

## **Effect of Atmosphere on Radiation-Induced Crosslinking of Polyethylene. Part III. Effect of Various Gases and Effect of Electric Field**

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### **INTRODUCTION**

The effect of atmosphere on the crosslinking of polyethylene was reported previously<sup>1,2</sup> for the following eight gases: NO<sub>2</sub>, O<sub>2</sub>, Cl<sub>2</sub>, SO<sub>2</sub>, NH<sub>3</sub>, CO, H<sub>2</sub>, and N<sub>2</sub>O. Polyethylene irradiated in N<sub>2</sub>O shows a decided increase in gel content over one irradiated in vacuum. For the effect of N<sub>2</sub>O, the dependency of crosslinking on dose rate, temperature, and gas pressure was reported recently.<sup>3</sup> The accelerating reaction of N<sub>2</sub>O is seemingly not an inhomogeneous one, for example a surface reaction, but a homogeneous one due to dissolved gas in the film.

In the present paper, the effect of thirteen other gases, i.e., He, Ne, Ar, Kr, Xe, N<sub>2</sub>, NO, H<sub>2</sub>O (vapor), CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, and C<sub>4</sub>H<sub>10</sub> besides the eight gases above mentioned is reported. Again, several gases among the thirteen were found to accelerate the crosslinking. As it is our view that the accelerating reaction is probably caused by the dissolved gas in polymer, we examined the gas solubility of various related gases. Michaels<sup>4</sup> measured the solubility of many gases, but not N<sub>2</sub>O. In the present study we measured the solubility of N<sub>2</sub>O by a simple volumetric method.

Secondly, the effect of electric field on the crosslinking during irradiation was studied in several gas atmospheres. At the beginning, we had planned this in order to clarify the dependency of the accelerating effect on electric field, but we could not draw a clear conclusion. On the other hand, the originally induced crosslinking was observed to be dependent on electric field. This dependency is probably related to the induced conductivity of polyethylene during irradiation as discussed by Fowler and Farmer.<sup>5,6</sup>

This paper is concerned only with our experimental results obtained recently. We will discuss the reaction mechanism of accelerating effect in a forthcoming paper. Again, the connection between film thickness and gel content will be studied more fully, since thicker film is generally higher in gel content than a thinner one.

## EXPERIMENTAL

### 1. Irradiation in Various Gases

The experimental procedure was almost the same as described in the previous paper.<sup>2</sup> The film used in the present case was limited to a commercial polyethylene (Sumikathene L-70) having a molecular weight of 28,000 and a density of 0.921 g./cc. Films 0.03 mm. and 0.3 mm. thick were used. Every gas used was commercially available except water vapor. The minimum purity of gas was 99.99% for N<sub>2</sub>, He, Ne, and Ar; 99.9% for CO<sub>2</sub>, Kr, Xe, CO, CH<sub>4</sub>, and N<sub>2</sub>O; 99.8% for C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, and C<sub>4</sub>H<sub>10</sub>; and 99.5% for NO and NO<sub>2</sub>.

In order to prepare the high pressure gas sample, a calculated volume of gas was enclosed in a test tube which was dipped in liquid nitrogen. Irradiation was carried out by  $\gamma$ -rays from a Co<sup>60</sup> source, and gel measurement was by extraction with xylene at 80°C. for 15 hrs.

Furthermore, the solubility of N<sub>2</sub>O in polyethylene was measured by a volumetric method, generally as described by Michaels<sup>4</sup> but ours was rather primitive. Again, the solubility of C<sub>3</sub>H<sub>8</sub> was measured so as to calculate volume fraction of the amorphous portion of polyethylene.

### 2. Application of Electric Field During Irradiation

An irradiation tube was fitted up with plane parallel plates 8.0  $\pm$  0.5 mm. apart, as shown in Figure 1. The plate was made of nickel and the lead wire was platinum. A folded film, 0.03 mm. thick (90 mg. for experiment

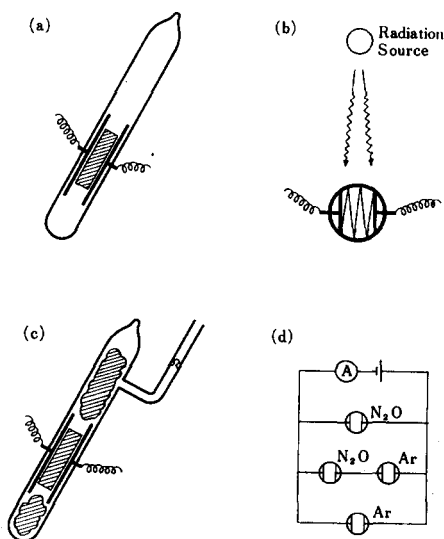


Fig. 1. Apparatus for application of electric field during irradiation: (a) irradiation tube for experiment 1; (b) plane view of irradiation system and cross section of irradiation tube; (c) irradiation tube for experiment 2; (d) circuit for experiment 2.

1, 120 mg. for experiment 2), was inserted between the plates as shown in Figure 1*b*. Moreover, in experiment 2, additional film (1.2 g.) was packed above and under the plates as shown in Figure 1*c* in order to diminish the effect of radiolytic products of  $N_2O$ .<sup>3</sup> Again, in experiment 2, the tube was fitted up with a breakseal joint so as to permit evacuation immediately after irradiation.

The tube was set vertically toward gamma-rays from a radiation source as shown in Figure 1*b*. For experiment 1, five tubes were linked in a parallel circuit and a 1500 v. charge was applied equally. However, in experiment 2, two tubes received 1600 v. and the other two tubes 800 v. in a circuit as shown in Figure 1*d*. In addition, the same tube was used for the control run (blank).

## RESULTS AND DISCUSSION

### 1. Irradiation in Various Gases

**Irradiation in Inorganic Gases.** Results of irradiations in He, Ne, Kr, Xe, CO, and  $H_2O$  are summarized in Figures 2 and 3. The gel content relative to that for vacuum irradiation is plotted against gas pressure. The accelerating effect of Xe and Kr is not so marked at 1 atm., but increases with gas pressure. In case of Xe, the relative gel content of the thinner film shows a decrease at high pressure. We cannot clearly explain this decrease from our present knowledge. In case of  $N_2O$ , the relative gel content decreases at high pressure as shown by the dashed line.

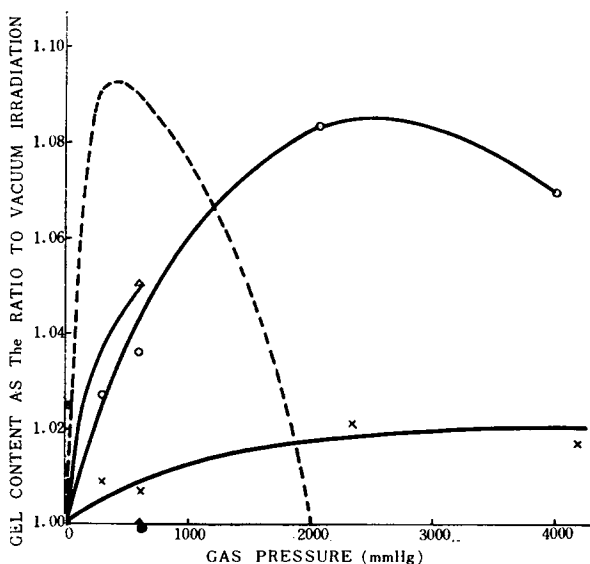


Fig. 2. Increase in gel content of 0.03 mm. thick film on irradiation in various atmospheres: (O) in Xe; (X) in Kr; ( $\Delta$ ) in CO; ( $\bullet$ ) in  $H_2O$ (vapor); ( $\bullet$ ) in He; ( $\blacktriangle$ ) in Ne.

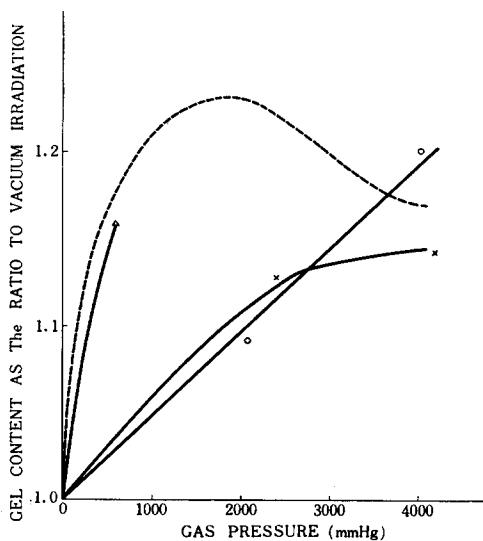


Fig. 3. Increase in gel content of 0.3 mm. thick film on irradiation in various atmospheres: (O) in Xe; ( $\Delta$ ) in CO; ( $\times$ ) in Kr.

This decrease seems to be due to the depressive effect of radiolytic products of  $N_2O$  as previously discussed.<sup>3</sup> In addition, the irradiation in Xe at 600 mm. Hg gives a relative gel content of 1.02 at a dose of  $8 \times 10^6$  r (0.03 mm. thick film).

We have few data on CO and  $H_2O$ , whose effect is also clearly accelerative. At a dose of  $5 \times 10^6$  r, a relative gel content of 1.03 was obtained for the irradiation in water vapor. However, we have some doubt about direct comparison of  $H_2O$  with other inert gases, since the vapor is much different in behavior. We previously assumed that CO is ineffective on the crosslinking,<sup>2</sup> but recent experience has thrown doubt on this assumption, and evidence is accumulating that the effect of CO is probably accelerative.

He and Ne appear to be ineffective. Ar and  $N_2$  also seem to be ineffective, and  $CO_2$  is depressive to some extent. (Our data for  $CO_2$  are not reported in this paper, however, since the data show moderate scattering owing to an accident during our measurements.) We will examine the effect of these three gases in more detail in the near future.

NO depresses the crosslinking markedly. While the gel content for the vacuum irradiation was 74.0% at a dose of  $2 \times 10^7$  r, the gel content for the irradiation in NO at 50 mm. Hg pressure was only 39.7%. NO is known to be an effective radical scavenger not only in general chemical reactions,<sup>7</sup> but also in some radiochemical reactions, as reported by Yang<sup>8,9</sup> and Kuri.<sup>10</sup> Therefore, NO probably reacts with the radicals in polyethylene and hinders the crosslinking.

Since no insoluble part was detected, the effect of  $NO_2$  was previously assumed to be analogous to  $O_2$ . However, the solution viscosity of poly-

ethylene stored in  $\text{NO}_2$  without irradiation showed a large decrease, as shown in Table I. The infrared spectrum of this polyethylene showed the formation of various substitution groups.<sup>11</sup> Polyethylene irradiated in  $\text{NO}_2$  has a solution viscosity higher than that of polymer in  $\text{NO}_2$ , but lower

TABLE I  
Viscosity of Xylene Solution of Film at 75°C.

Treatment	Radiation dose, r	$[\eta]$ , l./g.
Blank	0	0.049
Stored in $\text{NO}_2$	0	0.015
Irradiated in $\text{NO}_2$	$4 \times 10^6$	0.019
Irradiated in $\text{NO}_2$	$8 \times 10^6$	0.032

than that of the original polymer. Thus, the effect of  $\text{NO}_2$  during irradiation cannot be a pure radiation chemical reaction. Irradiated and un-irradiated film were stored in  $\text{NO}_2$  at 600 mm. Hg for the same period (about 3 months).

**Irradiation in Hydrocarbon Gases.** Results of irradiation in saturated hydrocarbon gas are shown in Table II. The relative gel content was more than unity in 0.3 mm. thick film, but was less than unity in 0.03 mm. thick films. At the present, we cannot clearly explain this, but we assume that the effect of the gases may be accelerative in itself.

TABLE II  
Relative Gel Content of Polyethylene Films Irradiated to Various Total Doses at a Dose Rate of  $2.3 \times 10^6$  r/hr. in Hydrocarbon Gases at Gas Pressure of 600 mm. Hg

Gas	Relative gel content of 0.03 mm. film		Relative gel content of 0.3 mm. film	
	$2 \times 10^7$ r	$8 \times 10^6$ r	$2 \times 10^7$ r	$8 \times 10^6$ r
$\text{CH}_4$	0.99	0.98	1.14	1.17
$\text{C}_2\text{H}_6$	0.98	0.98	1.15	1.16
$\text{C}_3\text{H}_8$	0.95	0.93	1.12	1.13
$\text{C}_4\text{H}_{10}$	0.90	0.87	1.08	1.07

Although the data are not shown in Table II, our study indicates that 0.03 mm. thick film evacuated after irradiation gives a relative gel content of more than 1.00. Usually, the irradiated films were stored for about 12 days in the gas before opening the ampule. This postirradiation effect will be treated more fully in a forthcoming paper.

We have no data for unsaturated hydrocarbon gases, but we assume that ethylene and propylene probably depress the crosslinking, because they are known to be effective radical scavengers.

**Gas Solubility in Polyethylene.** Table III shows our results for  $\text{N}_2\text{O}$  and  $\text{C}_2\text{H}_6$ , where  $k$  is the solubility constant, and  $k^*$  is the solubility constant

in a hypothetical, completely amorphous polyethylene, defined by Michaels<sup>4</sup> as:

$$k = \alpha k^*$$

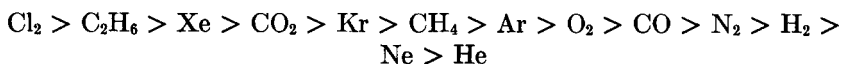
where  $\alpha$  is the volume fraction of amorphous polymer. This relationship indicates that gas is insoluble in the crystalline fraction of the polymer.  $\alpha$  of our polymer is equal to 0.57, calculating from  $k$  and  $k^*$  of  $C_3H_8$ . Thus,  $k^*$  of  $N_2O$  is equal to 0.86.

TABLE III  
Solubility Constant at 25°C.

	Solubility constant, cc. (STP)/cc. (atm.)	
	$k$	$k^*$
$N_2O$	0.49	0.86
$C_3H_8$	2.26	3.97*

\* Data of Michaels and Bixler.<sup>4</sup>

Since the gas dissolved in the film seemingly plays a main role in the accelerating reaction, we examined solubilities of gases concerned. Michaels' data are shown in Table IV. According to Hildebrand et al.,<sup>12</sup> on the other hand, the decreasing order of solubility in organic solvents, including liquid paraffin, is as follows:



This order of solubility can be applied usefully to our case, because liquid paraffin resembles the amorphous part of polyethylene with respect to gas solubility.

TABLE IV  
Solubility Constant  $k^*$  at 25°C.

Gas	$k^*$ , cc. (STP)/cc. (atm.)
He	0.012 ± 0.001
$N_2$	0.0412 ± 0.0013
CO	0.064
$O_2$	0.0769 ± 0.0026
Ar	0.103 ± 0.003
$CH_4$	0.203 ± 0.006
$CO_2$	0.451 ± 0.023
$C_2H_6$	1.28 ± 0.04
$C_3H_8$	3.97 ± 0.06

In conclusion,  $N_2O$  can be said to dissolve to the same degree as Xe, and to be among the most soluble of the inorganic gases. Kr dissolves moderately; CO is sparingly soluble. Hydrocarbon gases dissolve easily, except for  $CH_4$ .

The sorption of water vapor in polyethylene is 0.001 g./cc. at 18 mm. Hg,<sup>13</sup> and the calculated solubility constant  $k$  is equal to 1.2. However, the dissolved state of water would be much different than that of the other gases mentioned above. In addition, the gas solubility would be unchanged in the range of our irradiation dose.<sup>14</sup>

## 2. Effect of Electric Field

In experiment 1, irradiation was carried out in N<sub>2</sub>O, Ar, CO, Xe, and Kr atmospheres. Five irradiation tubes were linked in a parallel circuit and to each was applied 1500 v. The total current was about  $6 \times 10^{-11}$  amp. before irradiation, but this increased to a much higher value during irradiation as shown in Figure 4. There was a so-called plateau region from 400 v. to 2200 v.

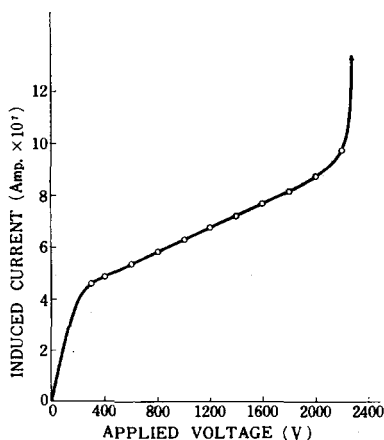


Fig. 4. Induced current in experiment 1 at dose rate of  $2.0 \times 10^6$  r/hr., gas pressure of 300 mm. Hg.

The results of experiment 1 are indicated in Table V. Whatever the atmosphere, the gel content decreases markedly. The value for the N<sub>2</sub>O

TABLE V  
Gel Content in Experiment 1 (Irradiated up to  $8 \times 10^6$  r, at a Constant Dose Rate of  $2 \times 10^6$  r/hr.)

Gas	Gel content, %
N <sub>2</sub> O <sup>a</sup>	68.6
N <sub>2</sub> O	44.4
CO	43.9
Ar	41.4
Xe	41.2
Kr	40.2

<sup>a</sup> Blank test (in the same irradiation tube). Usually, gel content was 66.5% for irradiation in N<sub>2</sub>O, 60.2% for irradiation in vacuum (conventional test tube).

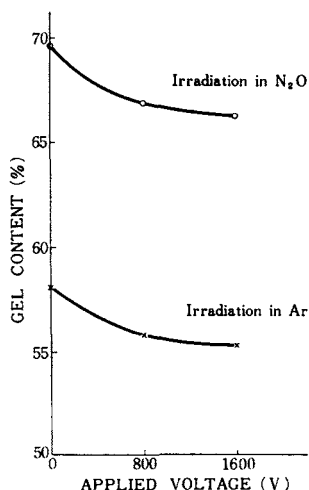


Fig. 5. Decrease in gel content with applied field at dose rate of  $1.8 \times 10^6$  r/hr., total dose of  $8 \times 10^6$  r, gas pressure of 300 mm. Hg.

blank in the irradiation tube was slightly higher than 66.5%, which was a mean value in case of conventional test tube. This higher value would be due to some scattering of  $\gamma$ -rays by electrodes. Irradiation in  $N_2O$  and CO gave higher values than irradiation in Ar, Xe, and Kr. This difference is probably caused by significant accelerating effect of  $N_2O$  and of CO. Although Xe and Kr must be also accelerative, they are not so effective at this pressure, as shown in Figures 2 and 3. Vacuum irradiation was impossible in our case, but we can assume that irradiation in Ar would serve as a substitute for the former.

In experiment 2, the decrease with applied field was examined for the irradiation in  $N_2O$  and in Ar, as shown in Figure 5. Four irradiation tubes were connected in the circuit as shown in Figure 1*d*, and the current in each is indicated in Table VI.

TABLE VI  
Current Before and During Irradiation in Experiment 2 at a Constant Dose Rate of  $1.8 \times 10^6$  r/hr.

	Current, amp.	
	Before irradiation	During irradiation
$N_2O$ (1600 v.)	$3 \times 10^{-11}$	$5 \times 10^{-8}$
Ar (1600 v.)	$7 \times 10^{-11}$	$6 \times 10^{-8}$
$N_2O$ and Ar (800 v.)	$1.5 \times 10^{-11}$	$1.8 \times 10^{-8}$

Our system was composed of a gas phase and solid polyethylene phase. The electrical state of our system during irradiation may be described as follows.



A great part of the current observed must be due to the induced conductivity of gas phase, and only a small part of it may come through polyethylene. It should be noted that both of our experiments were carried out in the so-called plateau region (ohmic-current region). On the other hand, the radiolysis of  $N_2O$  is known to be doubled at about this electric strength,<sup>15,16</sup> but no depressive effect due to the radiolytic products was observed, when the products were absorbed in the abundantly packed film in experiment 2.

The current through polyethylene could not be measured separately in our case, but some conductivity due to the motion of electron must be induced according to the theory of Fowler and Farmer.<sup>5,6</sup> According to these authors, the induced conductivity at equilibrium was  $3 \times 10^{-16}$  ohm./cm. at a dose rate of 8 r/min., when the static conductivity was  $5 \times 10^{-21}$  ohm./cm. Therefore, higher conductivity may be expected in our case, because the dose rate was higher than in the former case ( $2.0-1.8 \times 10^5$  r/hr.).

It is not so easy to explain this decrease in gel content, since this involves various complicated factors. However, we assume that the neutralization between conductive electron and positive ion induced by irradiation in the polymer may be the most probable case for this decrease. According to the ionic mechanism,<sup>17-20</sup> the positive ion (e.g., positively charged methylene group) should play a significant role in radiation-induced crosslinking. While recombination between the positive charge and electron may occur even without the field, the induced current would promote the neutralization to some extent and would depress the crosslinking. This depression, needless to say, may not be due to a radical mechanism such as the effect of  $O_2$  or  $NO$ , but may be due to an ionic mechanism.

On the other hand, in view of the electric strength in our system, this decrease would be difficult to explain in terms of polarization of polyethylene molecule or exposure of film to slightly accelerated gas ions or electrons. Furthermore, it is hardly possible that as a result of irradiation the electrode evolve a gas which depresses the crosslinking (for example  $O_2$  or  $NO$ ).

There was a considerable difference of the decreasing tendency between experiments 1 and 2. We cannot propose any explanation for this difference at the present; however, we want to point out several differences of experimental condition between two experiments, such as weight of inserted film between the electrodes, presence or absence of excess film, volume of evolved hydrogen, and total current, etc.

At the beginning, we had planned these experiments in order to study the dependency of the accelerating effect on electric field, but could not reach any clear conclusion. Although two curves are nearly parallel in Figure 5, in view of the precision of our data we cannot conclude that the accelerating effect, the difference between the two curves, may be independent of electric field. This is because that neither the decreasing ratio due to applied field and the difference of the two curves was so large that we would not be able to observe any appreciable change in the accelerating effect,

even if the effect were diminished in the same ratio as the originally induced crosslinking (normal crosslinking in the case of vacuum irradiation).

Irradiation in  $N_2O$  and CO indicated a rather higher value than irradiation in Ar, Xe, and Kr. This difference appears to be smaller than the difference between irradiation in  $N_2O$  and in vacuum without electric field, even if we take the nonlinear relationship between gel content and degree of crosslinking into consideration. There is some probability that the accelerating effect can be dependent on electric field.

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### References

1. Okada, Y., and A. Amemiya, *J. Polymer Sci.*, **50**, S22 (1961).
2. Okada, Y., *J. Appl. Polymer Sci.*, **7**, 695 (1963).
3. Okada, Y., *J. Appl. Polymer Sci.*, **7**, 705 (1963).
4. Michaels, A. S., and H. J. Bixler, *J. Polymer Sci.*, **50**, 393 (1961).
5. Fowler, J. F., *Proc. Roy. Soc. (London)*, **A236**, 464 (1956).
6. Fowler, J. F., and F. T. Farmer, *Brit. J. Radiol.* **29**, 338 (1956).
7. Laidler, K. J., *Chemical Kinetics*, McGraw-Hill, New York, 1950, p. 190.
8. Yang, K., and P. J. Manno, *J. Am. Chem. Soc.*, **81**, 3507 (1959).
9. Yang, K., *J. Phys. Chem.*, **65**, 42 (1961).
10. Kuri, Z., H. Ueda, and S. Shida, *J. Chem. Phys.*, **32**, 371 (1960).
11. Okada, Y., T. Ito, and A. Amemiya, *Kogyo Kagaku Zasshi*, **61**, 355 (1961).
12. Kobatake, Y., and J. H. Hildebrand, *J. Phys. Chem.*, **65**, 331 (1961).
13. Rouse, P. E., Jr., *J. Am. Chem. Soc.*, **69**, 1068 (1947).
14. Sobolev, I., J. A. Meyer, V. Stannett, and M. Szwarc, *J. Polymer Sci.*, **17**, 417 (1955).
15. Williams, N. T., and H. Essex, *J. Chem. Phys.*, **16**, 1153 (1948).
16. Burt, B. P., and J. F. Kircher, *Radiation Res.*, **9**, 1 (1958).
17. Williams, T. F., *Nature*, **186**, 544 (1960).
18. Weiss, J., *J. Polymer Sci.*, **29**, 425 (1958).
19. Dorfman, L. M., paper presented at the 30th Colloid Symposium, Madison, Wisconsin, 1956.
20. Libby, W. F., *J. Chem. Phys.*, **35**, 1714 (1961).

### Synopsis

Polyethylene film was irradiated in various atmospheres, i.e., He, Ne, Ar, Kr, Xe,  $N_2$ , NO,  $NO_2$ ,  $H_2O$ , CO,  $CO_2$ ,  $CH_4$ ,  $C_2H_6$ ,  $C_3H_8$ , and  $C_4H_{10}$  by  $\gamma$ -rays from a  $Co^{60}$  source. Xe, Kr, CO, and  $H_2O$  (vapor) accelerate the radiation-induced crosslinking as recently shown in case of  $N_2O$ . In addition, saturated hydrocarbon gases seem to be accelerative. NO depresses the crosslinking markedly and  $CO_2$ , mildly. On the other hand,  $NO_2$  decomposes the polyethylene molecule without irradiation. Every other gas appears to be inactive. In relation to the effect of gas atmospheres, we examined their solubilities in polyethylene.  $N_2O$  is one of the most soluble of the inorganic gases. An electric field was applied to polyethylene film during irradiation. The degree of crosslinking evidently decreases with applied field, whether the atmosphere is  $N_2O$  or Ar.

### Résumé

On a irradié un film de polyéthylène par rayons- $\gamma$  du  $Co^{60}$  dans diverses atmosphères, c'est à dire, He, Ne, Ar, Kr, Xe,  $N_2$ , NO,  $NO_2$ ,  $H_2O$ , CO,  $CO_2$ ,  $CH_4$ ,  $C_2H_6$ ,  $C_3H_8$ , et  $C_4H_{10}$ . Comme on l'a démontré récemment dans le cas du  $N_2O$ , le Xe, Kr, CO, et  $H_2O$  (vapeur)

accélèrent le pontage induit par irradiation. De plus, les gaz saturés d'hydrocarbures semblent avoir un effet d'accélération. NO diminue fortement le pontage, l'oxyde de carbone faiblement. D'autre part, NO<sub>2</sub> décompose la molécule de polyéthylène en absence d'irradiation. Tous les autres gaz semblent inactifs. Quant à l'effet des atmosphères gazeuses, nous avons examiné leurs solubilités dans le polyéthylène. N<sub>2</sub>O est l'un des gaz les plus solubles parmi les gaz inorganiques. On applique un champ électrique au film de polyéthylène durant l'irradiation. Le degré de pontage diminue évidemment proportionnellement au champ appliqué, que l'atmosphère soit du N<sub>2</sub>O ou de l'Ar.

### Zusammenfassung

Polyäthylenfolie wurde in verschiedenartiger Atmosphäre, nämlich He, Ne, Ar, Kr, Xe, N<sub>2</sub>, NO, NO<sub>2</sub>, H<sub>2</sub>O, CO, CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, und C<sub>4</sub>H<sub>10</sub> mit  $\gamma$ -Strahlen aus <sup>60</sup>Co bestrahlt. Xe, Kr, CO, und H<sub>2</sub>O (Dampf) beschleunigen die strahlungsinduzierte Vernetzung in gleicher Weise wie es kürzlich für N<sub>2</sub>O gezeigt wurde. Auch gasförmige gesättigte Kohlenwasserstoffe scheinen beschleunigend zu wirken. NO unterdrückt die Vernetzung merklich, CO<sub>2</sub> nur schwach. Andererseits zersetzt NO<sub>2</sub> das Polyäthylenmolekül ohne Bestrahlung. Alle anderen Gase scheinen inaktiv zu sein. In Zusammenhang mit dem Einfluss der Gasatmosphäre wurde die Löslichkeit der Gase in Polyäthylen untersucht. N<sub>2</sub>O ist eines der am besten löslichen anorganischen Gase. Bei Anlegen eines elektrischen Feldes an die Polyäthylenfolie während der Bestrahlung nimmt der Vernetzungsgrad sowohl in N<sub>2</sub>O als auch in Ar mit der angelegten Feldstärke ab.

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