Polymer(Korea), Vol. 33, No. 1, pp 91−95, 2009

# 내열성 전자기 노이즈 흡수 폴리(아미드-이미드)/연자성체 복합체 필름

**한지은 · 전병국 · 구본재 · 조승현* · 김성훈** · 이경섭** · 박연흠 · 이준영†**
성균관대학교 화학공학과, *성균관대학교 고분자기술연구소, **(주)창성기술연구소
(2008년 11월 4일 접수, 2008년 11월 11일 수정, 2008년 11월 11일 채택)

# Heat Resistant Electromagnetic Noise Absorber Films Using Poly(amide imide)/Soft Magnet Composite

**Ji Eun Han, Byung Kuk Jeon, Bon Jae Goo, Seung Hyun Cho*, Sung Hoon Kim**, Kyung Sub Lee**, Yun Heum Park, and Jun Young Lee†**
*Department of Chemical Engineering, Sungkyunkwan University, Suwon, Gyeonggi-do 440−746, Korea*
**Polymer Technology Institute, Sungkyunkwan University, Suwon, Gyeonggi-do 440−746, Korea*
***R&D Center, Chang Sung Corporation, 11−9 Namdong Industrial Area, Namdong-gu, Incheon 405−846, Korea*
*(Received November 4, 2008; Revised November 11, 2008; Accepted November 11, 2008)*

**초록:** 폴리(아미드−이미드)와 연자성체의 블렌딩에 의해 고온에서 이용 가능한 내열성 전자기 노이즈 흡수용 필름을 제조하였다. *N,N*−디메틸아세트아미드에 폴리(아미드 아믹 산)을 용해시킨 후 연자성체 파우더를 혼합하고 이 용액을 유리 기판에 캐스팅한 뒤 열을 가하여 이미드화하는 방법으로 전자기 노이즈 흡수 필름을 제조하였다. 제조된 필름의 열적 특성, 열 안정성 및 기계적 성질을 분석하고 마이크로 스트립 라인 법에 의해 전자기 흡수력을 측정하였는데, 1 GHz에서 150 μm 두께의 복합체 필름의 전력손실(power loss)은 약 25%였다.

**Abstract:** We fabricated the electromagnetic (EM) noise absorber films for high temperature use by blending a soft magnetic powder with poly(amide imide) (PAI). The EM noise absorber films of PAI/soft magnet composite were prepared by casting the solution of poly(amide amic acid)/soft magnet powder into glass substrate with casting applicator device and then thermal imidization. The obtained films were fully characterized and their physical properties including thermal behavior, thermal stability and mechanical properties were studied. The EM noise absorption ability was also investigated using micro-strip line method. At 1 GHz, the power loss of composite film with 150 μm thickness was about 25%.

**Keywords:** poly(amide imide), soft magnetic, composite films, electromagnetic noise absorption, micro-strip line.

## Introduction

The problems caused by electromagnetic (EM) noise has become more serious due to rapid growth of technologies and broad improvement of electronics industry in GHz frequency range such as mobile phones, local area networks, satellite-based broadcasting, etc.[1,2] There have been many researches about various suppressing and shielding materials focused on the use of standard metals, polymers containing magnetic materials and their composites,[3−10] which have disadvantages such as limited physical flexibility, heavy weight and limited commercial applications under varied temperatures.[11,12] Therefore the EM noise absorbers for the high temperature use are needed. The soft magnetic Fe−Si−Al composites have drawn much attention due to their high permeability, electrical resistivity, low magnetic anisotropy constant and magnetostriction.[13]

Recently, researches about heat resistant polymers have focused on the application of polyimides which are well known for their high performance properties such as high thermal stability and excellent mechanical properties.[14,15] However, it is difficult to process these polymers due to their poor solubility in most organic solvents. In order to overcome these drawbacks, several researches have been proposed

†To whom correspondence should be addressed.
E-mail: jylee7@skku.edu

to attach the flexible segments such as ester, amide, ether or urethane linkages in the polymer backbone, without damaging the thermal stability.[16,17] Poly(amide imide)s(PAI)s are known to have exceptional mechanical, thermal and chemical resistant properties due to synergic properties of both polyamides and polyimides such as high strength, high heat capability, and melt processability. And these polymers are finding applications in various fields, such as nanocomposites, microwave devices, electric wire enamels, aviation industry and military use, etc. Therefore, poly(amide imide) (PAIs) can offer suitable electromagnetic noise absorption materials with thermally stable EM noise absorbing film.

In this study, we report electromagnetic properties of PAI/soft magnet composite film having both extraordinarily high temperature stability and power loss for the purpose of electromagnetic noise absorbers in quasi-microwave frequency of GHz range. The properties of resulting composite films such as thermal stability, surface morphology, mechanical properties and Power loss were investigated. We showed the relationships between the power loss and the composite ratio or thickness of composite films.

## Experimental

Poly(amide amic acid) (PAAA, Kolon Ind. Inc.)/soft magnet

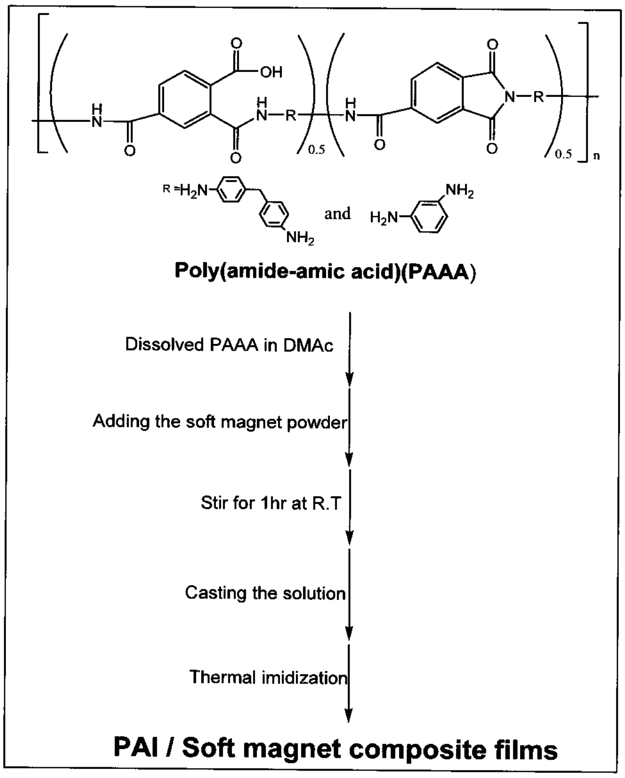


**Figure 1.** Schematic structure and procedures to prepare the PAI/soft magnet composite films.

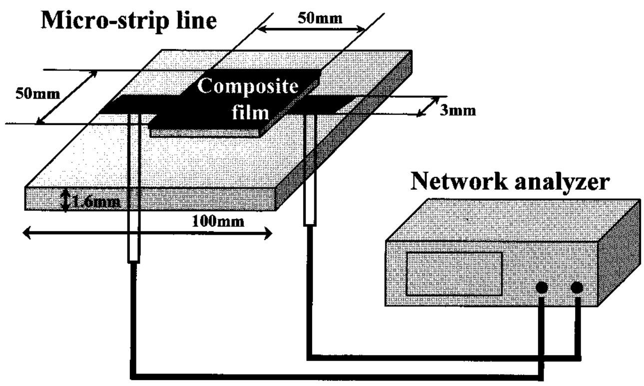


**Figure 2.** Measurement system (MSL).

(SENDUST: Fe-Si-Al alloy flaky powder) composite films were prepared as shown in Figure 1. The PAAA powders were dissolved in organic solvent such as *N,N*-dimethylacetamide (DMAc, Aldrich) to give 33 wt% solution. Composite solutions were prepared by adding the mixture of soft magnet powder into the PAAA solution and stirred for 1 h using ultra-sonification. After this period, PAAA/soft magnet composite solutions were cast into glass substrate (30 cm ×20 cm) using casting applicator device. After the solvent was evaporated in the oven at 100 ℃ and 150 ℃ for 1 h each, PAAA was thermally imidized at 200 ℃ for 1 h and then fully imidized at 250 ℃ for 30 min.

FT-IR spectra of the sample were recorded using Bruker IFS-66/S. Thermal characteristics of the EM noise absorber films were examined by differential scanning calorimeter (DSC, Seiko Exstar 6000) and thermogravimetry analyzer (TGA, Shin Do and Scientific Corp.) at a heating rate of 10 ℃/min. Also the morphology of the EM noise absorber films was analyzed by scanning electron microscopy (SEM, Hitachi).

The elongation at break, tensile strength at break and young's modulus of sample films were determined using universal testing machine (UTM, Instron 4482). The films were cut into uniform shapes according to the ASTM D882-02. The test was carried out at a crosshead speed of 15 mm/min. At least four samples were tested for each formulation.

For the suppression effect of conduction noise, the composite films (50 mm×50 mm, various thicknesses) were set onto micro-strip line (MSL) with a characteristic impedance of 50 Ω as shown in Figure 2.

We measured the scattering parameters corresponding to reflection and transmission in the frequency range from 0.05 to 10 GHz.

## Results and Discussion

The FT-IR spectra of the poly(amide amic acid) (PAAA) and its fully cured sample are shown in Figure 3. The characteristic peaks of PAAA at 3426 $cm^{-1}$ (O−H, carboxylic acid), at 1720 and 1657 $cm^{-1}$ (C=O stretchings, carboxylic acid (C=O−OH) and amide (C=O−NH) groups) are shown. The spectrum of PAI exhibited all characteristic peaks of both imide and amide, including the peaks at 1780 and 1722 $cm^{-1}$ (symmetric and asymmetric C=O stretchings, imide group), at 1667 $cm^{-1}$ (C=O stretching, amide group), at 1382 $cm^{-1}$ (C−N stretching, imide ring), at 727 $cm^{-1}$ (C=O bending, imide group) and at 3309 $cm^{-1}$ (N-H stretching, amide group).

The DSC thermograms shown in Figure 4 represent $T_g$ of PAAA as 207.04 ℃, however $T_g$ of PAI, which is normally about 277~326 ℃, seems higher than 300 ℃ since there is no peak shown until 300 ℃ for the PAI curve.[18]

The thermal stability of the pristine PAI and PAI/soft magnet composite films were characterized by TG analysis conducted at a heating rate of 10 ℃/min under nitrogen as shown in Figure 5. Both films show similar initial decomposition temperature ($T_0$) and temperature for 10% weight loss ($T_{10}$) that is important criterion for evaluation of thermal stability behavior. The $T_{10}$ values for both films are in the range of 478−484 ℃ and char yields of PAI film at 700 ℃ are above 57.5% and those of PAI/soft magnet composite film are about 65% due to the existence of soft magnet. The similar thermal stabilities of PAI films and PAI/soft magnet composite films mean that the feature of PAI has not been damaged during composite fabrication.

Figure 6 is a cross sectional surface morphology of composite film obtained by SEM. The soft magnet particles were uniformly aligned by mixing in the direction perpendicular to the eddy current. Thus, soft magnet particles in the direction parallel to the electromagnetic field increase the magnetized density and its anisotropic structure will increase the shielding property of the composite films.[19,20]

Table 1 shows the mechanical properties of composite film using UTM to measure a tensile strength at break, elongation at break and mean modulus, $E$. The values of tensile strength at break were from 89.99 to 118.58 MPa according to the content of soft magnet powders.

We confirm that the tensile strength and elongation at break

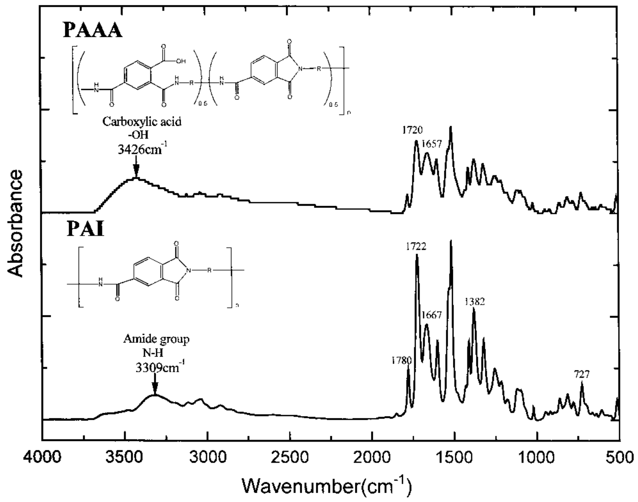


**Figure 3.** FT−IR spectra of the PAAA and PAI.

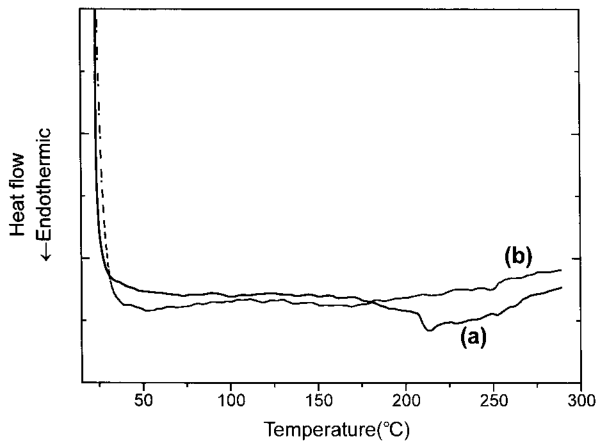


**Figure 4.** DSC curves of (a) PAAA and (b) PAI.

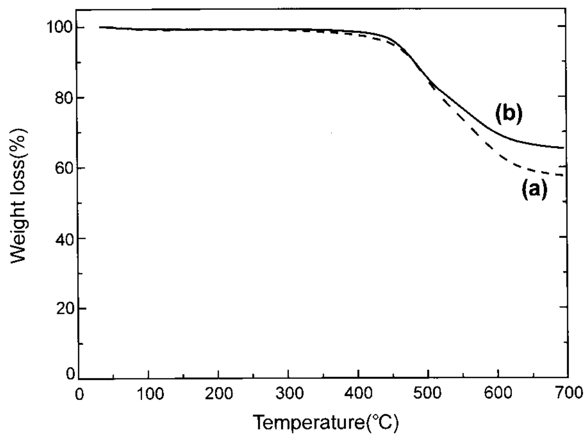


**Figure 5.** TGA thermograms of (a) PAI and (b) PAI/Soft magnet composite film.

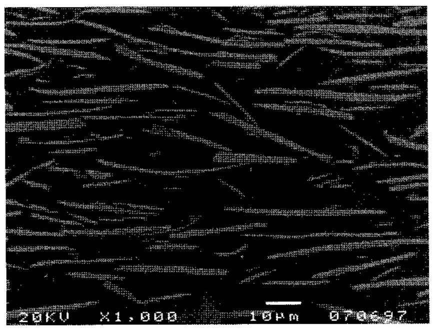


**Figure 6.** SEM photographs (×1000) of the composite film in cross section.

**Table 1. Mechanical Properties of Composite Films according to Soft Magnet Powder Contents**

| Films | Content of soft magnetic (%) | Elongation at break (%) | Tensile strength at break (MPa) | Mean modulus[a] (GPa) |
|---|---|---|---|---|
| A[b] | 0 | 9.80 | 118.58 | 1.21 |
| B | 20 | 5.03 | 91.90 | 1.83 |
| C | 50 | 2.73 | 89.99 | 3.29 |

[a]Mean modulus=Breaking stress/Breaking strain. [b]Obtain from Kolon Onlymide AP grade data sheet.

of the composite films decreased with increasing content of soft magnet powders and resulting composite films (B, C) were more stiff and rigid than the pristine PAI film (A). With increasing content of soft magnet powders, young's modulus of composite films increased.

Although the tensile strength and elongation at break of PAI/soft magnet composite films are lower compared to the pristine PAI film, they are much higher than those of general polymer films (PET, polystyrene, polycarbonate, etc.),[21] which means that the EM noise absorber films with high mechanical properties have been successfully manufactured.

To investigate the electromagnetic properties, the noise suppression effect is evaluated with the power loss ($P_{loss}/P_{in}$) as given in Eq. (1),[22]

$$P_{loss}/P_{in}=1-(S_{11}^2-S_{21}^2) \quad (1)$$

where $S_{11}$ and $S_{21}$ are reflection coefficient and transmission coefficient, respectively, which are determined from network analyzer.

Figure 7 shows the dependency of power loss on the thickness of composite film when the mixing ratio of soft magnet powders to PAI is 50 : 50 wt%. At 1 GHz, the power loss of PAI/soft magnet composite film with 50 μm and 150 μm are 15.9% and 25.27%, respectively. The value of power loss increased with increasing composite film thickness. The power loss corresponding to electromagnetic absorption loss at both near and far field is given by Eq. (2),

$$\text{Power loss} \propto t\,(f\mu_r\,\sigma_r)^{1/2} \quad (2)$$

The $t$, $\mu_r$, $\sigma_r$ and $f$ denote film thickness, relative magnetic permeability, relative conductivity, and frequency, respectively. According to the Eq. (2), the power loss is proportional to film thickness, $t$, and to square root of $\mu_r$ and $\sigma_r$. Since $\mu_r$ and $\sigma_r$ are nearly same for all the composite films, only the film thickness, $t$, affects the power loss. Therefore the differences in film thickness have effect on the electromagnetic absorption causing the difference in power loss.

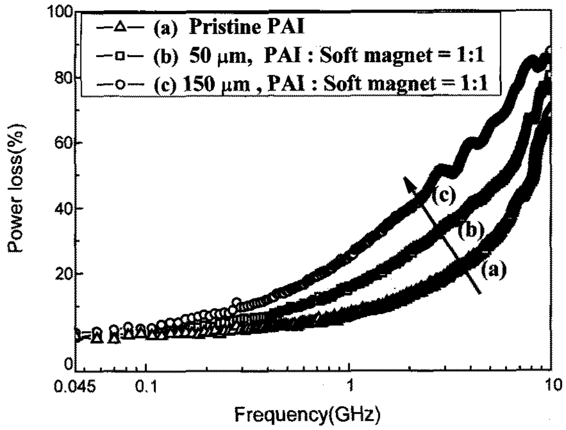


**Figure 7.** The power loss for PAI−soft magnet composite films with film thickness.

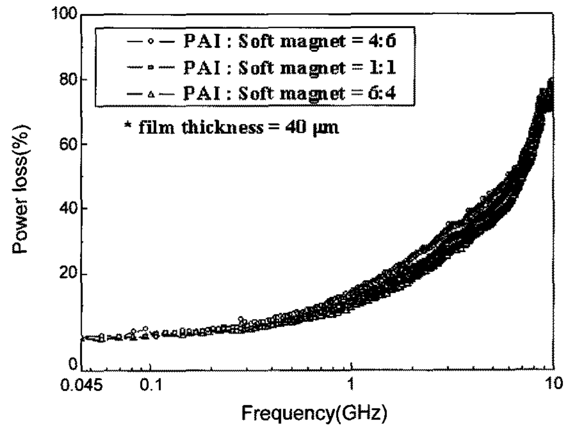


**Figure 8.** The power loss for PAI− soft magnet composite films with composite ratio.

The effect of mixing ratio of composite film is shown in Figure 8. When the thickness of composite films is 40 μm, the power loss of composite with 60 wt% soft magnet is 14.16%, that with 50 wt% soft magnet is 11.69% and that with 40 wt% soft magnet is 9.81% at 1 GHz, respectively. Namely, increasing the content of soft magnetic powder increased the value of the power loss. We believe that these results are caused by the characteristics of electromagnetic absorption of soft magnetic powder and therefore the composite films with high content of soft magnet powder show excellent electromagnetic absorption for electromagnetic shielding.

## Conclusions

We developed a new technique to fabricate the electromagnetic noise absorber film by blending a soft magnet powder with PAI.

The results obtained are concluded as follows:

1) The high $T_g$, $T_{10}$ and char yields values, high thermal stability and good mechanical properties of the fabricated EM noise absorber films make these films as promising processable EM noise absorber materials.

2) The power loss of PAI/soft magnet composite films has a strong dependency of the film thickness and composite ratio at 0.045−10 GHz range. The composite films with controlled composite ratio and thickness have approximately 25% power loss at 1 GHz and can be widely applicable as an electromagnetic shielding field especially where high temperature is needed.

**Acknowledgments:** This work was supported by Chang Sung Corporation Grant No. 2006−0904. This work was supported by the Korea Research Foundation Grant Funded by the Korean Government (MOEHRD) (KRF−2007−511−D00068).

## References


1. M. Mitial and B. K. Guha, *Chem. Eng. Technol.*, **16**, 325 (1993).
2. P. Chandrasekhar and K. Naishadham, *Synth. Met.*, **105**, 115 (1999).
3. M. Matsumoto and Y. Miyato, *IEEE Trans. Magn.*, **33**, 4459 (1997).
4. A. Tsaliovich, *Electromagnetic Shielding Handbook for Wired and Wireless EMC Applications*, Kluwer Academic, Boston, 1999.
5. J. W. Molyneux−Child, *EMC Shielding Materials*, Newnes, Oxford, 1997.
6. O. Hashimoto, *Recent Technologies for New Wave Absorber and its Applications*, Shiemushi, Japan, 1999.
7. K. Ishino and Y. Narumity, *Am. Ceram. Soc. Bull.*, **66**, 1469 (1987).
8. K. M. Lim, K. A. Lee, M. C. Kim, and C. G. Park, *J. Non−Crys. Solids*, **351**, 75 (2005).
9. J. R. Liu, M. Itoh, J. Jiang, and K. Machida, *J. Magn. Magn. Mater.*, **277**, 251 (2004).
10. S. S. Kim, S. T. Kim, J. M. Ahn, and K. H. Kim, *J. Magn. Magn. Mater.*, **271**, 39 (2004).
11. S. T. Shinn and Y. C. Fa, *Mat. Sci. Eng. A*, **302,** 258 (2001).
12. J. Joo and A. J. Epstein, *Appl. Phys. Lett.*, **65**, 18 (1994).
13. A. H. Kasama, C. Bolfarini, C. S. Kiminami, and W. J. Botta Filho, *Mat. Sci. Eng. A*, **449-451**, 375 (2007).
14. D. Wilson, H. D. Stenzenberger, and P. M. Hergenrother, Editors, *Polyimides*, Blackie, Glasgow and London, 1990.
15. M. K. Ghosh and K. L. Mittal, Editors, *Polyimides: Fundamentals and Applications*, Marcel Dekker, New York, 1996.
16. G. Liou, M. Maruyama, Y. Kakimoto, and Y. Imai, *J. Polym. Sci. Part A: Polym. Chem.*, **36**, 2021 (1998).
17. S. Mehdipour−Ataei and S. Keshavarz, *J. Appl. Polym. Sci.*, **89**, 2567 (2003).
18. D. Liaw, P. Hsu, W. Chen, and S. Lin, *Macromolecules*, **35**, 4669 (2002).
19. B. Lee, J. Lee, Y. Yun, I. Jeong, and J. Moon, *Mat. Sci. Forum.*, **534-536**, 1449 (2007).
20. K. S Lee, Y. C. Yoon, G. B. Choi, S. S. Kim, and J. Y. Lee, *J. Kor. Electr. Eng. Soc.*, **16**, 228 (2005).
21. J. Brandup and E. H. Immergut, *Polymer Handbook*, 2nd Ed., Wiley, New York, 1975.
22. S. Ohnuma, H. Nagura, H. Fujimori, and T. Masumoto, *IEEE Trans. Magn.*, **40**, 2712 (2004).