

# Electron-Induced Crosslinking of Polypropylene with the Addition of Hydrogen-Donating Hydrocarbons

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**ABSTRACT:** Electron-induced crosslinking of polypropylene with the addition of hydrogen-donating hydrocarbons produced from petroleum (HHAP) together with A-TMPT (trimethylpropanetriacrylate) was studied, and the following results were obtained: (1) Effective crosslinking was achieved at lower doses under 10 kGy. (2) HHAP functions as an effective radical scavenger to suppress scissions. (3) Distinguished improvements in properties such as elongation and heat distortion temperature were achieved by the irradiation. (4) Distinct diminution in melt flow rate and evident increase in average molecular weight with the irradiation at lower doses were observed. (5) It was confirmed that the oxidative degradation during mixing and molding was noticeably inhibited by the addition of HHAP. This effect as an antioxidant during these procedures along with the irradiation seems to be a cause to promote crosslinks. These effects are attributable to the high radical scavenging effects of HHAP and its pure hydrocarbon constitution without functional groups containing heteroatoms. © 1997 John Wiley & Sons, Inc. *J Appl Polym Sci* **64**: 311–319, 1997.

**Key words:** crosslinking of polypropylene; hydrogen-donating hydrocarbon; radical scavenger; pure hydrocarbon; physical properties

## INTRODUCTION

One of the most important problems in crosslinking of polypropylene (PP) seems to be that PP is liable to be fractured (scissioned); and, accordingly, to suppress these scissions seems to cause effective crosslinking.<sup>1–3</sup> The presence of radical scavengers to suppress these scissions together with polyfunctional monomers to accelerate crosslinkings is said to be preferable to achieve effective crosslinking.<sup>2,3</sup>

In these cases, however, the radical scavengers

suppress crosslinkings themselves, along with scissions; hence, the balance of these two suppressions seems to be decisive.

Various polyfunctional monomers such as silanes and vinylsilanes,<sup>4</sup> allylacrylates,<sup>4</sup> divinylsulfones,<sup>5</sup> and others<sup>4,6,7</sup> have been proposed and investigated. However, the studies on the radical scavengers to suppress scissions to achieve more effective crosslinking of PP have not satisfactorily been reported to date, notwithstanding that they play important roles in this technique. Although the conventional antioxidants have effectively been used,<sup>2,8</sup> the radical scavengers for this purpose are deemed desirable to be investigated from the viewpoints of radical scavenging effects, resistance to radiation, and morphology.

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Hydrogen-donating hydrocarbons have seldom been used as radical scavengers, except in processes such as coal liquifaction and heavy oil upgrading,<sup>9-11</sup> where they are used as hydrogen donor solvents in reductive atmospheres. However, it was certified that hydrogen-donating hydroaromatics (abbreviated here as hydroaromatics) can scavenge free radicals even in the presence of oxygen (that is, they function as antioxidants), although the partial pressures of oxygen at the reaction sites are closely related.<sup>12</sup>

It was also found that materials called heavy hydroaromatics (HHAPs), which are produced from petroleum fractions, have radical scavenging abilities as high as the conventional antioxidants, notwithstanding that they are pure hydrocarbons without functional groups containing heteroatoms.<sup>13,14</sup> The high radical scavenging effects of HHAPs were proven in the reactions with DPPH(2,2-diphenyl-1-picrylhydrazyl);<sup>15</sup> and the prominent inhibiting effects were also confirmed by the practical tests for the thermal, oxidative, and radiation-induced oxidative degradation of petroleum products,<sup>12,16</sup> rubbers,<sup>17</sup> plastics,<sup>18</sup> and thermoplastic elastomers.<sup>19</sup> The superior inhibiting effects on radiation-induced oxidation, including postoxidation of PP, were also verified.<sup>20,21</sup> In addition, the stability of HHAP toward radiation, which can be attributed to the pure hydrocarbon constitution without functional groups containing heteroatoms, was confirmed.<sup>22</sup>

Based on these facts, the additive effects of HHAP on electron-induced crosslinking of PP were investigated.

## EXPERIMENTAL

### Materials

The polypropylene (PP; density, 0.90–0.91; weight-average molecular weight by GPC, 324,000) used was in powder form, produced by Nippon Petrochemical Co., Ltd. The crosslinking agent was trimethylolpropanetriacrylate (296.3  $M_w$ , abbreviated as A-TMPT).

HHAP produced from petroleum was used as the stabilizer (radical scavenger). HHAPs are multicomponent hydrocarbons, derived from petroleum, which were produced by the hydrogenation of the highly aromatic heavy fraction from petroleum. Many components (more than 300 detected by gas chromatography) are contained in HHAP, and it is difficult to define the individual

**Table I Properties of HHAP Used in Crosslinking of Polypropylene**

Density at 15°C (g/cm <sup>3</sup> )	1.0562
Viscosity at 100°C (cST)	84.00
Softening point (°C)	24.0
Composition (wt %)	
Saturates	12.1
Aromatics	74.4
Resins	8.1
Asphaltenes	5.4
Elemental analysis (wt %)	
C	90.5
H	9.4
N	0.1
S	0.05
Average molecular weight	360
<sup>1</sup> H-NMR	
H <sub>a</sub>	13.4
H <sub>α</sub>	24.9
H <sub>β</sub>	42.1
H <sub>γ</sub>	19.6

components. Therefore, the properties as a mixture are shown in Table I. Most components contained in HHAP can be thought to have partly hydrogenated products of the condensed aromatic rings, and HHAP exhibits the high radical scavenging ability based on the hydrogen-donating property, as already confirmed.

The crosslinking agent and the stabilizer were mixed into PP powder at 170°C using Labo Plastomill and molded to sheet (0.5 mm thickness). The sheets were cut into dumbbells (ASTM-1822L) for the irradiation.

### Irradiation

The samples were irradiated with electron beam (EB) generated from 2 MeV accelerator (Japan Atomic Energy Institute, Takasaki Radiation Chemistry Research Establishment) at room temperature under vacuum ( $10^{-2}$  torr). Dose rates were 0.1 and 0.5 kGy/s, and were irradiated up to 300 kGy.

### Measurement

Tensile tests were conducted using an Instron Model 4302 at room temperature, and the crosshead speed was controlled at 100 mm/min. The elongation at the break, yield strength, tensile strength, and modulus at 10% elongation were

**Table II** Conditions of GPC

Column	GMH.HT (2)
Solvent	Dichlorobenzene
Temperature	135°C
Speed	1 mL/min
Concentration	0.25 wt/wt %
Injection	300 mL
Detector	RI
Calibration	Polypropylene

measured by these tests. The gel fraction of the irradiated samples were determined by the extraction in xylenes at 138°C for 24 h.

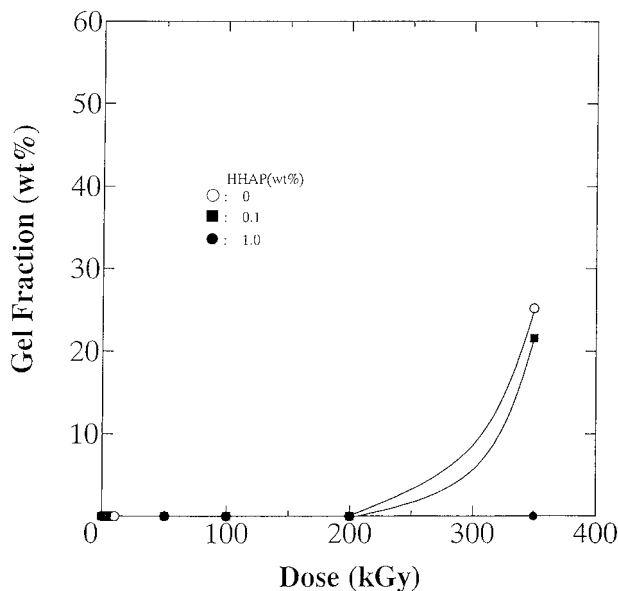
Mechanical properties such as elongation (JIS K7113), tensile strength (JIS K7113), Charpy impact value (JIS K7111), heat distortion temperature (JIS K7207), deflection temperature (JIS K7207), Vicat softening temperature (JIS K7206), melt flow rate (ASTM D1238), and Izod impact value (JIS K7110) were measured to infer the structural changes by the irradiation. The changes in molecular weight were also analyzed by gel permeation chromatography (GPC). The conditions for GPC are shown in Table II.

## RESULTS AND DISCUSSION

From our previous studies, A-TMPT exhibited most prominent effects as a polyfunctional monomer for crosslinking of PP, and the data with A-TMPT will be described hereafter.

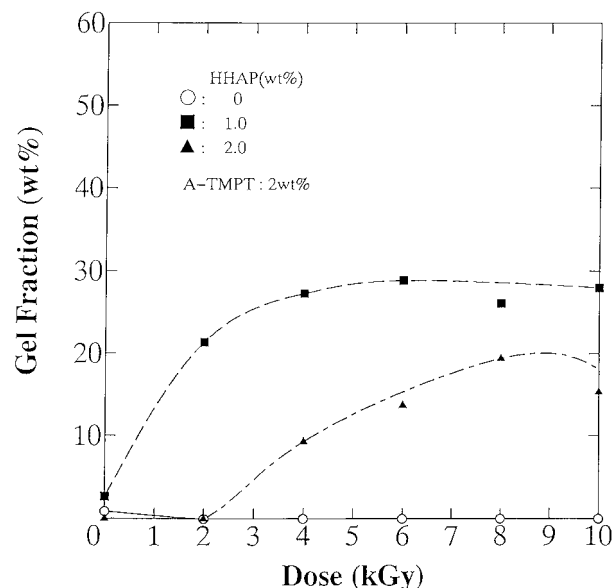
The changes in gel fraction by dose without A-TMPT are shown in Figure 1. From this figure, it can be observed that the formation of gel is suppressed until 200 kGy in all cases. However, above 200 kGy in dose, gel is formed in the cases in which radical scavengers are not added or the addition rate is low (0.1%). In contrast, gel is not formed at higher doses than 200 kGy when plenty of radical scavengers (1.0%) are added. From these data, it can be understood that the formation of gel is restricted by the addition of HHAP (this was also confirmed with a hindered phenolic antioxidant), and gel is not formed when the polyfunctional monomers are not added.

Next, the gel fraction by dose, when the polyfunctional monomer (A-TMPT) is added (2.0 wt %), is illustrated in Figure 2. The changes in gel fraction by dose at lower doses (10 kGy) are illustrated in this figure. It can be observed in this figure that gel is not formed up to 10 kGy when

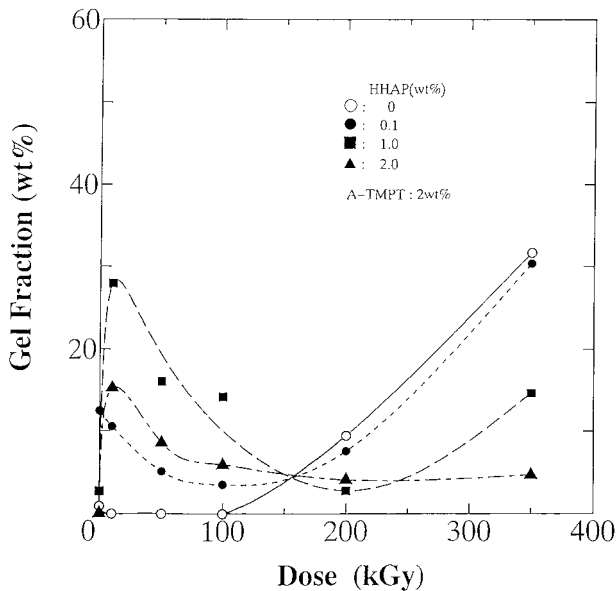


**Figure 1** Effects of the addition of HHAP on gel fraction without A-TMPT by EB irradiation at room temperature under vacuum ( $10^{-2}$  torr) at the rate of 0.5 kGy/s.

HHAP is not added; that when 1 wt % HHAP is added, gel is formed at lower doses, and 30 wt % gel fraction is attained at 4 kGy; (3) and that when 2 wt % HHAP is added, gel fraction was lower than the addition of 1 wt % HHAP. The



**Figure 2** Effects of the addition of HHAP on gel fraction with 2.0 wt % A-TMPT at lower doses by EB irradiation at room temperature under vacuum ( $10^{-2}$  torr) at the rate of 0.1 kGy/s.



**Figure 3** Effects of the addition of HHAP on gel fraction with 2.0 wt % A-TMPT at higher doses. Irradiation is the same in Figure 1.

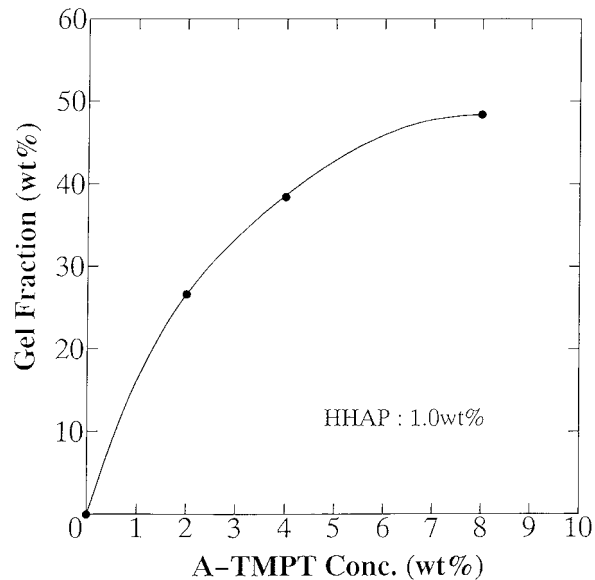
gel fractions in these data are not high enough compared with other reports<sup>2</sup>; however, they can be raised by selecting appropriate conditions, if desired.

From these observations, the following conclusions can be derived: a radical scavenger is definitely necessary to crosslink PP at lower doses, and an excessive radical scavenger is not effective to get higher crosslinking. That is, the existence of a radical scavenger is indispensable to suppress scissions during irradiation; however, the excessive addition of it causes the suppression of crosslinking itself.

Figure 3 shows an interesting tendency of the addition of HHAP at higher doses (above 200 kGy). It is obvious that crosslinking is suppressed by the addition of HHAP at the higher doses. When the addition of HHAP (1.0 wt %) and dose (6 kGy) are fixed, gel fraction is increased by raising A-TMPT concentration (Fig. 4), and 50% of gel fraction is attained at 8 wt % of A-TMPT.

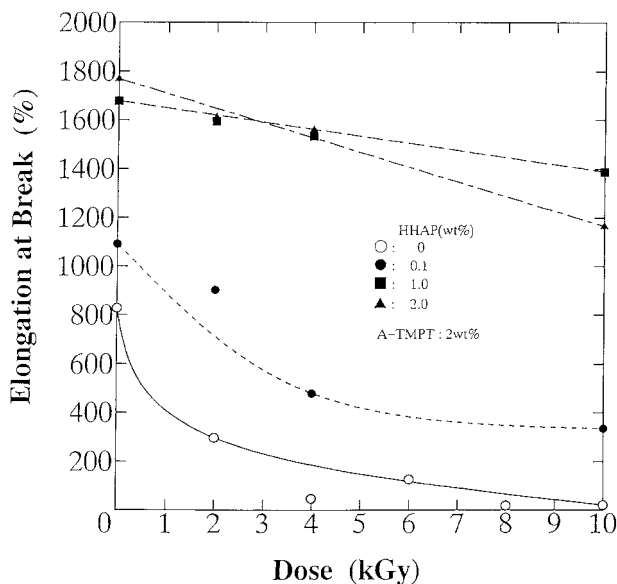
The changes in mechanical properties will be reviewed. The changes in elongation at the break by dose are illustrated in Figure 5. At dose zero, elongation is changed by the addition of HHAP. This means that oxidative degradation during mixing and molding is suppressed by HHAP.

This fact seems to be important in crosslinking of PP. That is, PP without antioxidant is notably scissioned during mixing and molding, as shown

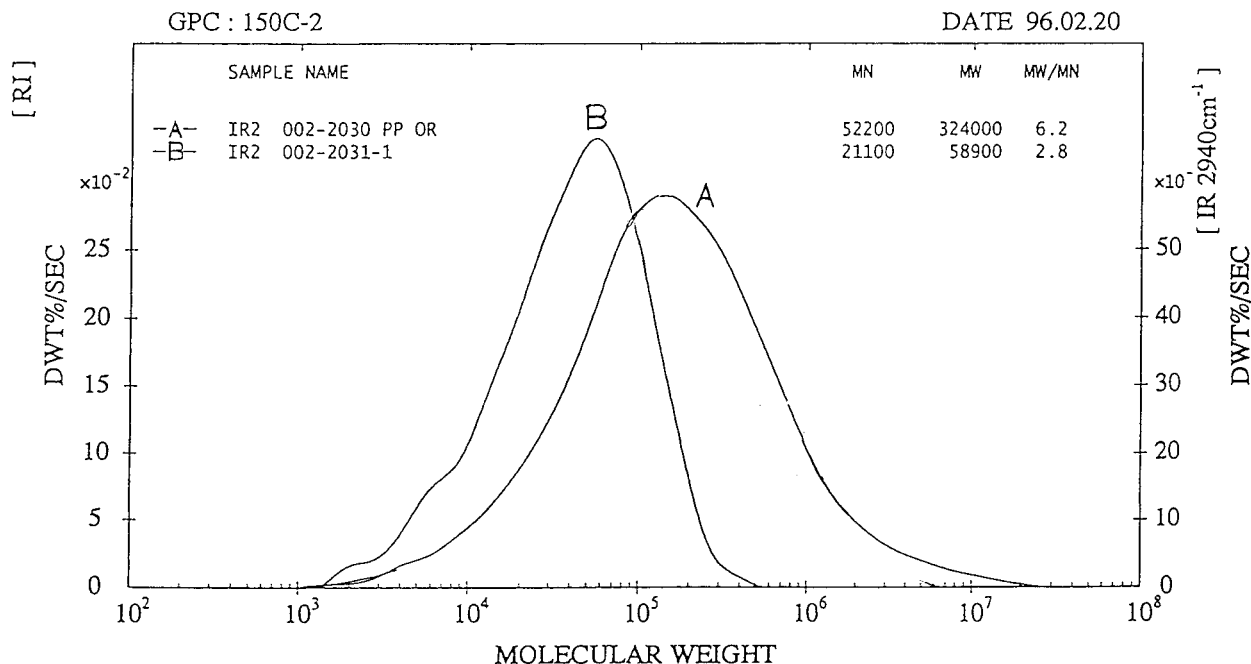


**Figure 4** Changes in gel fraction by the amount of A-TMPT at the fixed amount of HHAP (1.0 wt %) and the dose (6.0 kGy). Irradiation is the same as in Figure 2.

in Figure 6. This can also be certified in the changes in melt flow rate (MFR) described later. From these facts, one of the important roles of antioxidants in crosslinking of PP is deemed to be the suppression of scissions (oxidative degradation) during mixing and molding together with the ones during irradiation. At the same time, the



**Figure 5** Effects of the addition of HHAP on elongation at the break at the fixed amount of A-TMPT (2.0 wt %). Irradiation is the same as in Figure 2.



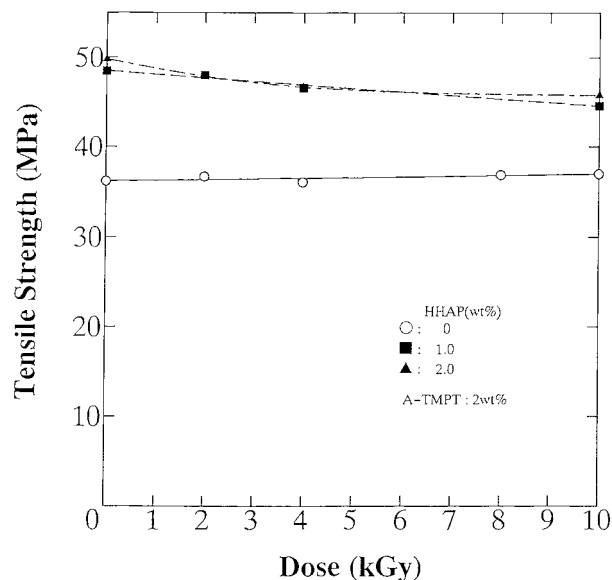
**Figure 6** Changes in molecular weight distribution by mixing (170°C, 6 min) and molding (by GPC): (A) original PP (powder); (B) original PP sheet (without additives and without irradiation).

effects on crosslinkings of the oxygen atoms that are taken in during these procedures cannot be disregarded.

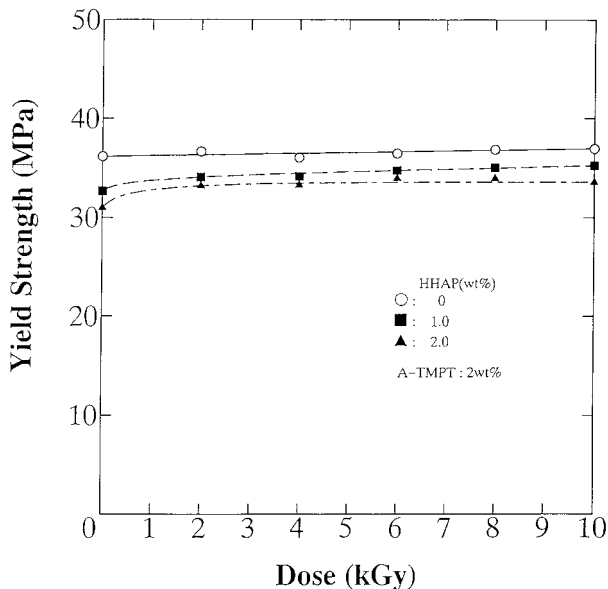
The changes in tensile strength by dose are illustrated in Figure 7. It can also be observed that tensile strength is remarkably improved by the addition of HHAP, the same as elongation. These are consistent with the gel fraction shown in Figure 3. Yield strength and modulus are shown in Figures 8 and 9, respectively. These values are decreased by the addition of HHAP. These data also seem to show the crosslinkings; however, the additive effects of HHAP on physical properties should be taken into account, at the same time. HHAP is a mixture of hydrocarbons whose properties are similar to oil, as shown in Table I; and the addition of it bestows plasticization on the base polymers, as exhibited in the increase in bending of rubbers.<sup>17</sup>

From these data, it is quite obvious that PP is crosslinked by the electron beam irradiation at lower doses in the presence of A-TMPT and HHAP. Furthermore, the changes in average molecular weight were investigated by GPC, and the results are shown in Figure 10. It can be observed that average molecular weight ( $M_w$ ) is increased by the irradiation in the presence of A-TMPT (2.0 wt %) and HHAP (1.0 wt %), extending over the original powder.

Then, the improvements in properties for practical uses were investigated, and the results are summarized in Table III. It can be seen from this table that the changes in such properties as elongation at the break, heat distortion temperature (HDT), and MFR are evident. Especially, MFR is



**Figure 7** Effects of the addition of HHAP on tensile strength at the fixed amount of A-TMPT (2.0 wt %). Irradiation is the same as in Figure 2.



**Figure 8** Effects of the addition of HHAP on yield strength at the fixed amount of A-TMPT (2.0 wt %). Irradiation is the same as in Figure 2.

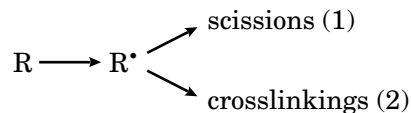
notably changed by the irradiation. That is, it can be seen from this table that MFR is remarkably diminished by the irradiation, and it seems that crosslinkings proceed, as illustrated in Figure 10. However, it should be noted that the MFR at dose zero is greatly reduced by the addition of HHAP and A-TMPT. This can be deemed that the oxidative degradation during mixing and molding is inhibited by HHAP, as described before.

From the results described so far, it can be concluded as follows:

1. The effective crosslinking can be achieved by electron beam irradiation at lower doses in the presence of A-TMPT and HHAP.
2. HHAP functions as a radical scavenger to suppress scissions together with crosslinkings, and it is definitely necessary to achieve effective crosslinking of PP at lower doses.
3. The increase in average molecular weight by the irradiation was also confirmed by GPC, and this was exhibited as diminution in MFR.
4. There are distinct improvements in properties such as elongation and HDT.
5. The oxidative degradation during mixing and molding was obviously inhibited by HHAP.

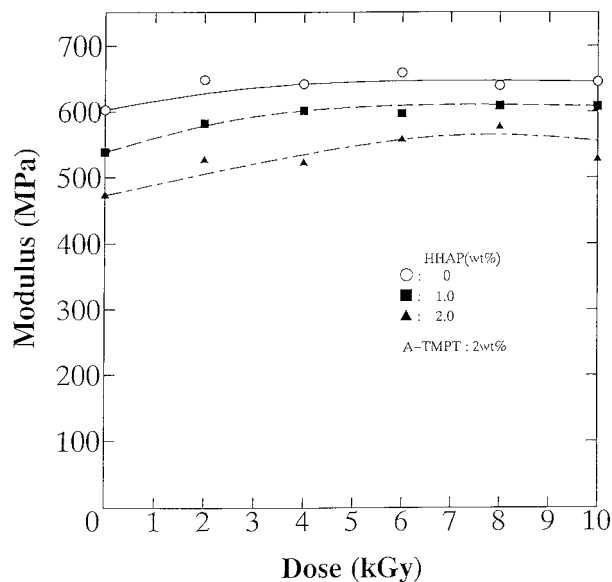
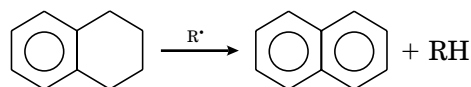
The effects of HHAP are obvious in these data,

described above. As judged from Figures 1–3, routes (1) (2) in the following are suppressed by HHAP:

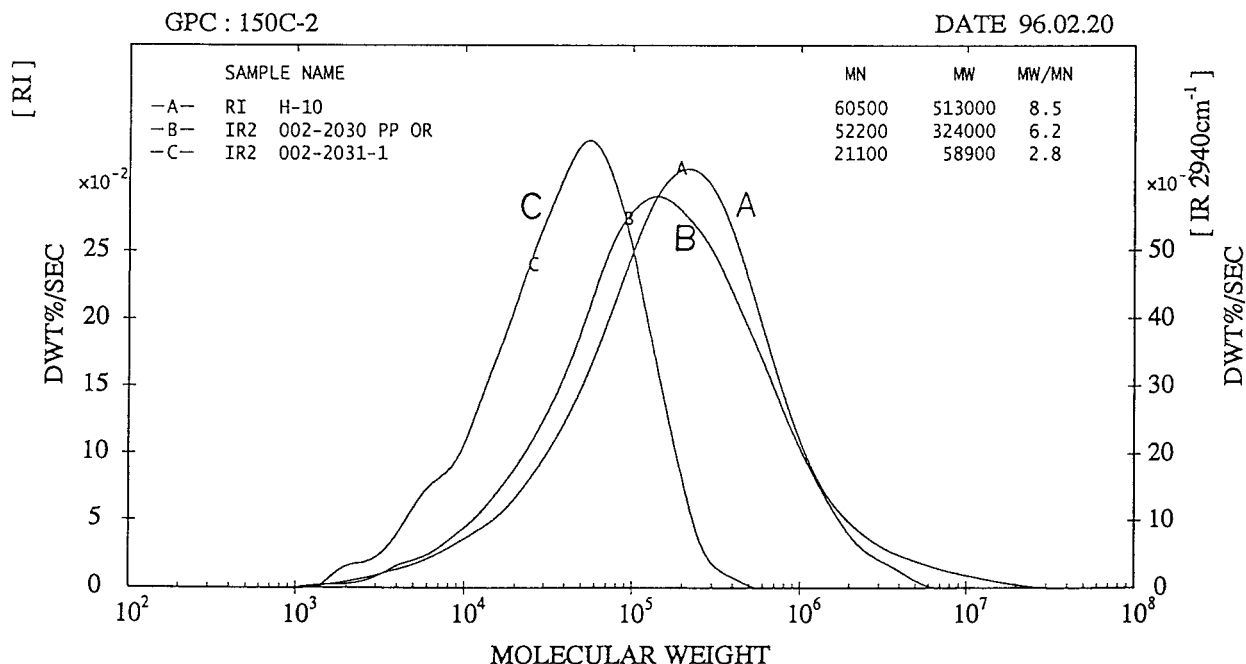


Scissions seem to be predominant,<sup>23</sup> and crosslinkings do not occur at lower doses when HHAP is not present. However, scissions are suppressed by HHAP, and crosslinkings occur when HHAP is present as shown in Figure 2. From these outcomes, the radical scavenging ability of HHAP is deemed to play important roles in this system.

HHAPs are multicomponent hydrocarbons. Most of the components have partially hydrogenated products of condensed aromatic rings, and they are deduced to exhibit radical scavenging effects based on their hydrogen-donating properties, as elucidated below<sup>12</sup> (tetralin is taken as the example for the explanation):



**Figure 9** Effects of the addition of HHAP on modulus at the fixed amount of A-TMPT (2.0 wt %). Irradiation is the same as in Figure 2.



**Figure 10** Changes in molecular weight distribution of crosslinked PP by GPC: (A) crosslinked sheet (A-TMPT 2.0 wt %, HHAP 1.0 wt %, irradiation 10 kGy); (B) original PP (powder); (C) original PP sheet (without additives and without irradiation).

The radical scavenging effects of HHAPs were already confirmed in the reactions with 2,2-diphenyl-1-picrylhydrazyl (DPPH), as already reported.<sup>12</sup> The reaction rate constants of HHAPs with DPPH at 50°C in air assuming first-order reactions are demonstrated in Table IV, compared with typical hydrogen-donating hydrocarbons,

conventional hindered phenolic antioxidants, and ultraviolet ray absorbents.<sup>13,14</sup> From this table, the following can be observed.

1. HHAPs have much higher radical scavenging abilities than tetralin and 1,2,3,4,5,6,7,8-octahydrophenanthrene (OHP).

**Table III** Properties of the Crosslinked PP at Various Conditions

Sample No.	A-6	B-0	B-2	B-4	B-6	B-8
PP	100	100	100	100	100	100
HHAP	0	1.0	1.0	1.0	1.0	1.0
A-TMPT	0	2.0	2.0	2.0	2.0	2.0
Dose (kGy)	0	0	2	4	6	8
Yield strength (kg/mm <sup>2</sup> )	—	3.15	3.13	—	3.15	3.07
Tensile strength (kg/mm <sup>2</sup> )	2.32	2.20	—	—	—	—
Elongation at the yielding point (%)	—	14.0	13.5	—	14.0	14.0
Elongation at the break (%)	5.5	86	>400	—	>400	>400
Bending strength (kg/mm <sup>2</sup> )	3.90	4.29	4.11	—	4.19	4.31
Bending modulus (kg/mm <sup>2</sup> )	144	145	141	—	153	157
Charpy impact value (kg cm cm <sup>-2</sup> )	21.1	25.7	40.6	—	39.6	48.0
HDT (°C)	100	105	114	—	115	118.3
Vicat softening temp. (°C)	150.7	152.5	152.0	—	153.5	152.5
MFR (g/10 min) at 230°C, 2160 g	135	0.59	0	—	0	0
MFR (g/10 min) at 230°C, 5000 g	—	—	0.06	—	0.173	0.512
Izod impact value (kJ/m <sup>2</sup> )	—	—	—	2.69	—	—
Deflection temperature (°C)	—	—	—	70.5	—	—

**Table IV Reaction Rate Constants of Antioxidants and Hydroaromatics with DPPH at 50°C in Air (Assuming Pseudo First Order)**

Antioxidant	$k$ ( $10^{-3} \text{ min}^{-1}$ )	Activation Energy (kcal/mol)
Tetralin	1.11	21.6
OHP (octahydrophenanthrene)	5.60	9.5
HHAP (A)	78.5	5.3
HHAP (B)	26.6	11.2
HHAP (C)	130	—
Irganox 1076	130	(24.2)
DBPC	215	5.2
Tinuvin (UV absorber)	0	—
Seesorb 101 (UV absorber)	0	—

2. Some of the HHAPs have radical scavenging abilities as high as the conventional hindered phenolic antioxidants.
3. The ultraviolet rays absorbents do not have radical scavenging abilities as expected.

It has not been known that pure hydrocarbons without functional groups containing heteroatoms can have such high radical scavenging abilities as the conventional hindered phenolic antioxidants, and, in addition, they can be obtained from petroleum. The reasons why such hydrocarbons can have such high radical scavenging abilities are not satisfactorily elucidated yet; however, some data to suggest this were already reported in our previous articles.<sup>13,14</sup>

The high radical scavenging abilities of HHAPs have already been certified in many practical tests as thermal and oxidative degradation of petroleum products; thermal-oxidative deterioration of rubbers, plastics, and thermoplastic elastomers; radiation-oxidation of petroleum products and polyolefins; and others. They have also been proven in inhibition of coke formation in heavy oil processing (nonoxidative atmospheres).<sup>24</sup> These outcomes seem to substantiate the high radical scavenging effects of HHAPs, which were exhibited in the reactions with DPPH. In the tests for crosslinking of PP reported here, however, HHAP(B) in Table IV was used to avail the appropriate radical scavenging effects.

The prominent inhibiting effects of HHAPs toward thermal-oxidative and radiation-oxidative (including postoxidation) degradation of PP were already confirmed.<sup>20,21</sup> Accordingly, the effects reported in this paper are quite acceptable.

HHAPs are pure hydrocarbons without func-

tional groups containing heteroatoms, and some characteristic properties different from the conventional antioxidants are derived from this fact. The following properties were found in our tests:

1. resistance to heat (this is already known in coal liquefaction and other processes)<sup>12,16</sup>;
2. resistance to radiation<sup>22</sup>;
3. low additive effects on electrical and dielectric properties of the base materials.<sup>13,14</sup>

In crosslinking of PP, resistance to radiation is closely related. As reported in our previous paper,<sup>22</sup> functional groups (OH group in our tests) are liable to be attacked by irradiation, and they are changed to carbonyl groups. Contrary to this, HHAPs are resistant to radiation because they don't have the functional groups to be attacked. It is not obvious that to what extent this property is contributed in these data reported here; however, in any case, this property should be beneficial for radiation processing, especially because conventional antioxidants are not resistant to radiation as many researchers have pointed out.<sup>25,26</sup>

In addition, HHAPs are liquid state in normal uses. The morphological studies have not been conducted, but some differences from the conventional antioxidants cannot be denied from this viewpoint. The distinguished improvements in the properties as shown in Table III could be partly caused by this factor. Furthermore, HHAPs are multicomponent, and the radical scavenging abilities and the properties of them can be controlled. In crosslinking of PP, the flexibilities of these properties seem to be beneficial, and this type of stabilizers seem to exhibit their own characteristics in this respect.

Conclusively, it was revealed that the effective



crosslinking can be achieved by the addition of HHAP, which is a pure hydrocarbon radical scavenger derived from petroleum, together with A-TMPT.

## CONCLUSIONS

Crosslinking of PP with the addition of HHAP, which is a pure hydrocarbon radical scavenger together with A-TMPT, was studied. The following conclusions were obtained.

1. Effective crosslinking was achieved at lower doses (under 10 kGy).
2. HHAP functions as a radical scavenger to suppress scissions.
3. Distinguished improvements in properties such as elongation and HDT were achieved.
4. Distinct diminution in MFR and an evident increase in average molecular weight were confirmed.
5. It was observed that the oxidative degradation during mixing and molding was noticeably inhibited by HHAP, and the effect as an antioxidant during these procedures seems to be a cause to promote crosslinking.

These effects are attributable to the high radical scavenging ability of HHAP and, probably, to its pure hydrocarbon constitution.

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