10°C/min. The homo mixer (T. K. homo mixer mark 2, model 2.5) was used to control the stirring speed. High-performance liquid chromatography (HPLC) was performed using an Alliance Dissolution System (Waters). Elemental analysis (EA) was performed using an ele-

mental analyzer (EA 1110, CE Instruments). Scanning electron microscopy (SEM) was performed using both a Hitachi S-2500C and Hitachi S-5200V scanning electron microscopes.

### Encapsulation of Imidazoles

The most representative method of encapsulation is shown below. In brief, 1.00 g of Im was dissolved in 10 mL of 1 wt % aqueous PVA solution. This solution was put into the solution which contained 1.00 g of PCL in 20 mL of DCM at 20°C. This mixture was stirred at 4000 rpm for 15 min. After that, the mixture was added dropwise quickly into 100 mL of 1.0 wt % aqueous PVA solution which was stirred at 200 rpm at 40° for 10 min. DCM was evaporated in this process, and the formed microcapsules were isolated through centrifugation at 2000 rpm for 5 min. Then, the microcapsules were dried in a vacuum oven for 10 h at 30°.

### The Determination of the Curing Time

The curing time was measured using an indentation method. The reaction vessel was heated to a desired temperature, and the mixture of the epoxy resin and the microcapsules was added to the vessel. Then, the surface of the resin mixture was pierced every second, and the time was recorded when the pin did not pierce the surface.

### The Measurement of the Permeability of the Microcapsules

A 100-mL round bottomed flask was filled with ethanol and 0.10 g of the microcapsules containing imidazole was added at 35°C. The solution was stirred with a magnetic stir bar at a very slow speed (one rounding every 2 s). Then, 1 mL of the sample solution was removed from the upper part of the solution at determined intervals, and the amount of the permeated imidazole was measured using HPLC.

## RESULTS AND DISCUSSION

### Encapsulation of Imidazoles

In this study, the microencapsulation was conducted by the solvent evaporation method using W/O/W emulsion. As the melting point of the PCL is very low, PCL will be melted very easily at high temperature. Therefore, PCL was selected as the polymer for the encapsulation of the curing agent.

As Im and 2MI were soluble in water, and water was used in solvent evaporation method, if we try to conduct the

**Table I.** The Content of Imidazole Estimated by EA in the Microcapsule That Was Prepared with Different Stirring Speeds During the Formation of the W/O/W Emulsion<sup>a</sup>

Imidazole	Stirring speed	EA (%)
Im	2000	11.2
	1000	15.3
	500	20.7
	200	22.1
2MI	2000	13.1
	1000	17.6
	500	21.7
	200	23.5

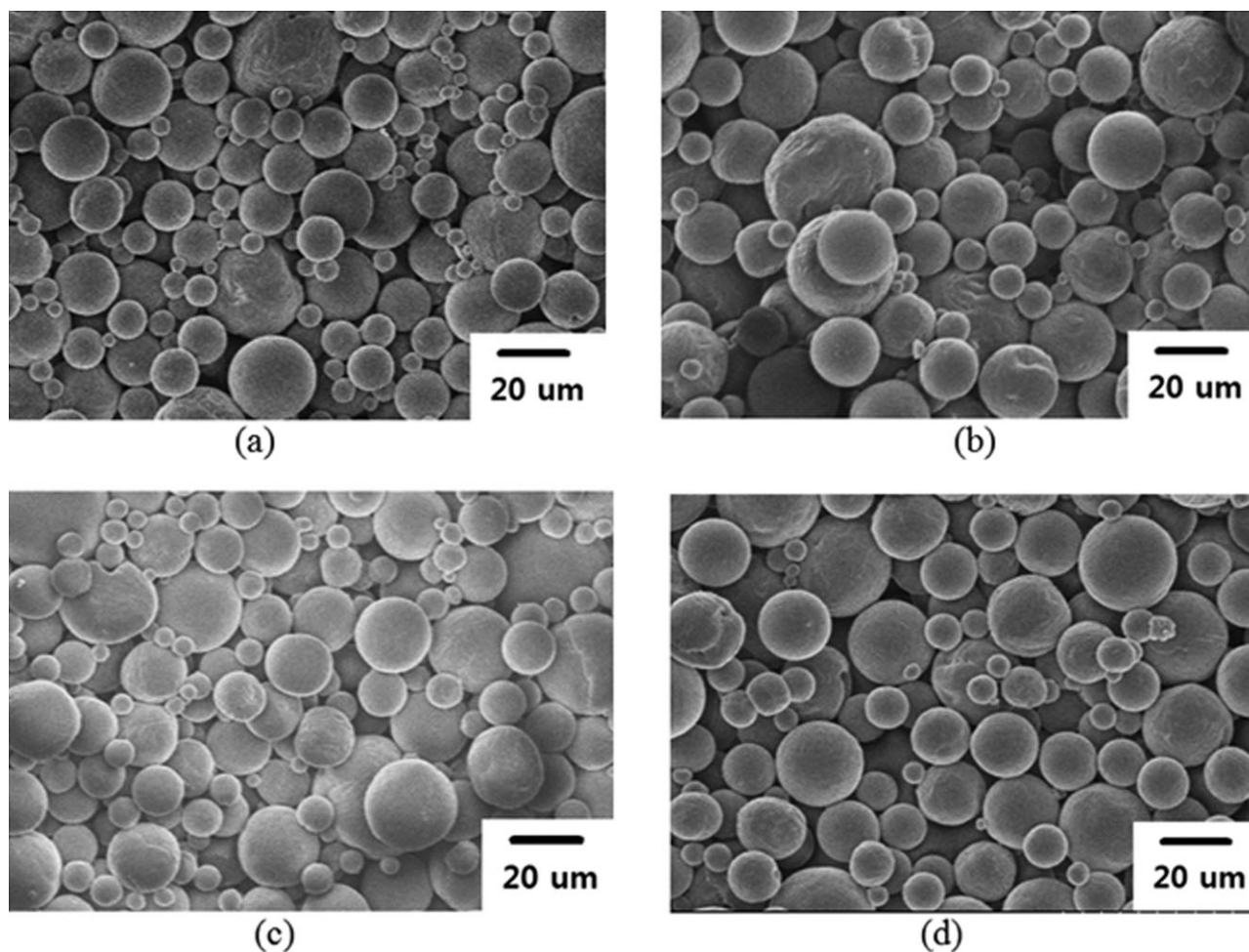
<sup>a</sup>Formulation (PCL/imidazole) 1.0/0.5,  $M_w$  of PCL 65,000.

microencapsulation by solvent evaporation method using simple organic solution, Im or 2MI is diffused out from the microcapsule to water during encapsulation process. Hence, Im and 2MI cannot be used in the microencapsulation by solvent evaporation method using a simple organic solution. In this study, we

used W/O/W emulsion instead of simple organic solution. In this process, at first W/O emulsion was formed using aqueous solution of imidazoles and DCM solution of PCL. This W/O emulsion was put into the large amount of water, and then W/O/W emulsion was formed for solvent evaporation. PVA was used as an emulsion stabilizer in all of the process.

#### The Effect of the Stirring Speed

The stirring speed is one of the main factors when forming fine microcapsules. We tried to conduct the process at several stirring speed. In this experiment, Im was dissolved in aqueous PVA solution, and this solution was put into the solution which contained PCL in DCM. This mixture was stirred at 4000 rpm to form the W/O emulsion. This 4000 rpm stirring speed was chosen to make the fine small emulsion, and this stirring speed is relatively higher than general experiment. But this stirring speed had not much effect on the formation of the microcapsule. When we tried the encapsulation at 2000 rpm or at 3000 rpm, the similar results were obtained comparing with that at 4000 rpm. But the results at 4000 rpm got a little better regular shape and more small size. Hence, the remaining experiments were performed at 4000 rpm.



**Figure 2.** SEM photographs of the microcapsules that were prepared with different stirring speeds during the formation of W/O/W emulsion, PCL/2MI = 1.0/0.5,  $M_w$  of PCL: 65,000. (a) 2000 (b) 1000, (c) 500, and (d) 200 rpm.



**Table II.** The Content of Imidazole Estimated by EA in the Microcapsule That Was Prepared with Different Ratios of PCL/Imidazole

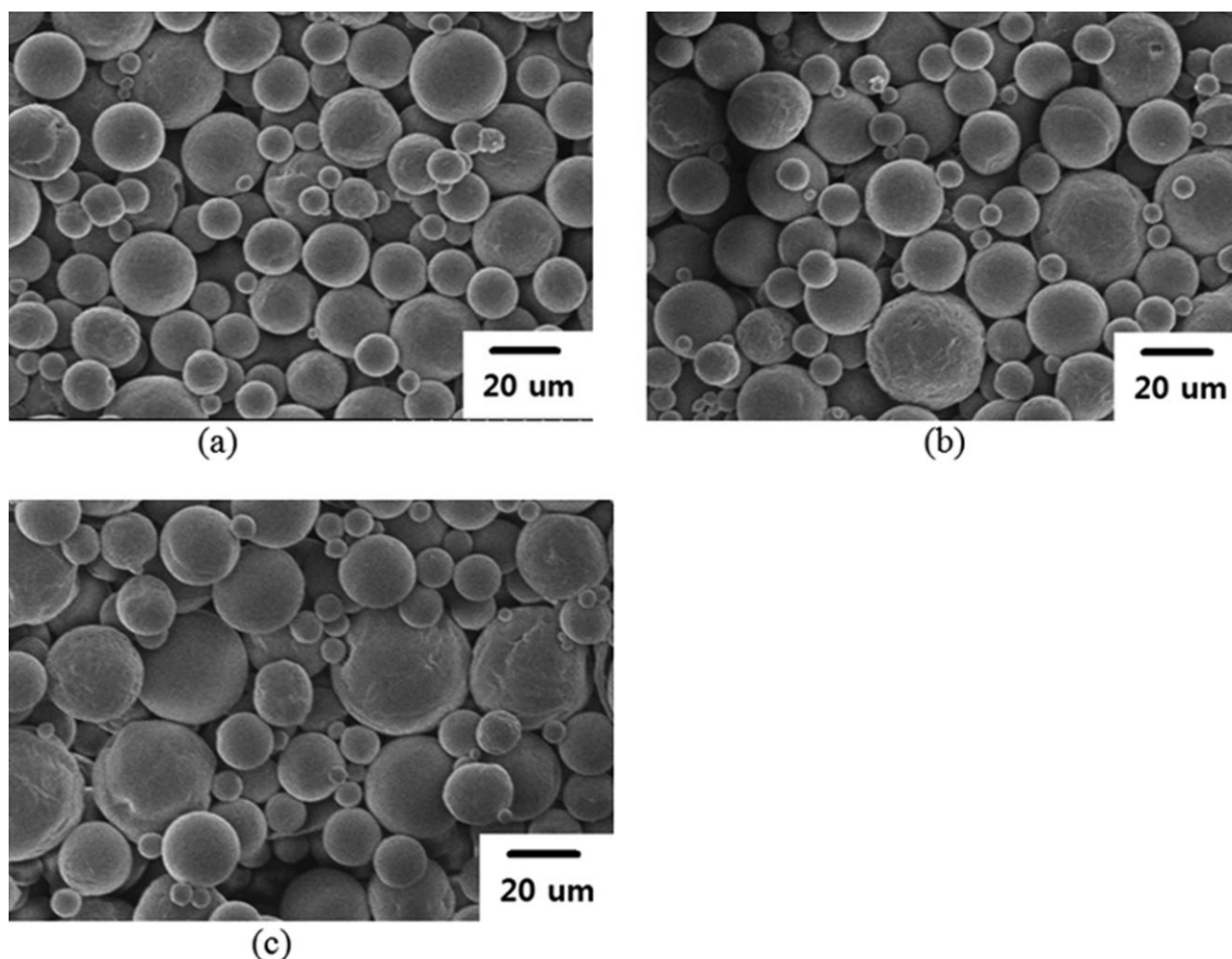
Imidazole	Formulation (PCL/imidazole, g/g)	EA (%)
Im	1.0/0.5	22.1
	1.0/0.75	27.4
	1.0/1.0	32.9
2MI	1.0/0.5	23.5
	1.0/0.75	28.2
	1.0/1.0	34.1

Then, the mixture was added dropwise quickly into aqueous PVA solution which was stirred at 200 rpm at 40°C. DCM was evaporated in this process. The stirring speed in this process has much effect on the formation of the microcapsule. Hence, we conducted the experiment by changing the stirring speed. After the microcapsules were formed, the amount of imidazole in the microcapsules was estimated from the ratio of nitrogen to carbon in EA experiments. The EA experiments were conducted

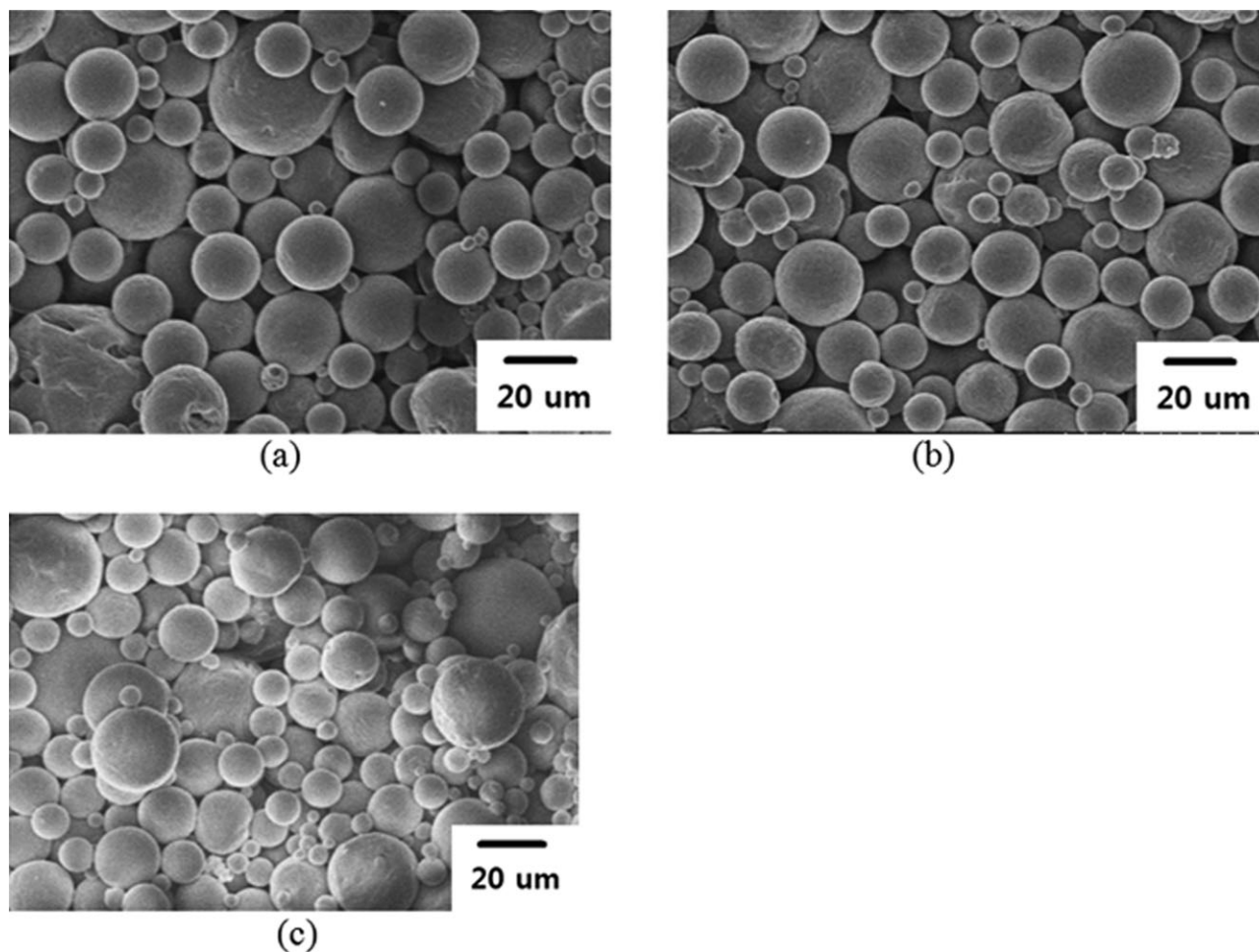
using the microcapsules from different imidazoles and other imidazole/PCL ratios. The results are summarized in Table I.

In Table I, the contents of imidazole in the microcapsules were reduced as the stirring speed was increased. The content of Im was 22.1 wt % at 200 rpm, and that was reduced to 11.2 wt % at 2000 rpm. The content of 2MI was 23.5 wt % at 200 rpm, and that was reduced to 13.1 wt % at 2000 rpm. This results mean that the formed W/O emulsions could be destroyed at the violent stirring when this emulsion was put into large amount of water. The estimated amount of imidazole in the microcapsule is smaller than the theoretical amount calculated from the feed amount. It means that some amount of imidazole was diffused out from the microcapsule to water in this process.

In Figure 2, the SEM photographs of the microcapsules that were prepared at different stirring speed are shown. All of the microcapsules had regular shape and size. The microcapsules had sizes of  $15.1 \pm 8.0$ ,  $14.5 \pm 8.0$ ,  $13.5 \pm 7.0$ , and  $12.7 \pm 6.5$   $\mu\text{m}$  for the 200, 500, 1000, and 2000 rpm, respectively. The size of the microcapsule was reduced with increasing the stirring speed but the reducing amount is small.



**Figure 3.** SEM photographs of the microcapsules that were prepared with different ratios of PCL/2MI, (a) 1.0/0.5, (b) 1.0/0.75, (c) 1.0/1.0,  $M_w$  of PCL: 65,000.

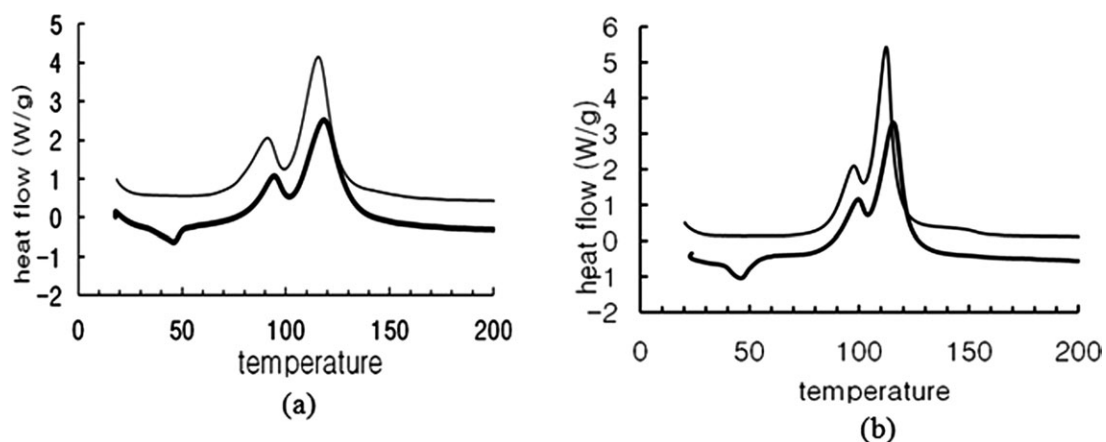


**Figure 4.** SEM photographs of the microcapsules that were prepared with different molecular weights of PCL, (a) 80,000, (b) 65,000, and (c) 14,000. PCL/2MI = 1.0/0.5.

#### The Effect of the Feed Ratio of Imidazole to PCL

The effect of varying the ratio of imidazole to PCL on the formation of the microcapsules was examined. The amounts of

PCL were fixed at 1.0 g and the amount of imidazoles was varied from 0.5 to 1.0 g. The molecular weight of PCL was 65,000. After the microcapsules were formed, EAs were conducted to



**Figure 5.** Scanning DSC curves for the curing of the epoxy resin. (a) (—) PCL/Im = 1.0/1.0, microcapsule/YDF-170 = 18/100, (---) Im /YDF-170 = 6/100, (b) (—) PCL/2MI = 1.0/1.0, microcapsule/YDF-170 = 18/100, (---) 2MI/YDF-170 = 6/100.

**Table III.** The Kick-off and Peak Temperatures of the Microcapsules

Imidazole	Formulation (PCL/imidazole)	Content of imidazole by EA (%)	Feed ratio (microcapsule/epoxy resin)	Kick-off temp. (°C)	Peak temp. (°C)
Im	0/1.0	100	6/100 (Im/epoxy resin)	68	116.6
	1.0/0.5	22.1	27/100	97	132.5
	1.0/0.75	27.4	21/100	94	128.9
	1.0/1.0	32.9	18/100	92	125.8
2MI	0/1.0	100	6/100 (2MI/epoxy resin)	82	112.7
	1.0/0.5	23.5	27/100	93	129.8
	1.0/0.75	28.2	21/100	92	125.5
	1.0/1.0	34.1	18/100	89	121.2

measure the amount of imidazole in the microcapsules. The EA experiments were conducted using the microcapsules from different imidazoles and other PCL/imidazole ratios and the results are summarized in Table II.

The SEM micrographs of the microcapsules that were prepared with different ratios of PCL/2MI are shown in Figure 3. In this experiment, PCL 65,000 and 2MI were used. The PCL/2MI ratios that were used to form the microcapsules were 1.0/0.5, 1.0/0.75, and 1.0/1.0. Almost all of the microcapsules were similar in shape. The microcapsules had sizes of  $12.7 \pm 6.5$ ,  $13.2 \pm 7.0$ , and  $14.7 \pm 8.0 \mu\text{m}$  for the 1.0/0.5, 1.0/0.75, and 1.0/1.0 ratios, respectively. In Figure 3, as the amount of the imidazoles–core material was increased, the microcapsule got a little larger size.

#### The Effect of PCL Molecular Weight

The encapsulations were conducted using PCL with different molecular weights of 14,000, 65,000, and 80,000 to examine the effects of the PCL molecular weight on the microcapsule formation. In the formation of the microcapsules, the PCL/2MI ratio was 1.0/0.5. The SEM photographs of the microcapsules using PCL 80,000, PCL 65,000, and PCL 14,000 are shown in Figure 4. The microcapsules were sized  $13.7 \pm 7.0$ ,  $12.7 \pm 6.5$ , and  $11.5 \pm 7.0 \mu\text{m}$  for Mw 80,000, 65,000, and 14,000 PCL, respectively. The microcapsules were almost uniform shapes, and the

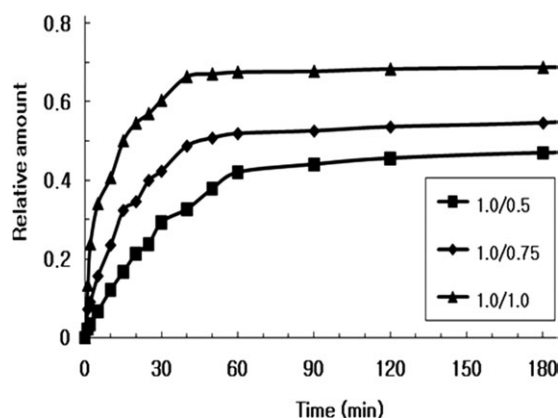
microcapsule size increased with increasing PCL molecular weight.

#### Curing Behavior of the Microcapsules for Epoxy Resin

DSC was used to investigate the curing behavior of the microcapsules for epoxy resin. In this experiment, the molecular weight of PCL was 65,000, the PCL/Im ratio was 1.0/1.0, and the microcapsule/YDF-170 ratio was 18/100. DSC was conducted from 15 to 200°C at a heating rate of 10°C/min. This result was compared with the result from the sample in which pure Im was used instead of the microcapsules. The Im/YDF-170 ratio was 6/100. These two samples had different feed ratios because the two samples had the same amount of Im for epoxy resin. The results are shown in Figure 5. In case of 2MI, DSC was also conducted under the same condition with the case of Im. The results are also shown in Figure 5. In this figure, the exothermic pattern was monitored using DSC because the curing reaction of the epoxy resin with imidazoles was exothermic. Similar patterns were observed for the two samples, but a delay of 8.5–17.1°C in the maximum peak temperature for the microcapsules because the polymeric wall material needed to melt. Two peaks were shown in the DSC data for both Im and 2MI: the first owing to the adduct formation of epoxy resin and imidazoles, and the second owing to the polymerization of epoxy resin.<sup>12</sup>

**Table IV.** The Curing Rates and the Shelf Lives of the Microcapsules

Imidazole	Formulation (PCL/imidazole)	Content of imidazole by EA (%)	Feed ratio (microcapsule/epoxy resin)	Curing time at 150°C (s)	Curing time at 180°C (s)	Shelf life at 20°C (day)
Im	0/1.0	100	6/100 (Im/epoxy resin)	13	8	1
	1.0/0.5	22.1	27/100	25	18	40
	1.0/0.75	27.4	21/100	22	16	35
	1.0/1.0	32.9	18/100	17	12	23
2MI	0/1.0	100	6/100 (2MI/epoxy resin)	9	6	0.5
	1.0/0.5	23.5	27/100	19	15	30
	1.0/0.75	28.2	21/100	17	13	26
	1.0/1.0	34.1	18/100	13	9	19



**Figure 6.** Release behaviors of the 2MI/PCL microcapsules that were prepared with different ratios of PCL/2MI (PCL:  $M_w = 65,000$ ).

In the case of the microcapsules, the endothermic peak at about 50°C was owing to the melting of PCL. Using the different PCL/imidazole ratios, DSC studies were conducted and the results are summarized in Table III. The starting temperature of curing (kick-off temperature) and the temperature of maximum peak (peak temperature) were described. These results were compared with the data using pure imidazoles. In Table III, the kick-off temperature and the peak temperature were increased with increasing relative PCL amount in the microcapsules. The peak temperatures of 2MI were lower than those of Im.

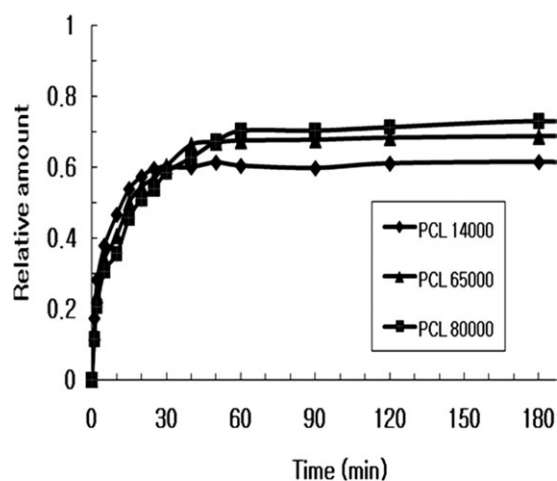
The curing times were estimated at 150 and 180°C using an indentation method, and the results are summarized in Table IV. The curing times at 150°C were increased with increasing PCL amount in the microcapsules, and the curing times at 180°C showed a similar trend. The curing time of 2MI was lower than that of Im at both 150 and 180°C.

After the epoxy resin and imidazoles were mixed at 20°C, this mixture was stored at the same temperature. The shelf life of the microcapsules was examined at 20°C by measuring the time before the curing started. This shelf life of the microcapsules was compared to that of the pure imidazoles, and the results are also summarized in Table IV. The curing reaction started at 20°C after 1 day had passed for pure Im. On the other hand, no curing occurred after storage for 40 days at 20°C for 1.0/0.5 (PCL/Im). The curing reaction started at 20°C after 0.5 days had passed for pure 2MI, but no curing occurred after storage for 30 days at 20°C for 1.0/0.5 (PCL/2MI).

#### Permeability of the Microcapsules

The permeability of the microcapsules was examined by measuring the amount of 2MI permeating from the microcapsules in ethanol at various time intervals. In this experiment, the microcapsules were formed using different ratios of PCL/2MI (1.0/0.5, 1.0/0.75, and 1.0/1.0). The results are shown in Figure 6. In the case of 1.0/0.5, the release rate was slower than that of the 1.0/1.0 case.

The permeability of the microcapsules was also examined by changing the molecular weight of PCL. In this experiment, the microcapsules were formed using PCL molecular weights of 80,000, 65,000, and 14,000, and the PCL/2MI ratio used for



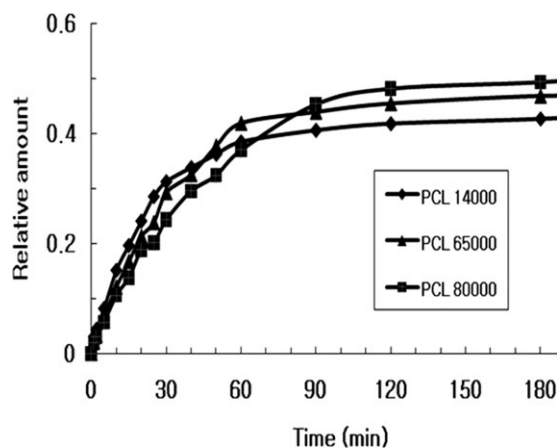
**Figure 7.** Release behaviors of the 2MI/PCL microcapsules that were prepared with different molecular weights of PCL, (PCL/2MI = 1.0/1.0).

these experiments was 1.0/1.0. The results are shown in Figure 7. In Figure 7, the release rates of all the samples showed similar results. But the release rate was a little slower as the molecular weight of PCL was increased. In case that the PCL/2MI ratio was 1.0/0.5, the results of release behavior by changing the molecular weight of PCL are shown in Figure 8. Almost similar pattern is shown in Figure 8. But the results in Figure 8 are much slower than that in Figure 7.

#### Comparison of These Present Results with Those of the Previous One

We compared these present results with the results of our previous report on the encapsulation of the imidazole curing agent with PCL by the solvent evaporation method and the spray-drying method. The results are summarized in Table V.

We tried to form the microcapsules using four-type method, solvent evaporation method with simple organic solution (SE1),<sup>17</sup> spray-drying method with simple organic solution (SD1),<sup>18</sup> spray-drying method with W/O emulsion (SD2),<sup>19</sup> and solvent evaporation method with W/O/W emulsion (SE2).



**Figure 8.** Release behaviors of the Im/PCL microcapsules that were prepared with different molecular weights of PCL, (PCL/2MI = 1.0/0.5).



**Table V.** The Comparison of These Present Results with Those Previous One

Method	Imidazole	Formulation (PCL/imidazole)	Content ratio in microcapsule (wt %)	Feed ratio (microcapsule/ epoxy resin)	Curing time at 150°C (s)	Curing time at 180°C (s)	Shelf life at 20°C (day)
a	2PhI	1.0/1.0	10	-	-	-	30
b	Im	5/5	48.5	12/100	14	9	4
		7/3	28.6	20/100	16	10	7
	2MI	5/5	48.8	12/100	10	7	2
		7/3	28.9	20/100	11	8	3
	2PhI	5/5	49.1	12/100	27	17	8
		7/3	29.0	20/100	32	21	15
c	Im	5/5	47.1	12/100	15	8	4
		10/5	30.9	18/100	17	11	13
	2MI	5/5	47.5	12/100	10	7	2
		10/5	31.1	18/100	12	9	10
	Im	5/5	32.9	18/100	17	12	23
		10/5	22.1	27/100	25	18	40
2MI	5/5	34.1	18/100	13	9	19	
	10/5	23.5	27/100	19	15	30	

$M_w$  of PCL: 65,000.

a: Solvent evaporation method with simple organic solution.<sup>17</sup>

b: Spray-drying method with simple organic solution.<sup>18</sup>

c: Spray-drying method with W/O emulsion.<sup>19</sup>

d: Solvent evaporation method with W/O/W emulsion.

Using SE1, we could use only 2PhI as a core material but Im or 2MI could not be used. The content in the microcapsule was only 10 wt %. The shelf life was more than 30 days. Using SD1, Im and 2MI could be used as core materials, and the content in the microcapsule was increased up to 49 wt %. But the shelf life was very much shortened. Using SD2, the shelf life grew longer two or three times comparing SD1. Using SE2, the shelf life was longer greatly. SEM photographs and release behaviors showed that the microcapsule by SE2 was encapsulated very well.

The future study will focus in raising the content of core material in the microcapsule, because the polymeric wall material can give an influence to the physical property of the final epoxy polymer.

## CONCLUSIONS

In this study, imidazole curing agents were encapsulated with PCL by a solvent evaporation method using W/O/W emulsion to create a one-pot system for an epoxy-imidazole system. Im and 2MI could be used as core materials with the formation of W/O/W emulsion. The amount of imidazole in the microcapsule was measured using EA. The stirring speed during the formation of W/O/W emulsion was very much effective to the content of imidazole in the microcapsule. As the stirring speed was increased, the content of imidazole in the microcapsule was decreased. The permeability of the microcapsules was measured in ethanol. The release rate was slower at higher molecular weights of PCL. In the curing reaction with the epoxy resin, the microcapsules of imidazoles exhibited a delayed kinetic behavior compared to pure imidazoles. The curing times were estimated

at 150 and 180°C using an indentation method. These microcapsules exhibited a long shelf life. When comparing the results of the previous methods with the results of this study, finer microcapsules were formed and the microcapsule has longer shelf life using the W/O/W emulsion.

## ACKNOWLEDGMENT

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