

Surface Photografting Polymerization of Vinyl Acetate, Maleic Anhydride, and Their Charge-Transfer Complex. IX. Charge-Transfer Complex (5)*

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ABSTRACT: The two-step method was employed to obtain insight into the grafting mechanism of (MAH-VAC)/LDPE film in the preceding investigation, where photoreduction of BP (the first step) was investigated and the subsequent grafting polymerization (the second step) was studied from the viewpoint of kinetics. The present article is devoted to investigation of the living polymerization performance, the compositions of the grafted chains, and the relevant grafting copolymerization mechanism of the (MAH-VAC)/LDPE system. Both grafting copolymerization and nongrafting copolymerization performed living polymerization characteristics to some degree. Regarding the nongrafting copolymerization at a monomer concentration, $[MAH] = [VAC] = 2M$, MAH and VAC mainly underwent alternating copolymerization, especially at low temperatures; however, the grafting copolymerization mostly exhibited random copolymerization, which became obvious with the elevation of temperature. The composition of the monomer

feed largely affected the composition of the grafted copolymer and the nongrafted copolymer. When $[MAH]/[VAC]$ was either 2.5/1.5, 2/2, or 1.5/2.5, MAH/VAC in nongrafted copolymers stayed approximately at 1/1; while when $[MAH]/[VAC]$ was 2.5/1.5, MAH/VAC in the grafted copolymer was nearly 1/1; but in the case of $[MAH]/[VAC]$ being 2/2 and 1.5/2.5, the content of MAH in grafted chains was somewhat lower than that of VAC. All these results demonstrated that the grafting copolymerization on the substrate occurred randomly, which was greatly affected by the affinity of the monomer toward the substrate. This performance of the grafting copolymerization was much different from the nongrafting copolymerization in the solution.

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Key words: charge transfer; graft copolymers; living polymerization; surface

INTRODUCTION

Polyolefins, such as polyethylenes and polypropylenes, are extensively applied as materials owing to their light weights, appropriate flexibility, high solvent resistance, and low cost. However, their inert surface gives rise to a series of tough problems, ranging from poor hydrophilicity, wettabilities, and printability, to poor adhesivity. To overcome these problems, a number of surface modification techniques have been developed, including both chemical and physical processes.¹ The common strategy of nearly all these methods is to introduce polar groups onto the surface of substrates of interest to improve their surface energies. Among these methods, grafting

achieved by plasma discharge,^{2,3} UV irradiation,^{4–7} and so forth drew much attention because of the relatively permanent modification effects. Regarding the monomers used to perform grafting polymerization, maleic anhydride (MAH) was frequently applied due to its low cost and the highly hydrophilic properties of its derivative polymeric products. In literature,^{8–15} to graft MAH polymeric chains onto substrates, free radical initiated grafting polymerization was widely used and conducted in extruders or rheometers. In addition, comonomers or additives were widely used because of the relatively low reactivity of MAH alone, which caused the grafted chains on substrates to generally contain only one MAH unit.^{16–18} These grafted products were attempted to be used as compatibilizing agents, for example, in blends of polyolefins and polyamides.

In our previous studies,^{19,20} MAH was successfully grafted onto low density polyethylene (LDPE) film with UV irradiation, and the grafting polymerization performed high grafting conversion percentage and high grafting efficiency. Furthermore, MAH exhibited a unique self-initiation performance, that is, it could

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undergo photografting polymerization even in the absence of typical photoinitiators.²¹ To accelerate the grafting polymerization, another monomer, vinyl acetate, was employed as the comonomer, and it was found that the second monomer afforded increased photografting polymerization rate and grafting efficiency simultaneously.^{22,23} These studies possess the potential to be put into practice for improving the surface energies of polyolefins. However, the above photografting polymerization of MAH/VAC binary monomer systems was too complex for us to study its grafting copolymerization mechanism, which is certainly important for further design of new grafting polymerization systems. Therefore, in the preceding article,²⁴ we employed a two-step method to study the relevant copolymerization mechanism, and some aspects of it had been addressed; in the present study, the two-step method was still used and other aspects of the grafting copolymerization were investigated further, including the living copolymerization performance of MAH and VAC binary grafting copolymerization systems, the compositions of the grafted copolymer chains, and the involved grafting copolymerization mechanism.

EXPERIMENTAL

Materials

Monomers. Maleic anhydride (MAH), analytically pure, was produced by Tianjin Chemical Reagent Plant No. 6 (Tianjin, China) and purified by recrystallization; vinyl acetate (VAC), analytically pure, was obtained from Tianjin Tiantai Chemical Reagent Plant (Tianjin, China) and purified by distillation in advance.

Photoinitiators. Benzophenone (BP), chemically pure, from Shanghai Reagent Plant No. 1 (Shanghai, China) was purified by recrystallization from ethanol.

Film substrates. Commercial low density polyethylene (LDPE, 63 μm in thickness) films were subjected to Soxhlet extraction with acetone as solvent for 5 h to eliminate the impurities and additives before use.

Other reagents. Benzopinacol, from Aldrich (Germany), and acetic anhydride, from Tianjin Tiantai Chemical Reagent Plant (Tianjin, China), were of analytically pure grade and used without further purification. All other reagents were analytically pure and used directly.

Preparation of LDPE films with semibenzopinacol dormant groups

The equipment for UV irradiation to introduce semibenzopinacol dormant groups onto LDPE film was described previously.^{25,26} The main procedures are briefly described as follows: A given amount of BP

solution was deposited between two film samples with a microsyringe and an appropriate pressure was applied to make the BP solution a thin and even layer. The assembly was covered with a piece of quartz plate, put on the holder, and then irradiated by UV radiation (1 kW high-pressure mercury lamp). After irradiation, the films were taken out, separated, and then subjected to Soxhlet extraction with acetone to remove the residual BP and possibly generated benzopinacol. The detailed description of the above procedures was reported elsewhere.²⁷ The content of semibenzopinacol dormant groups was determined by UV-vis spectroscopy.

Procedures for thermally-induced grafting copolymerization

The LDPE films with semibenzopinacol dormant groups were placed in a self-made glass reactor, to which the monomer solution containing a certain amount of MAH and VAC was added. The above system was deaerated by purging nitrogen for about 10 min, and then placed in a water bath at a temperature for a given period of time under nitrogen protection. After polymerization, the films were taken out and subjected to extraction with acetone for 8 h to exclude the residual monomers, the homopolymers, and the nongrafted copolymers. Then the films were dried and weighed to constant weight to determine the grafting yield.

To follow the polymerization that occurred in the solution paralleling to grafting polymerization on the surface of the LDPE film, a certain amount of the reaction solution was taken out from the reactor at different intervals and added into a large amount of *n*-hexane as the precipitant, which was then heated to the boiling point of *n*-hexane to precipitate out the nongrafted polymer. To eliminate the residual MAH thoroughly, the obtained polymer was dissolved in THF and precipitated with *n*-hexane again. The above processes for dissolving and precipitating the polymer were repeated three times, and then the polymer was dried to constant weight.

To characterize the grafting copolymerization, conversion percentage in solution (CH) and grafting percentage (GP) are defined as follows:

$$\text{CH} = (W_H/W_M) \times 100\% \quad (1)$$

$$\text{GP} = (W_G/W_F) \times 100\% \quad (2)$$

where W_H , W_M , W_G , and W_F are the weight of nongrafted polymer formed in the solution, the added monomer, the grafted polymer on the LDPE film, and the pure LDPE film, respectively. Other parameters, including total conversion percentage (CP), conversion percentage on LDPE film surface (CG), and graft-

ing efficiency (GE), were discussed in the preceding article.

Determination of the intrinsic viscosity and molecular weight of the non-grafted copolymers

A certain amount of nongrafted MAH/VAC copolymer was dissolved in tetrahydrofuran (THF) (0.25g/dL). The intrinsic viscosity of the solution was calculated by measuring the relative viscosity with an Umstatter viscometer ($\phi = 0.3\text{--}0.47$ mm) at 25°C. Relative molecular weights were measured by GPC (Shodex KF-850 column) calibrated by using polystyrene as the standard and THF as the eluent.

Determination of the compositions of the non-grafted copolymers

The compositions of the nongrafted copolymers were determined by titration,²⁸ which is based on the method reported by A. Brown and coworkers.²⁹ The main procedures are as follows: A certain amount of the sample (about 0.035 g) was dissolved in 5 mL of acetone; and after its thorough dissolution, 5 mL of aniline was added into the solution, which was stirred for 30 min at r.t. Then, 10 mL ethanol was added into the solution. The solution was titrated to blue color with 0.075N NaOH aqueous solution, using bromethymol blue as the indicator. More details were described earlier.²⁸

Determination of the compositions of the grafted copolymer chains

The method for determining the compositions of the grafted copolymer chains was established and reported in detail previously.²⁸

RESULTS AND DISCUSSION

Living polymerization performance

The present (MAH-VAC)/LDPE binary monomer grafting copolymerization systems performed living polymerization features to some degree in both grafting polymerization and nongrafting polymerization in the solution. These results are related closely to the employed two-step technique.²⁴ This technique is composed of the following two main steps: first, semibenzopinacol dormant groups were introduced onto the LDPE film surface by photoreduction of BP, and then the dormant groups were reactivated by heating to produce free radicals, which initiated polymerizations of the monomers.³⁰ Here, the semibenzopinacol free radicals could work both as active species to initiate polymerization of the monomers and as termination reagents to the propagating chains. Thereby, either the grafting polymerization or the nongrafting

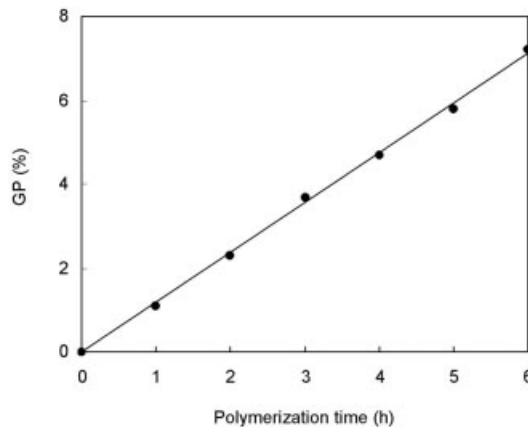
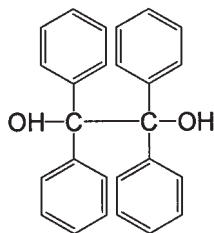


Figure 1 GP as a function of grafting polymerization time. UV irradiation conditions: concentration of BP, 0.3 wt % of LDPE film; irradiation time, 120 s; intensity of UV radiation, 5040 $\mu\text{W}/\text{cm}^2$; protected by nitrogen. Grafting polymerization conditions: [MAH] = [VAC] = 1 mol/L; temperature, 83°C; with acetic anhydride as solvent.

polymerization performed living polymerization characteristics to some extent. At first, the living performance was observed in the grafting polymerization on LDPE film, and the relevant results are presented in Figure 1.

Figure 1 shows that GP increased linearly with the elongation of polymerization time. When grafting polymerization proceeded for 1 h, GP was about 1.1%; when it was 3 h, GP increased to 3.7%; when grafting polymerization time was 6 h, GP increased to 7.2%. These results demonstrated that the grafting propagating chains on the LDPE film were terminated with much difficulty due to the steric effects of the substrate; on the other hand, semibenzopinacol free radicals could play a role as terminators and then as dormant groups, which also made some contribution for the living polymerization performance of the binary monomer systems. These two factors made the living feature of the grafting polymerization more apparent than that of the nongrafting polymerization in solution, which is addressed below.

According to the above discussion, semibenzopinacol free radicals should be responsible for the living performance to some degree. Therefore, the concentration of semibenzopinacol free radicals might be a significant factor. On the other hand, two semibenzopinacol free radicals could couple to form benzopinacol (see Structure 1), which could cleave to form two semibenzopinacol free radicals owing to its high steric repulsion among the four benzene groups. Accordingly, the content of benzopinacol directly affected the living polymerization. To clarify it, benzopinacol was deliberately added into the grafting polymerization system, and the results are shown in Figure 2. In Figure 2, after the photoreduction of BP, if the LDPE films were washed with acetone, both GP and CH



Structure 1 Molecular structure of benzopinacol.

were obviously higher than those without washing with acetone. We know that after UV irradiation, some BP might be left, but it did not affect the subsequent thermally induced polymerization. Thus, the resulted benzopinacol was assumed to exert a large influence on the grafting polymerization and nongrafting polymerization. This view was further confirmed by adding benzopinacol into the grafting system, as shown in Figure 2. When benzopinacol was added into the system, both GP and CH became somewhat lower compared with those without adding benzopinacol, and obviously much lower than that washing with acetone. These results identified the inhibition effects of too much benzopinacol in the grafting polymerization system and, in the meantime, the living polymerization characteristic caused by the proper amount of semibenzopinacol groups. The molecular structure of benzopinacol is presented in Structure 1, and the possible reactions involving it are described in eqs. (3)–(6):

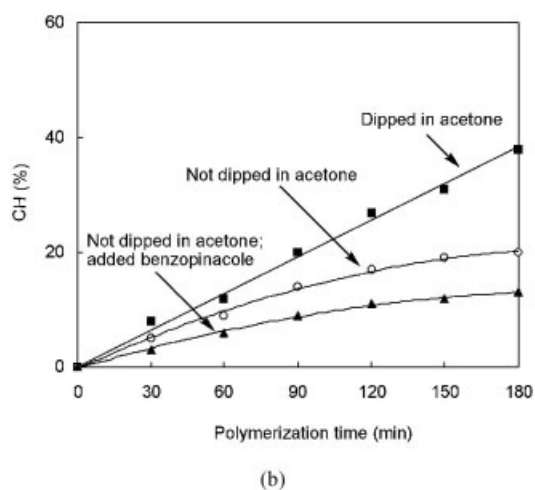
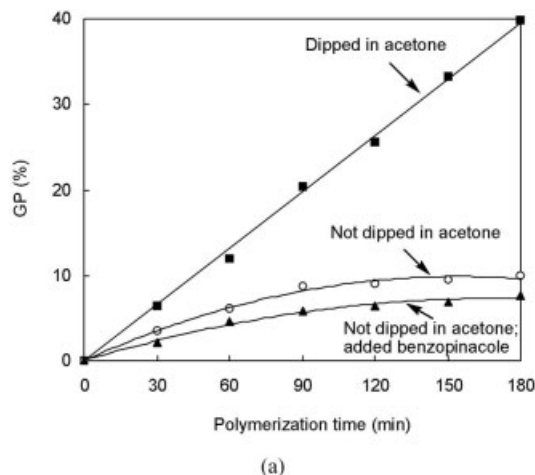
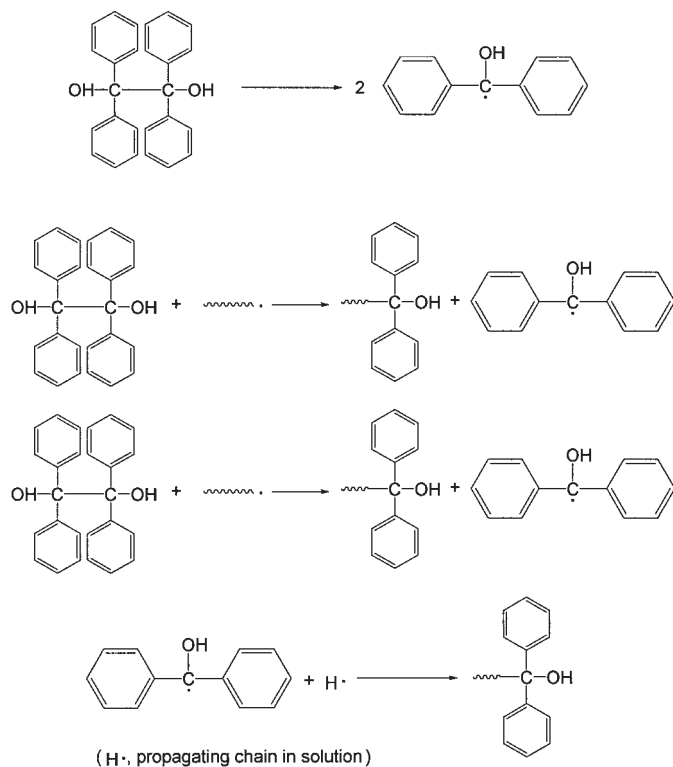


Figure 2 Effects of adding benzopinacol on (a) grafting and (b) nongrafting copolymerization. (Dipped in acetone: after UV irradiation, the films were dipped in a large amount of acetone for 5 min to exclude the formed benzopinacol; Not dipped in acetone: after UV irradiation, the films were not dipped in acetone and were used to conduct grafting polymerization directly; Not dipped in acetone, added benzopinacol: after UV irradiation, the films were not dipped in acetone and were used to conduct grafting polymerization directly, to which benzopinacol (about 1 wt % of BP) was added.). UV irradiation conditions: concentration of BP, 0.3 wt % of LDPE film; irradiation time, 120 s; intensity of UV radiation, 5040 $\mu\text{W}/\text{cm}^2$; protected by nitrogen. Grafting polymerization conditions: $[\text{MAH}] = [\text{VAC}] = 2 \text{ mol/L}$; temperature, 85°C; with acetic anhydride as solvent.

The living polymerization performance was also observed in the intrinsic viscosity of the nongrafted copolymers, which directly reflected the molecular weights of the nongrafted copolymers and indirectly reflected the growing of the grafted copolymer chains. The results are shown in Figure 3, where it can be seen that intrinsic viscosity increased with increasing the monomer concentration; it also increased along with elongation of the polymerization time. To examine this conclusion, molecular weight of the nongrafted

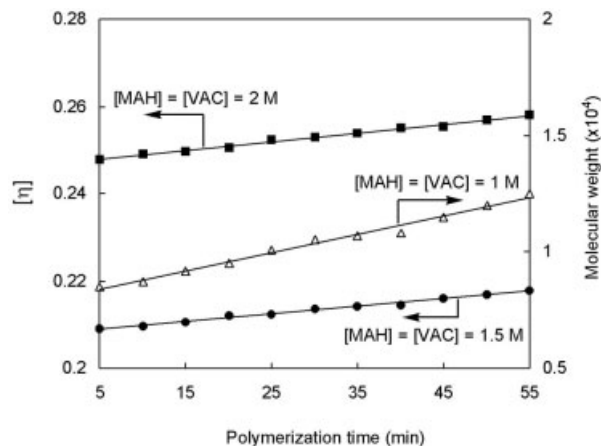


Figure 3 Increase of intrinsic viscosity ($[\eta]$) and molecular weight of the nongrafted copolymers along with elongation of the polymerization time. UV irradiation conditions: concentration of BP, 0.3 wt % of LDPE film; irradiation time, 120 s; intensity of UV radiation, $5040 \mu\text{W}/\text{cm}^2$; protected by nitrogen. Grafting polymerization conditions: temperature, 83°C ; with acetic anhydride as solvent.

copolymers was determined by GPC, and the results are also presented in Figure 3. Even though these data of molecular weight (M_n) are relative values, the plot shows a trend similar to that by intrinsic viscosity, that is, molecular weight increased with increasing the polymerization time. The exact molecular weights of the grafted copolymers and even the nongrafted copolymers could not be experimentally determined. However, it is still reasonable to conclude that the molecular weights of the nongrafted copolymers nearly linearly increased with polymerization time under the experimental conditions; furthermore, it can be inferred that the molecular weights of the grafted copolymer chains also increased with the polymerization time. These observations provided evidence for the living polymerization of the presently studied grafting polymerization systems to some degree.

Compositions of the grafted and non-grafted copolymers

The compositions of the grafted and nongrafted copolymers were determined and are presented in Figure 4. According to Figure 4, the following conclusions could be made: (1) Regarding the nongrafted copolymers, MAH and VAC mainly underwent alternating copolymerization at low temperatures ($75, 83^\circ\text{C}$); at high temperatures ($91, 99^\circ\text{C}$), the copolymerization deviated from alternating copolymerization, and the higher the temperature, the larger the deviation. (2) Regarding the grafted copolymerization, it was quite different from the nongrafted copolymerization; either at low or at high temperature, it mostly performed random copolymerization.

The relationship between the compositions of the copolymers, including grafted copolymer and nongrafted copolymer, and the compositions of the monomer feeds is illustrated in Figure 5. As far as the nongrafted copolymer is concerned, the mole fraction of MAH nearly always remained at 50% with $[\text{MAH}]/[\text{VAC}]$ being either 2.5/1.5, 2/2, or 1.5/2.5 in the monomer feed. In the case of grafted copolymer, it changed with varying $[\text{MAH}]/[\text{VAC}]$; with $[\text{MAH}]/[\text{VAC}]$ being 2.5/1.5, the composition of the grafted copolymers almost stayed at MAH/VAC being 1/1; in the cases of $[\text{MAH}]/[\text{VAC}]$ being 2/2 and 1.5/2.5, the content of MAH in the grafted copolymers was lower than that of VAC to different degrees. These findings demonstrated that at 75°C , the nongrafting copolymerization mainly followed alternating copolymerization, while the corresponding grafting copolymerization mostly underwent random copolymerization. This conclusion is the same as that drawn from Figure 4.

The results could be understood partly in terms of the affinity between the monomers and the substrate, just as discussed in previous articles.^{24,31} The solubility parameter (SP) of LDPE film is 7.9; it is 9.0 and 10.3 for VAC and acetic anhydride (used as the solvent for grafting polymerization), respectively. From the values of SP, the affinity of VAC to LDPE is much higher relative to that of acetic anhydride to LDPE film. Therefore, at the same concentration of MAH and VAC, more VAC attached to or even diffused into the LDPE film; but on the other hand, the copolymerization rate between VAC and MAH was much higher than the homopolymerization rate of VAC. Thus, even though the content of VAC in the grafted copolymers was higher than that of MAH, the difference between them was not fairly obvious. When the content of MAH was higher than that of VAC, the difference in concentration offset the difference in affinity, and therefore the contents of MAH and VAC in the grafted copolymers became similar (Fig. 5a). Another point is worth pointing out. The grafting copolymerization rate was so low, compared with that of nongrafting copolymerization, that it consumed a little amount of monomers and could not affect the copolymerization type of the nongrafting copolymerization.

Mechanism for grafting copolymerization

On the basis of the above discussion, we know that the grafting copolymerization on the substrate was quite different from the nongrafting copolymerization in the solution. More detailed, the grafting copolymerization rate was much slower than that of nongrafting copolymerization; the grafting copolymerization exhibited the characteristics of random copolymerization. In the present study, the two-step method was employed and indeed the grafting copolymerization was a hetero-polymerization system. In this grafting polymerization system, the substrate LDPE film existed in solid

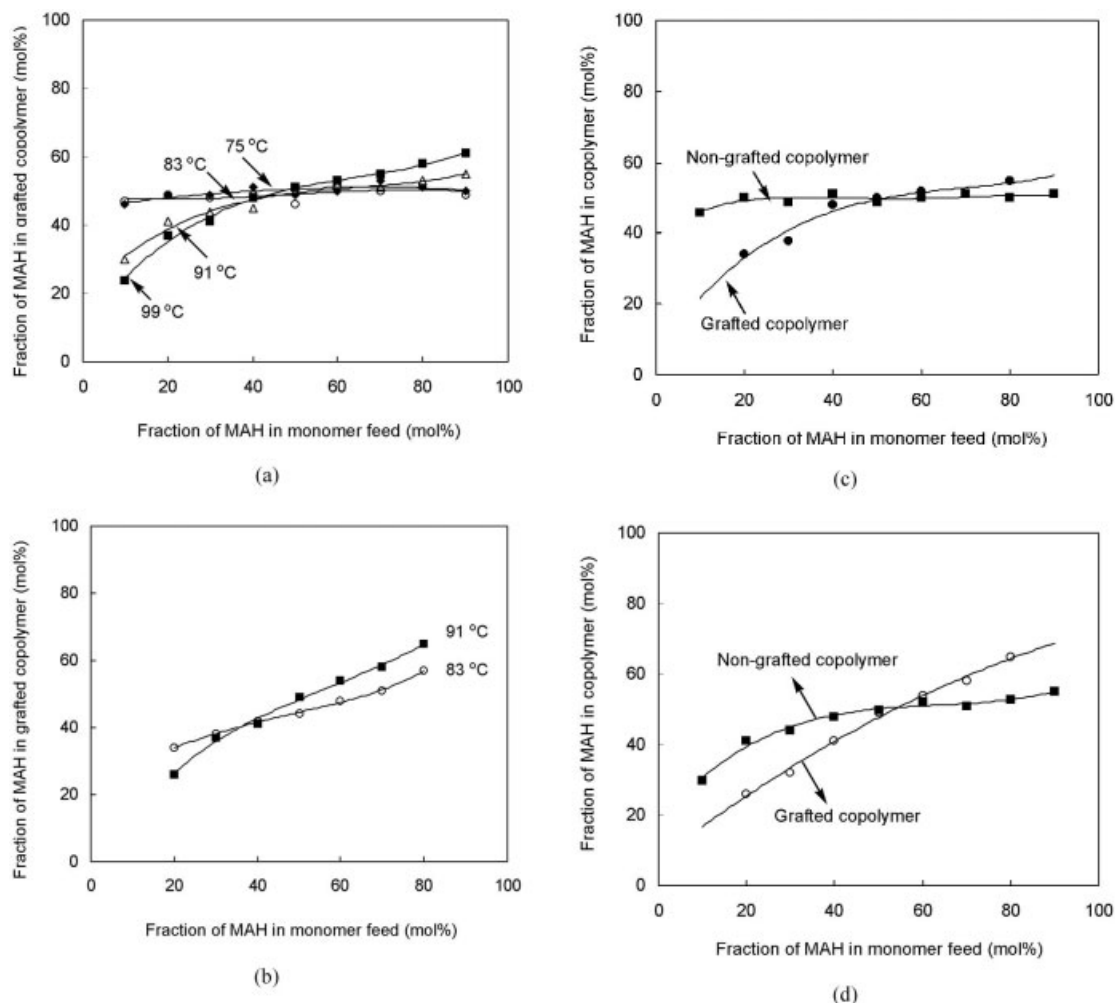


Figure 4 Fraction of MAH in the grafted and nongrafted copolymers versus fraction of MAH in the monomer feed. UV irradiation conditions: concentration of BP, 0.3 wt % of LDPE film; irradiation time, 120 s; intensity of UV radiation, 5040 $\mu\text{W}/\text{cm}^2$; protected by nitrogen. Grafting polymerization conditions: $[\text{MAH}] = [\text{VAC}] = 2 \text{ mol/L}$; with acetic anhydride as solvent; (a) composition of the copolymer formed in solution; (b) composition of the grafted copolymer; (c) composition of the copolymer; (d) composition of the copolymer.

form. Therefore, the grafting copolymerization was influenced not only by the interaction between the two monomers and the interaction between the propagating species and the monomers, but also by the affinity of the monomers toward LDPE film and the diffusion of the monomers into the substrate. In fact, the latter may be more important. To explain the processes, a model was put forward, as described in Figure 6.

In the model, the LDPE macromolecule free radicals existed on the surface or subsurface of the LDPE film; only those monomer molecules absorbed by the substrate or those distributed into the subsurface had the chance to be initiated to polymerize, so the affinity of the monomers to the substrate played a crucial role in the grafting copolymerization. The distribution of the monomers into the substrate proceeded slowly, which led to the low grafting copolymerization rate. Additionally, the concentration of the monomers affected the grafting co-

polymerization drastically. The used monomers, MAH/VAC, could form a CT complex, which diffused into the substrate with more difficulty, relative to VAC, and therefore, they could not exert so great an influence on the grafting copolymerization as they did on the non-grafting copolymerization. This also resulted in a slower grafting copolymerization compared with the non-grafting copolymerization. We think that this model is also applicable to some degree to the photografting copolymerization of the (MAH-VAC)/LDPE system carried out by the one-step method,^{22,23} and provides important information for us to design other new photografting systems in the future.

CONCLUSIONS

The presently studied grafting polymerization systems performed the living polymerization character-

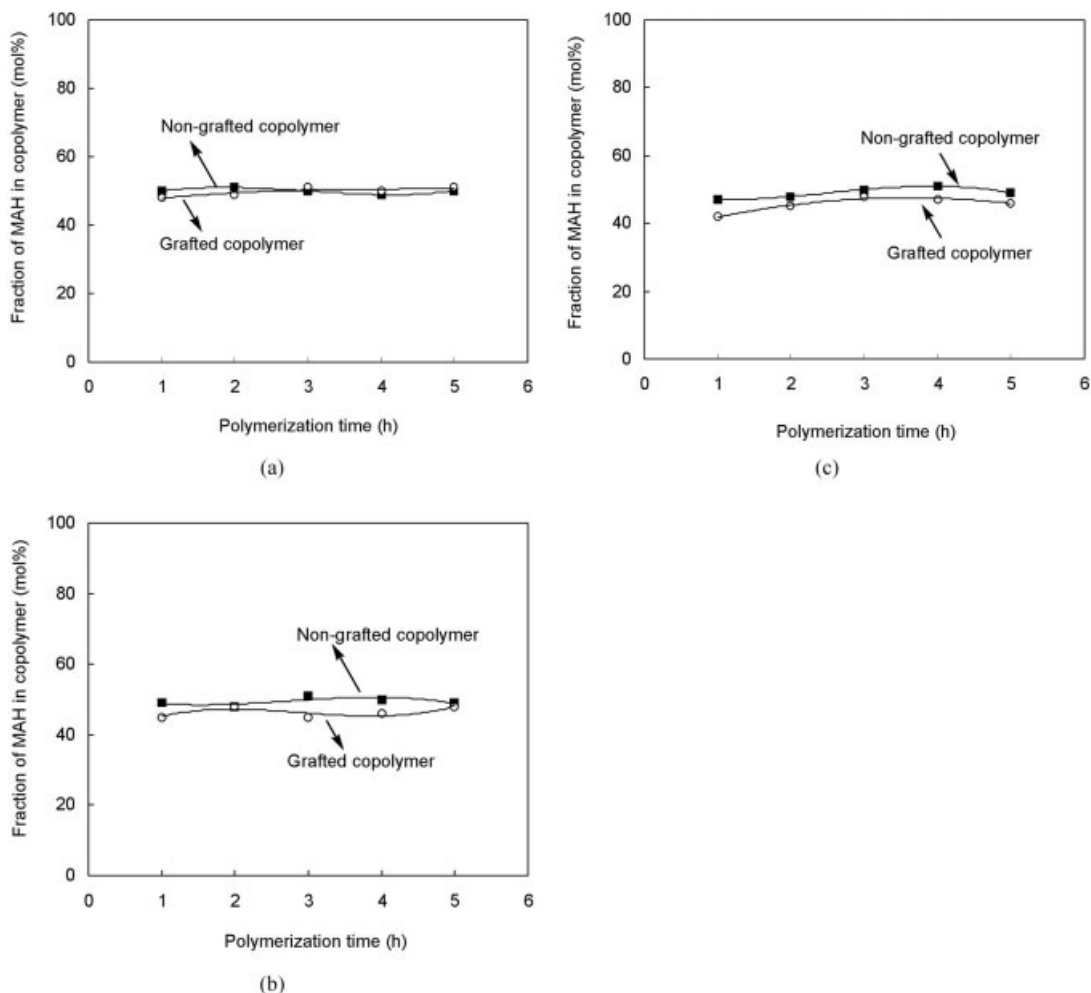


Figure 5 Fraction of MAH in the grafted and nongrafted copolymer versus fraction of MAH in the monomer feed. UV irradiation conditions: concentration of BP, 0.3 wt % of LDPE film; irradiation time, 120 s; intensity of UV radiation, $5040 \mu\text{W}/\text{cm}^2$; protected by nitrogen. Grafting polymerization conditions: temperature, 75°C ; with acetic anhydride as solvent; (a) $[\text{MAH}]/[\text{VAC}] = 2.5/1.5$ (mol/L); (b) $[\text{MAH}]/[\text{VAC}] = 1.5/2.5$ (mol/L); (c) $[\text{MAH}]/[\text{VAC}] = 2/2$ (mol/L).

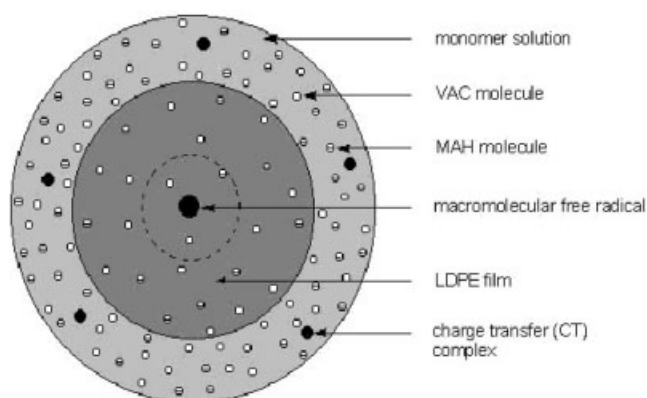


Figure 6 Model for grafting copolymerization of MAH/VAC binary monomers on LDPE film.

istics to some degree, which could be observed in the linearly increasing grafting percentage of the monomers on the substrate and the increase in intrinsic viscosity of the nongrafted copolymers along with grafting polymerization time; the influence of benzopinacol on the grafting copolymerization and nongrafting copolymerization also provided evidence for it. The grafting copolymerization showed random copolymerization performance, and with an increase of the temperature, the grafting copolymerization was inclined to undergo random copolymerization. At low temperature, the nongrafting copolymerization showed alternating copolymerization, but at high temperature, it deviated from alternating copolymerization. In systems with $[\text{MAH}]/[\text{VAC}]$ being 1.5/2.5, 2/2, and 2.5/1.5, nongrafting copolymerization also showed alternating copolymerization; while in the grafted copolymers, the ratio of MAH/VAC remained at nearly 1:1 when $[\text{MAH}]/[\text{VAC}]$ was 2.5/1.5, but at

[MAH]/[VAC] of 2/2 and 1.5/2.5, the content of MAH was lower than that of VAC. These results indicated that the affinity of the monomers toward LDPE film affected the grafting copolymerization, and the diffusion of monomers into the substrate might be a decisive factor influencing the grafting copolymerization rate and the composition of the grafted copolymer chains.

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