

Radiation-Induced Grafting of Glycidyl Methacrylate onto High-Density Polyethylene (HDPE) and Radiation Lamination of HDPE

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Received 12 April 2004; accepted 2 August 2004

DOI 10.1002/app.21508

Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Radiation-induced grafting of glycidyl methacrylate (GMA) onto high-density polyethylene (HDPE) and the radiation lamination of HDPE by bulk grafting of GMA were reported. The effects of irradiation dose, monomer concentration, and atmosphere on grafting were investigated. The extent of grafting initially increased with irradiation dose and then remained almost constant. The extent of grafting was higher in 2M GMA than in 1M GMA at the same irradiation dose. The extent of grafting in nitrogen was higher than that in air. The grafted samples were characterized with FTIR spectrometry and thermogravimetric (TG) analysis. A carbonyl group was found on grafted HDPE samples, and the carbonyl index increased with the extent of grafting. TG analyses proved

the existence of grafted materials on HDPE and the grafted GMA thermally decomposes at a temperature lower than that of HDPE. Strong adhesion could be obtained with radiation lamination of HDPE by bulk grafting of GMA. Benzophenone facilitates the grafting in a proper concentration range. The adhesion mechanism of the laminated samples was the entanglement of the grafted chains. © 2005 Wiley Periodicals, Inc. *J Appl Polym Sci* 96: 772–779, 2005

Key words: graft copolymers; high-density polyethylene (HDPE); adhesion; radiation lamination; thermogravimetric analysis (TGA)

INTRODUCTION

Polyethylene (PE) is one of the most widely used polymer materials because it has many good properties, such as high chemical resistance, high impact strength, and flexibility with low cost. However, the surface of PE is chemically inert in nature and its surface energy is low, which restrict practical applications of PE materials.^{1,2}

Radiation-induced grafting is a very common and powerful technique to modify the physicochemical (e.g., wettability, adhesion, adsorption, and surface reaction) properties of PE, because of its significant advantages over other methods, such as ambient reaction temperature, high penetration of polymer matrix, and so on.^{3–5}

Yang and Rånby developed a method for lamination of polymer films by a bulk (no solvent) surface photografting process, in which a thin layer of the monomer containing dissolved photoinitiator was sandwiched between two thin films. Most important, photolamination occurred simultaneously during the photografting process, resulting in good adhesion between the two films.^{6–10} This photolamination tech-

nique can be applied to a wide variety of plastic films and it gives laminates of high mechanical strength and high and selective barrier properties for different gases and vapors. However, this method has an unavoidable drawback: it can be used only for thin and UV transparent films.

High-energy irradiation such as gamma irradiation can penetrate thick polymeric materials, whether they are or are not transparent. Therefore if we use high-energy irradiation as the radiation source, then the shortcoming of photolamination can be overcome. Here we use a new term, "radiation lamination," to describe the lamination of polymeric materials by high-energy irradiation-induced bulk grafting.

In the former work,^{8,11} very strong adhesion could be obtained when acrylic acid (AA) and a combination of AA and glycidyl acrylate (GA) were used as the monomer(s) to photolaminate low-density (LDPE) and high-density polyethylenes (HDPE). However, there was no adhesion at all when AA was used as the monomer to laminate HDPE under γ -irradiation. Strong adhesion could be obtained when using glycidyl methacrylate (GMA) as the monomer to laminate HDPE by γ -irradiation-induced bulk grafting.

GMA has shown good potential as a reactive monomer for polyolefin modification. The grafting of GMA onto several polymers has been done with different initiation methods. For example, the thermal photografting of GMA onto PE and polytetrafluoroethyl-

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ene was initiated by the peroxide formed on the PE surface by Ar plasma¹²⁻¹⁴ and corona¹⁵ pretreatment, the grafting of GMA onto poly(vinyl chloride),¹⁶ polypropylene (PP),¹⁷ and cellulose¹⁸ initiated by peroxide. In recent years, the melt grafting or the reactive extrusion grafting of GMA onto polyolefins (PE, PP) has attracted substantial attention.¹⁹⁻²⁷

Work done on the radiation-induced grafting of GMA is quite rare.^{28,29} To our knowledge, no work has been done on the surface modification of HDPE for adhesion enhancement by radiation-induced grafting of GMA. In this study, the radiation-induced grafting of GMA in acetone onto HDPE and the radiation lamination of HDPE by mutual bulk grafting of GMA were carried out.

EXPERIMENTAL

Materials

HDPE (melt flow index: 0.39 g/10 min; density: 0.949g/cm³) was supplied by Nova Chemicals Ltd. (Sania, Ontario, Canada). The HDPE film was cut into 10 × 20 mm (~ 0.3 mm in thickness, for grafting study) and 20 × 70 mm (~ 0.6 mm in thickness, for radiation lamination) rectangle samples, and then subjected to Soxhlet extraction with acetone for 24 h to remove impurities and additives before use.

Monomer glycidyl methacrylate (GMA, 97%) was from Mitsubishi Gas (Tokyo, Japan). The photoinitiator benzophenone (BP, chemically pure grade) was from Yuanhang Chemicals Co. Ltd. (Tianjin, China) and solvent acetone (AR grade) was from Beijing Beihua Fine Chemicals Co. Ltd. (Beijing, China). All the chemicals were used without further purification.

Grafting

Polyethylene samples were irradiated in a ⁶⁰Co gamma chamber. The dose rate was determined by a Fricke dosimeter.

Because it is not easy to quantitatively study the bulk grafting of GMA that occurs in radiation lamination, the grafting of GMA in acetone solution was carried out to understand the grafting process. The PE samples were placed in test tubes for radiation experiments, and then 5 mL GMA solution was added to each test tube, after which the test tubes were sealed. For the study of the effect of atmosphere on grafting, some of the solutions were deaerated with nitrogen two times (30 min each time) to remove oxygen dissolved in solution before irradiation. All the samples were irradiated with different doses at a dose rate of 213 Gy/min. The reaction temperature was not strictly controlled. Room temperature was taken as the reaction temperature, which varied with seasons from 10 to 35°C. After grafting, the samples were Soxhlet ex-

tracted with acetone for at least 48 h to remove unreacted monomer and homopolymer, and then dried at 50°C for 24 h.

The extent of grafting was expressed as the weight increase per surface area of the sample, and was calculated from the following equation:

$$G = \frac{W_g - W_0}{S} \quad (\mu\text{g}/\text{cm}^2)$$

where W_g and W_0 are the weights of the PE sample after and before grafting, respectively, and S is the surface area of the PE sample.

The average value of five samples was taken as the final extent of grafting. Typically, the five samples gave values within 10%.

Radiation lamination

The assembly for the radiation lamination experiment was similar to that used by Yang and Rånby.⁶ The monomer GMA, with or without benzophenone, was coated onto one HDPE sample, and then the other HDPE film sample was placed on top. The sandwich was then pressed with suitable pressure to spread the solution into an even and thin liquid layer. After irradiation, the samples were dried at 50°C for 24 h.

FTIR characterization

FTIR spectra were obtained from pristine HDPE film and HDPE films grafted with GMA on an Avatar-360 spectrometer (Nicolet Analytical Instruments, Madison, WI). The number of scans was 32 at a resolution of 4 cm⁻¹.

The carbonyl index (CI) was calculated using the following equation:

$$\text{CI} = A_{\text{C=O}}/A_{\text{C-H}}$$

where $A_{\text{C=O}}$ is the absorbance of the carbonyl C=O stretching vibration peak at about 1731 cm⁻¹, which is the characteristic peak of carbonyl group in GMA, and $A_{\text{C-H}}$ is the absorbance of the C-H out-of-plane vibration at 908 cm⁻¹ of polyethylene units, used as the internal standard peak.³⁰ These peaks were auto-baselined before calculating the absorbance.

Thermogravimetric analyses

The samples for thermogravimetric analysis were dried before use. Thermogravimetric analyses were carried out in a TG 50 of a TA-3000 thermoanalysis system (Mettler International, Zurich, Switzerland). The purge gas was N₂, the flow rate was 10 mL/min, and the heating rate was 20°C/min. The STEP analysis

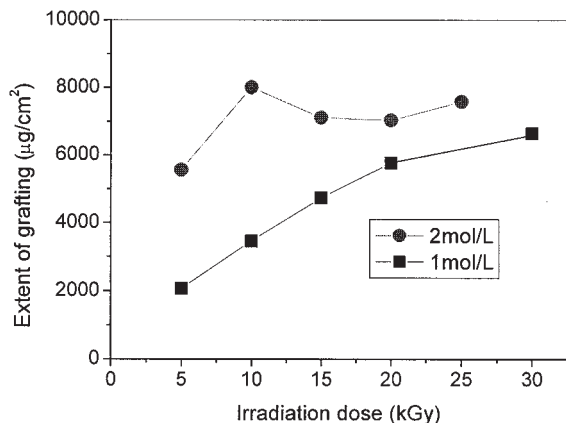


Figure 1 Extent of grafting as a function of irradiation dose. Grafted in nitrogen; reaction temperature, 30–35°C.

of each sample was carried out. The TG and differential thermogravimetric (DTG) curves of the samples were also obtained. Relative thermal stability of the grafted and pristine HDPE samples was evaluated by comparison of the decomposition temperature T_{sr} , T_{pr} and T_{er} , where T_{sr} , T_{pr} and T_e are the temperatures at which samples start to decompose, decompose most quickly, and the end of decomposition, respectively.

Peel test

Peel strength was measured at room temperature by a 180° T peel test using a CSS-2202 electronic universal testing machine (Changchun Institute of Experimental Machines, Chang Chun, China) with a crosshead speed of 50 mm/min. The average force after the initial peak load was taken as the peel strength. The peel strength is expressed in N/m.

RESULTS AND DISCUSSION

Effect of the GMA concentration and reaction temperature

Figures 1 and 2 show the results of the radiation grafting carried out in 1 and 2 mol/L GMA acetone solutions. The solutions were deaerated by bubbling nitrogen. The results shown in Figure 1 were obtained at 30–35°C and those in Figure 2 were obtained at 10–15°C.

Different from the low reactivity of GMA in the graftings initiated by other methods, the γ -ray irradiation-induced grafting of GMA onto HDPE occurs very easily.

As a general rule, the extent of grafting initially increased quickly with the total irradiation dose and then remained almost constant. As shown in Figure 1, for the grafting carried out in 1 mol/L GMA acetone solution, the extent of grafting was about 2000 $\mu\text{g}/\text{cm}^2$

after just 5 kGy irradiation. It increased almost linearly with irradiation dose up to 20 kGy, at which the extent of grafting was about 6000 $\mu\text{g}/\text{cm}^2$ and then increased slowly; the extent of grafting after 30 kGy irradiation was about 6500 $\mu\text{g}/\text{cm}^2$. For the grafting carried out in 2 mol/L GMA acetone solution, the extent of grafting reached about 5500 $\mu\text{g}/\text{cm}^2$ after just 5 kGy irradiation and reached about 8000 $\mu\text{g}/\text{cm}^2$ after 10 kGy irradiation. Then the extent of grafting decreased slightly and remained constant. At the same irradiation dose, the extent of grafting of the sample grafted in 2 mol/L GMA solution is higher than that in 1 mol/L GMA solution, especially when the irradiation dose is low. Therefore, the monomer concentration has a significant effect on the extent of grafting.

As shown in Figure 2, the situation for the grafting carried out at 10–15°C is quite similar to that at 30–35°C. The extent of grafting increased very quickly with irradiation dose up to 10 kGy and then increased slowly or remained almost constant. Also at the same irradiation dose, the extent of grafting of the sample grafted in 2 mol/L GMA solution is higher than that in 1 mol/L GMA solution. When the irradiation dose was <10 kGy, at the same irradiation dose, the extent of grafting of the HDPE sample grafted in 2 mol/L GMA solution was approximately twice that of the sample grafted in 1 mol/L GMA solution.

The obvious difference between the grafting carried out at 30–35°C and that carried out at 10–15°C is that the extent of the grafting is higher at the higher reaction temperature than that at the lower temperature at the same irradiation dose, which indicates that the reaction temperature also affects the radiation-induced grafting of GMA onto HDPE. A higher reaction temperature is beneficial to the grafting.

The effect of monomer concentration and reaction temperature on the radiation-induced grafting of GMA onto HDPE could be explained as follows:

As a general rule, the grafting reaction rate increases with the increase of monomer concentration. From the

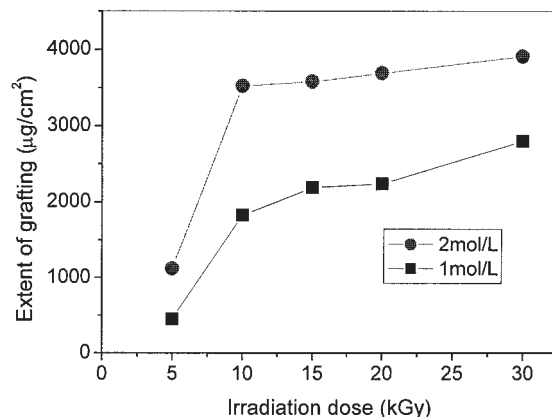


Figure 2 Extent of grafting as a function of irradiation dose. Grafted in nitrogen; reaction temperature, 10–15°C.

perspective of kinetic of polymerization, the propagation rate of grafting chain can be expressed as

$$R_p = k_p[M \cdot][M]$$

where k_p is the rate constant of propagation, $[M]$ is the monomer concentration, and $[M \cdot]$ is the concentration of primary free radical formed on the HDPE surface.

All the samples were irradiated at the same position of the radiation chamber, where the dose rate was the same. As we know, the concentration of macromolecular free radicals formed on the HDPE surface, which can initiate grafting, is dominated by the radiation dose rate, which means that the $[M \cdot]$ should be the same in all of the experiments. Then R_p is determined by the molar concentration of monomer $[M]$ and the propagation rate constant k_p . When the reaction temperature is the same and k_p is a constant, then R_p is proportional to $[M]$. So when $[M]$ is higher, then R_p is also higher.

$$R_p \propto [M]$$

The extent of grafting (G) should be proportional to the propagation rate R_p and the reaction time t :

$$G \propto R_p t$$

$$G \propto [M] t$$

and at the same irradiation time

$$G \propto [M]$$

The extent of grafting (G) is proportional to the monomer concentration at the same irradiation time.

When the monomer concentration $[M]$ and the concentration of macromolecular free radicals $[M \cdot]$ are the same, the change of reaction temperature will also affect the grafting. Because k_p usually increases with the increase of temperature, the increase of reaction temperature usually leads to the increase of grafting speed and the extent of grafting.

Effect of atmosphere

Reaction atmosphere also has a significant effect on the radiation-induced grafting of GMA onto HDPE. As shown in Figure 3, at the same irradiation dose, the extent of grafting of the sample grafted in 1 mol/L GMA acetone solution deaerated with nitrogen is higher than that of the sample grafted in an undeaerated solution. The effect of oxygen on radiation-induced grafting has been well studied. The oxygen molecules dissolved in solution can terminate the growing grafting chains, thus leading to the low ex-

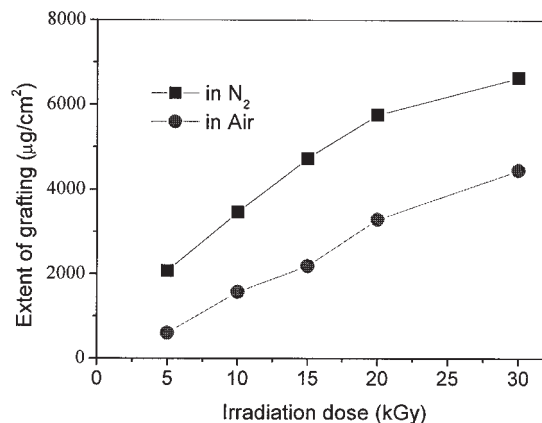


Figure 3 Extent of grafting as a function of irradiation dose for the graftings carried out in air and in nitrogen. 1 mol/L GMA acetone solution; reaction temperature, 30–35°C.

tent of grafting. Oxygen acts as an inhibitor in the grafting process.

FTIR characterization

Because the surface of the grafted films with high extent of grafting is too rough for infrared rays to penetrate, only the FTIR spectra of the grafted films with low extent of grafting could be obtained. The samples used were grafted at 10–15°C in nitrogen and in air.

The grafting of GMA onto HDPE was confirmed by FTIR spectroscopy, as illustrated in Figure 4(a) for pristine HDPE film and in Figure 4(b) and (c) for the grafted HDPE films. Figure 4(a) shows the typical HDPE spectrum without an absorption band near 1731 cm^{-1} , which is the characteristic absorption band of the carbonyl group (C=O). In the FTIR spectrum of the grafted sample [Fig. 4(b) and (c)] an absorption band occurring at about 1731 cm^{-1} appears, demonstrating that radiation-induced grafting of GMA on HDPE films has taken place.

The calculated carbonyl index (CI) of the HDPE sample grafted with 450 $\mu\text{g}/\text{cm}^2$ GMA is 0.56 and the CI of the HDPE sample grafted with 900 $\mu\text{g}/\text{cm}^2$ GMA is 0.69.

Thermogravimetric analyses

The TG and DTG curves of pristine HDPE and HDPE grafted with GMA are shown in Figure 5. The thermogravimetric analysis results of pristine HDPE and three HDPE samples grafted with GMA are listed in Table I.

There is only one decomposition peak in the DTG curve of pristine HDPE, whose T_s is 316°C, T_p is 486°C, and T_e is 518°C. However, there are two decomposition peaks in the DTG curve of HDPE

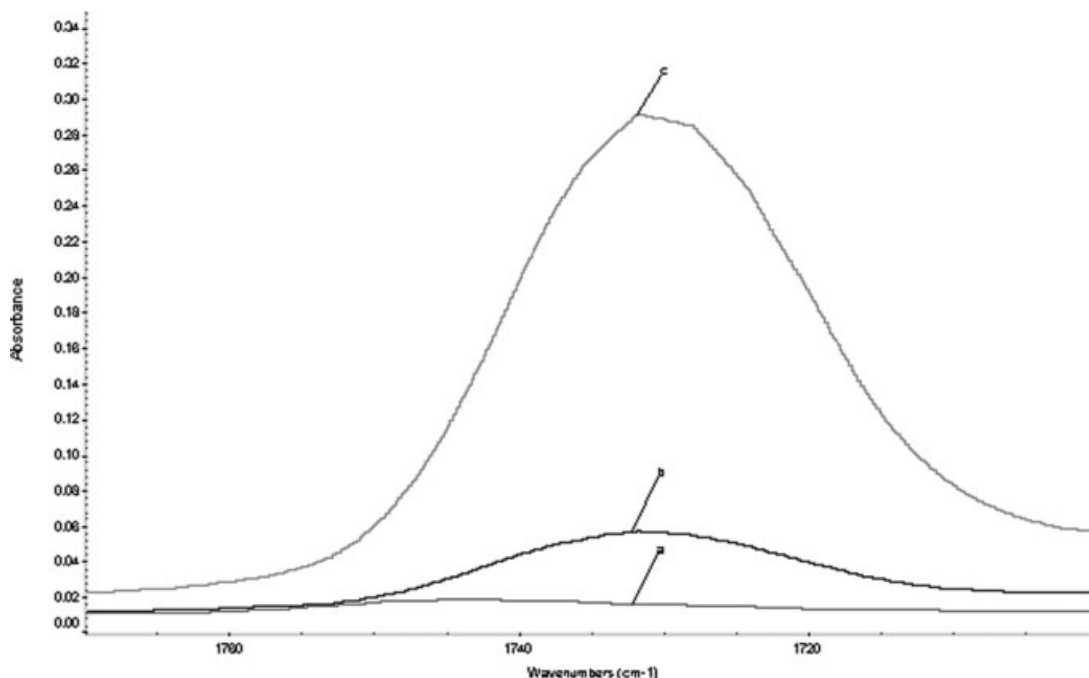


Figure 4 FTIR spectra of (a) pristine HDPE film, (b) HDPE film grafted with $450 \mu\text{g}/\text{cm}^2$ GMA, and (c) HDPE film grafted with $900 \mu\text{g}/\text{cm}^2$ GMA.

grafted with GMA. For the sample grafted with $1500 \mu\text{g}/\text{cm}^2$ GMA, in the first decomposition peak, T_s is 227°C , T_p is 301°C , and T_e is 350°C ; and in the second decomposition peak, T_s is 350°C , T_p is 486°C , and T_e is 518°C . In the second decomposition step, the T_p and T_e were almost the same as those of pristine HDPE, and thus can be attributed to the decomposition of HDPE. So, the first decomposition peak should be attributed to the decomposition of grafted GMA. The material loss of the first decomposition peak obtained from STEP analysis is 7.6%, which is a little lower than the calculated percentage grafting (8.4%) of this sample. This further proves that the first decomposition peak is attributed to the decomposition of grafted GMA. Similarly, for other grafted samples tested, the first decomposition peak is attributed to the decomposition of grafted GMA and the second one is attributed to the decomposition of HDPE.

There is an obvious increase in the T_p and T_e of the thermal decomposition of grafted GMA with increasing extent of grafting. The T_p values of the samples grafted with 1500, 3250, and $8250 \mu\text{g}/\text{cm}^2$ were 301, 312, and 321°C , separately; and the T_e values of these samples were 350, 367, and 382°C , separately. Therefore, the thermal stability of the grafted GMA increases with the extent of grafting. The reason is still unknown, however; possibly it is caused by the grafting that occurred in the inner portions of the HDPE sheets.

Peel test

When using bulk GMA, the grafting can be studied quantitatively in only a short irradiation time because liquid GMA can be polymerized to solid polymer quickly under irradiation. Therefore, the bulk grafting of GMA onto HDPE was not studied. However, from the results of the grafting of GMA in acetone solution, it can be deduced that the bulk grafting of GMA onto HDPE is faster than that carried out in acetone solution because of the higher monomer concentration of bulk GMA.

In the radiation-induced grafting of GMA in acetone solution, no photoinitiator or any other additives were used. In the radiation lamination, benzophenone (BP) was used in some of the bulk grafting systems.

Strong adhesion could be obtained when two HDPE films were radiation laminated by the bulk grafting of GMA with or without BP. Figure 6 shows the peel test results of HDPE samples radiation laminated by GMA without or with 4% BP by weight.

Quite similar to that reported in the photolamination of HDPE by bulk grafting of acrylic acid (AA),¹¹ as shown in Figure 6, the adhesion of the HDPE samples radiation laminated by bulk grafting of GMA jumps from zero to maximum in a very short time. There was no adhesion before the total conversion of liquid GMA to solid poly-GMA during the radiation lamination of HDPE films. Without BP, there was no adhesion when the irradiation dose was less than 15 kGy. The peel strength jumped from zero at 15 kGy to

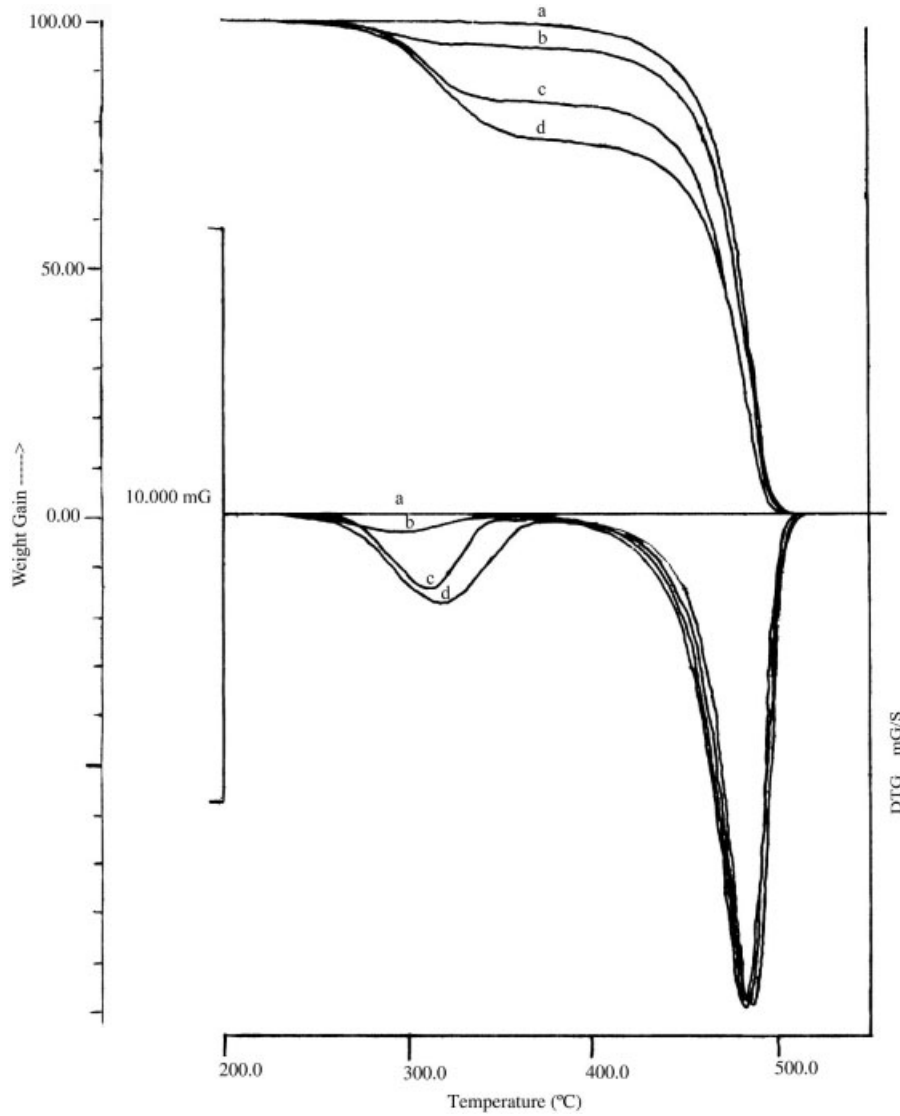


Figure 5 TG and DTG curves of (a) pristine HDPE and HDPE grafted with (b) 1500 $\mu\text{g}/\text{cm}^2$, (c) 3250 $\mu\text{g}/\text{cm}^2$, and (d) 8250 $\mu\text{g}/\text{cm}^2$ GMA.

2650 N/m at 18 kGy and then remained constant. When there was 4% BP in GMA, the maximum peel strength appeared at a lower irradiation dose (11.7 kGy). In other words, less irradiation time is needed to achieve strong adhesion when using 4% BP in the bulk grafting. The maximum peel strengths of the HDPE

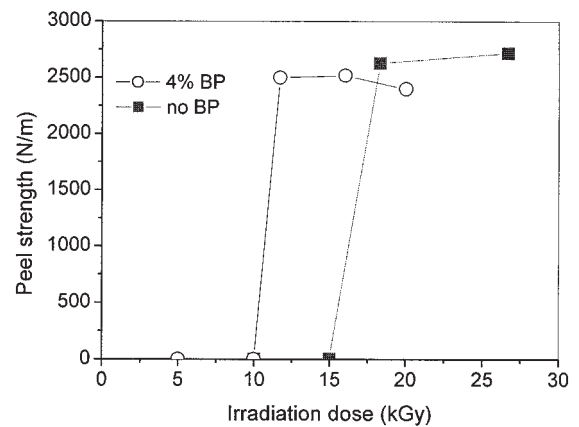


Figure 6 Peel strength of HDPE samples radiation laminated by GMA with 4% or without BP as a function of irradiation time.

TABLE I
 T_s , T_p , and T_e of Pristine and Grafted HDPE Samples

Sample	Peak 1			Peak 2		
	T_s	T_p	T_e	T_s	T_p	T_e
Pristine HDPE				316	486	518
1500 $\mu\text{g}/\text{cm}^2$ (in air)	227	301	350	350	486	518
3250 $\mu\text{g}/\text{cm}^2$ (in air)	244	312	367	367	484	515
8250 $\mu\text{g}/\text{cm}^2$ (in N_2)	229	321	382	382	485	519

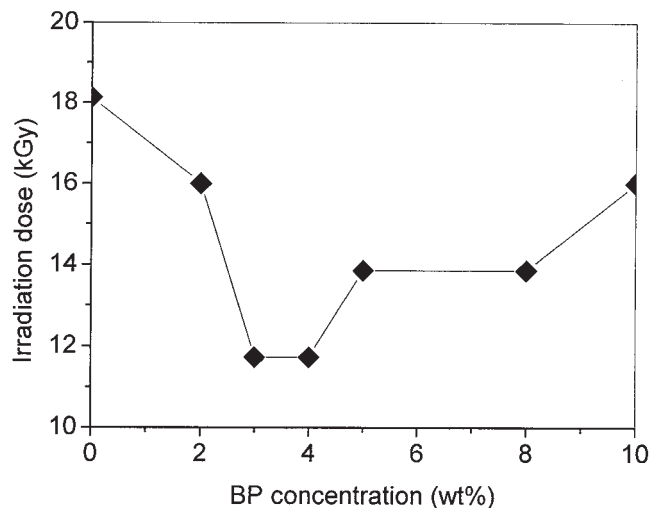


Figure 7 Effect of BP concentration on the minimum irradiation dose.

samples radiation laminated by bulk grafting of GMA without BP or with 4% BP are almost the same.

The preceding results show that photoinitiator BP has a positive effect on the radiation lamination of HDPE by bulk grafting of GMA when the concentration of BP is appropriate. The effect of BP concentration on the minimum irradiation dose required to achieve maximum peel strength is shown in Figure 7.

As shown in Figure 7, the minimum irradiation dose required to achieve maximum peel strength initially decreases with BP concentration up to 3%. When there is no BP, at least 18.1 kGy is required to achieve maximum peel strength. Only 11.7 kGy is required to achieve maximum peel strength when the BP concentration is 3 or 4%. When the BP concentration is >4%, the minimum irradiation dose increases with increasing BP concentration. The appropriate concentration of photoinitiator is between 3 and 4%.

A photoinitiator is usually not required in radiation-induced grafting because high-energy irradiation acts as a photoinitiator itself. Benzophenone is a photoinitiator commonly used in surface photografting. It absorbs UV light in the range of 200–360 nm. When BP absorbs a UV photon, it is excited to the short-lifetime singlet state and then relaxes to a more stable triplet state. Benzophenone in the triplet state can abstract a hydrogen atom from a polymer substrate (PH) and create an active site for surface graft polymerization. In this work, however, it cannot function as that under UV light. As well known, excited state(s) or superexcited state(s) of molecules can be formed under high-energy irradiation. Possibly, the energy of the excited state(s) of GMA can be transferred to BP, BP is then excited to a singlet state, and then relaxes to a triplet state. BP in the triplet state abstracts hydrogen from the HDPE surface and forms a macromolecular free

radical that can initiate the grafting of GMA onto HDPE. Therefore, a greater number of graftings can occur on the HDPE surface, which is favorable for adhesion. However, the excited BP can also abstract hydrogen from the grafted poly-GMA chain, leading to branching of the grafted chain. It can abstract hydrogen even from a monomer molecule, leading to the homopolymerization of GMA. In addition, the hydrogen free radicals formed and the growing homopolymer free radicals can terminate the growing grafting chains. These factors have a negative effect on adhesion. Therefore, a proper BP concentration is very important to balance the positive and negative effects.

Discussion of possible adhesion mechanism

The adhesion mechanism of photolamination was studied by Yang⁸ and Wang.¹¹ Yang and Rånby⁸ suggested that the lamination method involved the formation of hyperbranched graft macromolecules and a crosslinked macromolecular network obtained by adding multifunctional monomers. Wang and Brown¹¹ studied the photolamination of thicker HDPE strips and proposed that the stronger adhesion is generated by a less-branched grafted-chain structure that permits chain entanglement.

There are two factors affecting the adhesion strength of the radiation laminated HDPE samples by bulk grafting: the grafting density and the microstructure of grafted chains.

Because of the high penetration capacity of γ -ray irradiation, there is only a slight difference in the irradiation intensity between the two surfaces to be grafted and laminated. Therefore, the grafting density on both surfaces should be the same. During the peel test of the HDPE samples radiation laminated by the bulk grafting of GMA, the failure mode of the adhesion is mainly breakage of the grafted chains at the position where they are entangled rather than on the PE surface, which indicates that the grafting density is sufficiently high.

After immersion in acetone for about 1 week, the radiation laminated HDPE samples became separated automatically and there was no longer any adhesion, which means that the adhesion between the two laminated HDPE samples is caused by the entanglement rather than the crosslinking of the grafted chains.

CONCLUSIONS

The radiation-induced grafting of glycidyl methacrylate (GMA) in acetone solution onto high-density polyethylene (HDPE) occurs very easily. Radiation grafting was affected by many factors, such as irradiation dose, monomer concentration, reaction temperature and atmosphere, and so on. FTIR and thermo-

gravimetric analyses proved the existence of grafted GMA onto HDPE.

A new method of radiation lamination of high-density polyethylene (HDPE) by bulk grafting has been developed. Strong adhesion could be obtained with radiation lamination of HDPE by bulk grafting of GMA. Possibly, the radiation lamination can be applied to a variety of polymers.

References

1. Chan, C. M. *Polymer Surface Modification and Characterization*; Hanser: Munich, Germany, 1994.
2. Uchida, E.; Ikada, Y. *Curr Trends Polym Sci* 1996, 1, 135.
3. Chapiro, A. *Radiat Phys Chem* 1977, 9, 55.
4. Stannett, V. *Radiat Phys Chem* 1981, 18, 215.
5. Chapiro, A. *Radiat Phys Chem* 2002, 63, 207.
6. Yang, W. T.; Rånby, B. *J Appl Polym Sci* 1996, 62, 533.
7. Yang, W. T.; Rånby, B. *J Appl Polym Sci* 1996, 62, 545.
8. Yang, W. T.; Rånby, B. *J Appl Polym Sci* 1997, 63, 1723.
9. Rånby, B.; Yang, W. T.; Tretinnikov, O. *Nucl Instrum Meth B* 1999, 151, 301.
10. Rånby, B. *Int J Adhes Adhes* 1999, 19, 337.
11. Wang, H. L.; Brown, H. R. *J Appl Polym Sci*, to appear.
12. Mori, M.; Uyama, Y.; Ikada, Y. *J Polym Sci Part A: Polym Chem* 1994, 32, 1683.
13. Liu, Y. X.; Kang, E. T.; Neoh, K. G.; Tan, K. L. *J Polym Sci Part A: Polym Chem* 2000, 38, 80.
14. Zhang, M. C.; Kang, E. T.; Neoh, K. G.; Han, H. S.; Tan, K. L. *Polymer* 1999, 40, 299.
15. Zhang, J. F.; Kato, K.; Uyama, Y.; Ikada, Y. *J Polym Sci Part A: Polym Chem* 1995, 33, 2629.
16. Lee, W. F.; Lai, C. C. *J Appl Polym Sci* 1995, 55, 1197.
17. Pesetskii, S. S.; Jurkowski, B.; Makarenko, O. A. *J Appl Polym Sci* 2002, 86, 64.
18. Kubota, H.; Ujita, S. *J Appl Polym Sci* 1995, 56, 25.
19. Sun, Y. J.; Hu, G. H.; Lambla, M. *Angew Makromol Chem* 1995, 229, 1, 1.
20. Sun, Y. J.; Hu, G. H.; Lambla, M. *J Appl Polym Sci* 1995, 57, 1043.
21. Cartier, H.; Hu, G. H. *J Polym Sci Part A: Polym Chem* 1998, 36, 1053.
22. Cartier, H.; Hu, G. H. *J Polym Sci Part A: Polym Chem* 1998, 36, 2763.
23. Al-Malaika, S.; Kong, W. *J Appl Polym Sci* 2001, 79, 1401.
24. Torres, N.; Robin, J. J.; Boutevin, B. *J Appl Polym Sci* 2001, 81, 581.
25. Wei, Q.; Chionna, D.; Galoppini, E.; Pracella, M. *Macromol Chem Phys* 2003, 204, 1123.
26. Pracella, M.; Chionna, D. *Macromol Symp* 2003, 198, 161.
27. Pesneau, I.; Champagne, M. F.; Huneault, M. A. *J Appl Polym Sci* 2004, 91, 3180.
28. Choi, S. H.; Nho, Y. C. *Radiat Phys Chem* 2000, 58, 157.
29. Kim, M.; Saito, K. *Radiat Phys Chem* 2000, 57, 167.
30. Zhang, A. M.; Li, C. *Eur Polym J* 2003, 39, 1291.