

## 재활용 PET를 이용한 고분자 몰타르의 제조 및 흡음 특성

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### Preparation of Polymer-modified Mortars with Recycled PET and Their Sound Absorption Characteristics

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**초록:** 재활용 PET와 fly-ash를 이용하여 두 가지 종류의 고분자 몰타르(PMM)를 제조하였다. 재활용 PET의 조성을 변화시켜 경성(rigid) polymer-modified mortar(PMM)와 연성(flexible) PMM을 제조하였다. 마찰계수 측정을 포함하는 기계적 특성과 흡음 특성을 조사하였고 상업적 PMM인 에폭시 PMM과 PET PMM을 비교하였다. 기계적 특성들의 결과로부터 경성 PMM이 상업적인 PET PMM에 비하여 우수함을 알 수 있었다. 흡음계수 측정에서 경성 PMM과 연성 PMM 모두 상업적인 PMM에 비하여 매우 우수한 흡음 특성을 보여주었다. 또한, 마찰계수 측정결과는 경성 PMM이 바닥재의 활용에 적절하다는 것을 보여주었다.

**Abstract:** Two different types of polymer-modified mortars(PMM) were prepared with recycled PET and fly-ash. One is rigid PMM and the other is flexible PMM which are based on the composition of recycled PET. Their mechanical properties including friction coefficient measurement and damping characteristics such as sound absorption were investigated and compared with the commercial PMM such as epoxy PMM and PET PMM. The result from mechanical properties indicated that the rigid PMM could be competitive with the commercial PET PMM. The measurement of sound absorption coefficient showed that both rigid PMM and flexible PMM had much better damping capacity than commercial PMM. However, the friction coefficient of rigid PMM revealed that it would be suitable for the use as floor material.

**Keywords:** polymer-modified mortar, recycled PET, fly-ash, sound absorption coefficient.

### Introduction

Recently the research and development of polymer-modified mortar (PMM) has been focused on the utilization of recycling materials.<sup>1–3</sup> PMM has several advantages in comparison with Portland cement mortar. They are the short curing time, high bond strength, high modulus, good thermal stability, and other advantages. Generally the polymer mortar consists of sand, filler and polymeric additives such as latexes, polymer powders, water-soluble polymers, liquid resins, and monomers. As polymeric additives there are SB rubber, polyacrylic ester latex, epoxy, polyester, methyl methacrylate, methyl cellulose, and etc. However, the unsaturated polyester and epoxy resins have been widely used as polymer mortars.

Especially the unsaturated polyester has been known as an excellent polymer mortar for the construction materials due to its rapid setting, high bond strength, and good durability including water resistance, chemical resistance, abrasion resistance, and thermal resistance in comparison with those of latex-modified systems.<sup>4–6</sup> Moreover, the unsaturated polyester can be recycled to reduce the environmental problems and it has another functional advantage such as sound absorption behavior.

As a recycling system for the PET three different systems have been reported:<sup>7,8</sup> (1) Material recycling converts the recycled PET to the polymer materials with use of the melt extrusion system. (2) Thermal recycling makes the heat usable which is produced by the combustion of recycled PET. (3) Chemical recycling produces the chemicals with recycled PET using the chemical treatment. The chemical recycling

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system also has three different methods: (1) Hydrolysis uses water to produce monomer or oligomer with recycled PET, (2) Glycolysis uses the glycol, such as ethylene glycol (EG), propylene glycol (PG), neopentyl glycol (NPG), (3) Methanolysis uses methanol. Methanolysis and hydrolysis normally are used to produce monomers while the glycolysis is used to make unsaturated PET, polyurethane foam, and etc. In this study we used the glycolysis to produce unsaturated PET, which is a more suitable method to prepare PMM.

We also used fly-ash as filler in the PMM in order to replace more common fillers like quartz flour. Fillers are used in the production of PMM not only to reduce the cost of the material, but to improve its dimensional stability. However, recent studies have reported that fly-ash has an excellent compatibility with polymers such as polyolefins, nylon 6.6, epoxy, and polyurethane resins and poly(vinyl chloride). Fly-ash also has numerous advantages over quartz flour such as low density, strong filling ability, excellent fluidity, and good processibility of the filled materials.<sup>8</sup> Moreover, because fly-ash is a by-product of coal combustion, the cost of fly-ash is very low. However, we expect that the fly-ash may contribute to the sound absorption characteristic of PMM based on recycled PET because of its various size fraction and its ability to give compactness.

In this study we prepared PMM with recycled PET and fly-ash, and characterized it in terms of mechanical properties and sound absorption property. Comparison with commercial PMM such as epoxy mortar (PMM-EP, Sikadur30, Sika Korea Co.) and polyester mortar (PMM-PET, Tecroc, U. K.) will be also discussed in detail.

## Experimental

**Materials.** The recycled PET was prepared from PET mineral water bottles. As a glycolysis solvent styrene monomer (Aldrich) was used. To prepare unsaturated polyester resin, diethylene glycol (Gana Chemical) and unsaturated diacids such as maleic anhydride (Gana Chemical) and phthalic anhydride (Junsei) were used without further purification. Table 1 shows the composition of the unsaturated polyester resins. We prepared two different types of resins which are flexible

**Table 1. The Composition of Unsaturated Polyester Resins<sup>9,10</sup>**

	FR(wt%)	RR(wt%)
Recycled PET	21	22
Diethylene glycol	28	19
Styrene monomer	30	33
Maleic anhydride	10.5	17.0
Phthalic anhydride	10.5	9.0

**Table 2. The Composition of Fly-ash**

Chemicals	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	SO <sub>3</sub>	MgO
wt%	56-66	21-23	4-8	0.2-0.4	0.1-0.3	0.9-1.2

resin (FR) and rigid resin (RR).

Table 2 shows the composition of fly-ash (Boryung E&C) used in this study. We used fly-ash which was passed through 10 mesh sieve in order to get the homogeneous size distribution of fly-ash.

**Preparation of PMM (Glycolysis).** FR can be prepared by the melting of recycled PET mixed with diethylene glycol, maleic anhydride, and phthalic anhydride in a round bottomed flask under N<sub>2</sub> gas at 220 °C for 3 to 4 hrs. Afterwards the completely melted resin is cooled down to 40~50 °C, and finally the styrene monomer is added as a solvent.

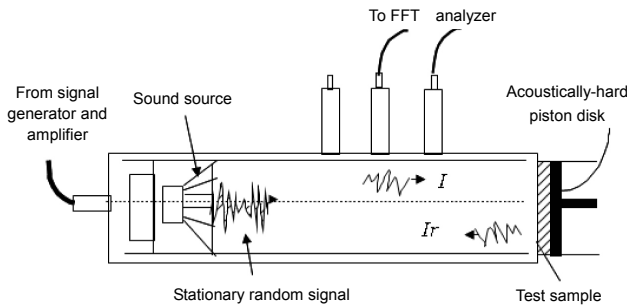
The PMM-FR was prepared by the mixing 60 wt% of melted FR and 40 wt% of filler (fly-ash/sand=1/1 wt%/wt%). We used a mold (L×W×H; 80 mm×40 mm×30 mm) to prepare the sample, and 7 days curing at the ambient temperature in order to obtain enough strength in the samples. However, since the curing starts in a few minutes we had to be cautious not to make bubbles during the preparation of sample because bubbles directly affect the mechanical strength.

In the case of RR the melting of recycled PET is carried out at 190 °C for 5 hrs. The styrene monomer is added at the temperature of 110~120 °C. The procedure to make PMM-RR was the same as the PMM-FR preparation method. When the resins were cured it was easily recognized that RR was harder than FR.

**Mechanical Characteristics.** In this study KS F2405<sup>11</sup> (ASTM C1439) was used to measure the compressive strength. The samples were cured for 7 days before the measurement of compressive strength and an average of 7 measurements was obtained. We used universal testing machine (Instron 4202) to measure the tensile strength according to ASTM D638.

In the case of measurement of the friction coefficient out there was no specific regulation. We used the KS F2429<sup>12</sup> which is the measurement of abrasion resistance for the concrete. In this study the ball on plate sliding equipment for the measurement of friction coefficient was used. The ceramic ball (3/10 inch) was used as a reference and the sliding speed of 134 rpm and 3.5 mm stroke of the equipment was fixed to each 100 g sample.

**Sound Absorption Characteristic.** The impedance tube (Tube type 4206) prepared by B & K company was used to measure the signal with a wide frequency, and the signal analysis was followed with a multichannel signal analyzer Type 3550. Figure



**Figure 1.** The schematic diagram of the sound absorption measurement tube with microphone.

1 shows the simple diagram of the impedance tube. Two different samples were prepared for the use of low frequency and high frequency, and their diameters were 2.9 cm and 10 cm respectively.

## Results and Discussion

**Mechanical Properties(Compressive Strength and Tensile Strength).** Table 3 shows the result of mechanical properties of PMM-FR and PMM-RR compared with the commercial polymer mortar, i.e. epoxy mortar and polyester mortar. The average values of compressive strength for each sample were expressed by the reduced values which were obtained by  $P/V$ . P and V mean measured value and volume of sample respectively. It can be easily found that PMM-RR shows an excellent compressive strength similar to that of PMM-PET. In addition, PMM-RR shows the higher compressive strength than that of PMM-EP. This revealed that PMM-RR could be a substitute for the commercial PMM even though recycled PET and fly-ash were used. However, PMM-FR shows the lowest compressive strength due to the lower number of methyl group in the ethylene glycol compared with that in the propylene glycol during the glycolysis, resulting in the low cohesive energy.

In the case of tensile strength both PMM-FR and PMM-RR prepared with recycled PET and fly-ash were showing the lower values than the commercial PMM, i.e. PMM-PET (Table 4). This is due to the fly-ash in the PMM-FR and PMM-RR, resulting in lowered adhesive power between polymer and filler. Even though fly-ash contributes to the high density of mortar it does not mean that this will enhance the adhesion between polymer and fly-ash. However, both PMM-FR and PMM-RR showed the much higher tensile strength than that of PMM-EP, indicating that they can be used for the building material. But it can be concluded from the result of mechanical properties that PMM-FR and PMM-RR is more suitable if they are used as the floor material

**Table 3. Compressive Strength of Various PMM**

Sample code	Measurement	Compressive strength ( $\text{kg}/\text{cm}^2$ )	Avg. value ( $\text{kg}/\text{cm}^2$ )
PMM-FR	1	39.45	40.02
	2	35.27	
	3	43.65	
	4	39.85	
	5	41.88	
PMM-RR	1	425.48	412.47
	2	400.77	
	3	415.20	
	4	409.95	
	5	410.95	
PMM-EP	1	139.31	145.85
	2	155.27	
	3	136.62	
	4	151.56	
	5	146.50	
PMM-PET	1	439.88	437.53
	2	464.09	
	3	421.61	
	4	427.53	
	5	434.55	

**Table 4. Tensile Strength of Various PMM**

Sample code	Measurement	Tensile strength ( $\text{kg}/\text{cm}^2$ )	Avg. value ( $\text{kg}/\text{cm}^2$ )
PMM-FR	1	32.55	31.06
	2	30.95	
	3	29.21	
	4	31.75	
	5	30.85	
PMM-RR	1	81.59	77.13
	2	75.74	
	3	76.21	
	4	77.54	
	5	74.55	
PMM-EP	1	20.63	20.46
	2	19.48	
	3	19.27	
	4	21.60	
	5	21.34	
PMM-PET	1	106.33	105.17
	2	102.20	
	3	104.29	
	4	106.07	
	5	106.96	

instead of the construction repairing material.

The friction coefficient was measured with ball on plate equipment for 5 min. The measured values at the first 2 sec and the last 2 sec were excluded, and then the average values at every second were taken.

Table 5 shows the friction coefficients of various PMM.

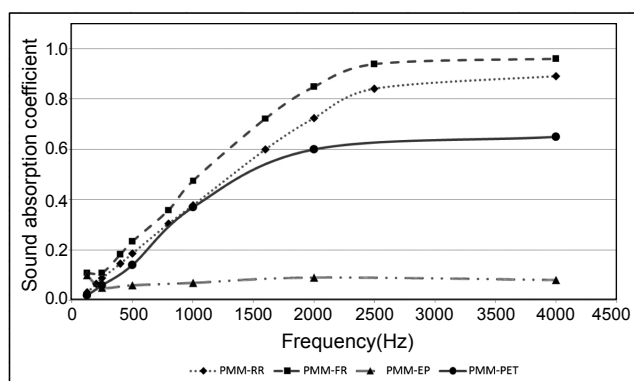
It reveals that PMM-FR and PMM-RR show the relatively high friction coefficient, indicating that they are able to be a substitute for the commercial PMM. However, the result of friction coefficient shows that PMM-FR and PMM-RR will be better for using of a floor material which agrees well with their mechanical characteristics.

**Sound Absorption Characteristics.** The sound absorption coefficients of various PMM are summarized in Figure 2. Figure 3 shows the noise reduction coefficient (NRC) of PMM. The NRCs are the average values of the sound absorption coefficients at the important frequencies such as 250, 500, 1000, 2000 Hz. Both PMM-FR and PMM-RR prepared in this study showed the better sound absorption coefficients than that of commercial PMM, PMM-PET and PMM-EP. It was revealed that the PMM-EP did not have any sound

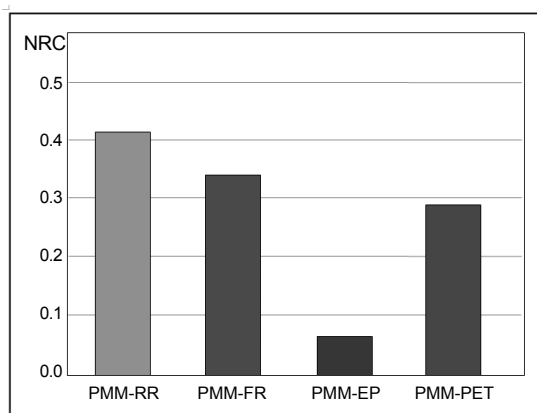
absorption property indicating that epoxy in the PMM-EP was used as a binder only. However, PMM-PET showed good sound absorption property, especially at the high frequency. This is due to the excellent acoustic characteristic of PET, which has been known as an excellent sound absorption material.<sup>13</sup> When PET was manufactured to the fiber it was found that the sound absorption coefficient improved up to 20–30% than glass fiber. However, it was also reported that the density of material with PET played an important role to enhance the sound absorption coefficient. It was generally known that PET with high density had a high sound absorption coefficient.<sup>14</sup> As seen in Figure 2, both PMM-FR and PMM-RR showed better sound absorption property in the all frequencies than that of commercial PMM. This is attributed to the fly-ash which is mostly infiltrated by a great deal of polymer matrix and has good interfacial adhesion with the matrix, resulting in the high density of PMM. It was reported that the damping mechanisms include not only the contribution of matrix viscoelasticity, but also grain boundary sliding (filler/filler) and interfacial sliding (filler/matrix) friction.<sup>15</sup> The composite filled with fly-ash has many contributions coming from friction loss and hollow structure, which may remarkably increase the sound absorption capacity. However, for the PMM filled with fly-ash, which also has an excellent damping capacity, the hollow structure is rare. So it can be concluded that in the case of PMM filled with fly-ash a frictional loss affects the sound absorption capacity more than the matrix viscoelasticity. The results from NRC also show that PMM filled with fly-ash has better sound absorption capacity than the commercial PMM, and they agree very well with the results from the sound absorption coefficient.

**Table 5. The Friction Coefficients of Various PMM**

Sample Value	PMM-FR	PMM-RR	PMM-PET	PMM-EP
Avg.	0.6563	0.7012	0.6017	0.4729
Max.	0.6851	0.7487	0.6723	0.5715
Min.	0.5184	0.5816	0.5655	0.2596



**Figure 2.** Sound absorption coefficient of various polymer modified mortars.



**Figure 3.** NRC of various PMM.

### Conclusions

A new PMM with recycled PET and fly-ash was prepared and its physical properties and sound absorption capacity were investigated. Two different types of PMM with recycled PET, flexible and rigid PMM, showed better sound absorption capacity than the commercial PMM due to the fly-ash infiltrated with polymer matrix, resulting in high friction loss and high density of PMM. The NRC result also confirms the better sound absorption capacity of PMM prepared in this study, and it agrees well with the results of measurements of sound absorption coefficients. The results from determination of the mechanical properties indicate that rigid PMM based on recycled PET and fly-ash can be used commercially. In particular, the high friction coefficient of rigid PMM based on recycled PET and fly-ash indicates that it will be better for use as floor material.

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### References

1. F. Mahdi, A. A. Khan, and H. Abbas, *Cem. Concr. Comp.*, **29**, 241 (2007).
2. E. H. Hwang and Y. S. Ko, *J. Korean Ind. Eng. Chem.*, **14**, 644 (2008).
3. M. C. Bignozzi, A. Saccani, and F. Sandrolini, *Compos. Part A*, **31**, 97 (2000).
4. Y. W. Choi, D. J. Moon, Y. J. Kim, and M. Lachemi, *Constr. Build. Mater.*, **23**, 2829 (2009).
5. K. S. Rebeiz, D. W. Fowler, and D. R. Paul, *Polym. Composite*, **1**, 27 (1993).
6. Y. Li, D. J. White, and R. L. Peyton, *Conservation and Recycling*, **24**, 87 (1998).
7. Abdel-Azim and Abdel-Azim A, *Polym. Eng. Sci.*, **36**, 2973 (1996).
8. F. Awaja and D. Pavel, *Eur. Polym. J.*, **41**, 1453 (2005).
9. K. S. Rebeiz, J. W. Rosett, S. M. Nesbit, and A. P. Craft, *J. Mat. Sci. Lett.*, **15**, 1273 (1996).
10. U. R. Vaidya and V. M. Nadkarni, *Ind. Eng. Chem. Res.*, **26**, 194 (1987).
11. Korean Standard Information Center, <http://www.standard.go.kr>, (Testing Method for Compressive Strength of Concrete), 12, 2005.
12. Korean Standard Information Center, <http://www.standard.go.kr>, (Testing Method for Abrasion Resistance of Concrete) 07, 2007.
13. H. S. Byun and T.G. Lee, *Polymer(Korea)*, **25**, 427 (2001).
14. N. Kino and T. Ueno, *Appl. Acoustics*, **69**, 575 (2008).
15. J. H. Gu, X. N. Zhang, and M. Y. Gu, *J. Alloy Compd.*, **372**, 306 (2004).