

# Effect of Container Diameter on $\beta$ -Radiation-induced Polymerization of Acrylamide

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## Synopsis

Beta particles emitted by radioactive nuclei have a finite depth of penetration in a given medium and hence a certain linear energy transfer value depending upon their initial energy. This property can be utilized to achieve different rates of radiation induced reactions. Various diameter capillaries containing saline solutions of acrylamide and radioactive phosphorus-32 were used in the experiment and the change in viscosity of acrylamide solution was used as an indicator of the amount of energy transferred. Distinctly different rates were observed which were roughly proportional to the diameters of the capillaries containing the solutions. It also appears that irradiation of the polymer solution at 80°C even in the absence of oxygen has an adverse effect on the viscosity. The experimental conditions were chosen to approximate those existing in oil reservoirs.

## INTRODUCTION

It is a common practice in the petroleum industry in conjunction with enhanced oil recovery operation to inject heat-sensitive mixtures into the oil reservoirs to plug up faulty geological formations or thief zones. These mixtures form a gel after a predetermined time at the elevated temperatures existing in the oil reservoirs. The formation of a tough crosslinked gel in the general area inhibits water or water-polymer-surfactant, or any other pusher fluid from penetrating the faulty formation. This improves the sweep characteristics and stops the undesirable loss of the sweeping fluid. Various combinations of chemicals, e.g., lignosulfonates with sodium dichromate and salt,<sup>1,2</sup> partially hydrolyzed poly(acrylamide) with aluminum sulfate and citrate ions,<sup>3</sup> sodium dichromate with sodium bisulfite,<sup>4</sup> and many different types of clays and biopolymers,<sup>5,6</sup> have been used for this purpose. Such a process has a disadvantage in that the formation of a gel in the general area of the fault will indiscriminately plug up all the openings, whether big or small. Consequently, the oil contained in the smaller pores cannot be recovered. A need, therefore, was evident to design a process by which only those cracks or holes that are bigger than a certain size can be closed, leaving the fine porous and oil rich surroundings open to the sweeping fluid.

The problem was defined as a process in which different extents of reaction of crosslinking could be achieved in different containers. Stated otherwise, higher rates of reaction are desired in the bigger pores and correspondingly lower rates of reaction in the smaller pores, so that in a given period of time significant amount of reaction would have occurred only in those holes which exceed a cer-

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tain diameter. An additional restraint implicit in the problem was that the reacting system should not be activated simply by thermal means. Briefly, then, the agent activating the chemical reaction should be capable of controlling the rate of reaction according to the diameter of the container independently of the reaction temperature. A possible solution to the problem was devised by combining two facts: (1) alpha and beta radiations have finite depths of penetration in a given medium; and (2) certain chemical reactions, e.g., polymerization and crosslinking, etc., can be induced by radiation.

The choice of the monomer acrylamide was made because poly(acrylamides) in various forms are already extensively used in enhanced oil recovery processes,<sup>7-13</sup> and because this monomer can be polymerized easily by radiation to a high molecular weight product. Monomer and polymer both are water soluble. The choice of <sup>32</sup>P as the source of  $\beta$  radiation was prompted by its short half life (14.22 days), easy availability, and the fact that the  $\beta$  particles emitted by <sup>32</sup>P nuclei have a moderate depth of penetration (7.9 mm maximum) in water. This radioactive isotope of phosphorus-32 decays according to the following scheme:



with the energy of disintegration (1.71 MeV) being randomly divided between the emitted  $\beta$  and the antineutrino. Because the interaction cross section of a neutrino is extremely small and it passes through matter virtually without any interaction, energy deposition from the neutrino may be neglected. The only energy available for interaction is, therefore, that of the electron, and its energy distribution function<sup>14(a)</sup> is similar to the normal distribution, with a maximum and average energy of 1.71 and 0.70 MeV, respectively. These electrons interact with the surrounding molecules in a variety of ways to yield ions, free radicals, and molecules in excited states. After reaching thermal energy, the electron may reduce a cation or may remain temporarily solvated in the medium. The average linear depth an electron can travel in a medium will depend upon its initial energy and the density of the medium, although the actual length of the zig-zig path could be several folds higher. The maximum range in water of a monoenergetic electron of energy  $E$  up to 2.5 MeV can be calculated by the Katz and Penfold expression<sup>14(b)</sup>

$$\text{Range (cm)} = 0.412 E^n \rho^{-1} \quad (1)$$

in which  $n$  is  $(1.265 - 0.0954 \ln E)$  and  $\rho$  is the specific gravity of the medium.

The depths of penetration for average- and maximum-energy electrons emitted by <sup>32</sup>P are calculated to be 0.25 and 0.79 cm, respectively, in water.

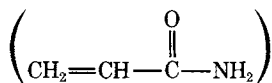
It should be noted that because of frequent scattering and large losses, a high energy electron may not always travel as deep as a low energy electron. The probability of any electron of a given energy penetrating a given depth in water is also well-established experimentally.<sup>15</sup> However, one can make a general statement that on an average the  $\beta$  particles emitted from <sup>32</sup>P nuclei will penetrate approximately 0.25 cm in water, and that the number of  $\beta$  particles that would penetrate beyond twice this depth will be negligible. Future considerations will be based on these assumptions, and the distance beyond which the number of particles is not significant will be defined as  $r_c$  or the critical depth of penetration.

Imagine a uniform concentration of these radioactive particles dissolved in water and imagine a small volume element in this medium [Figs. 1(A), (B)]. Only those electrons that are emitted by radioactive nuclei lying within a sphere of radius  $r_c$  will be able to reach this fluid element and transfer part of their energy to it. Any radioactive source beyond this critical distance will have no effect on this volume element. Now, if this element were situated in an infinitely long cylinder such that the diameter of the cylinder is less than this critical distance  $r_c$  [Fig. 1(B)] then, since there are no radioactive sources in the hypothetical sphere beyond the wall of the cylinder, the fluid element in effect has lost the energy that could have come from the radioactive sources contained in the void  $V_V$ . In other words, the same volume element in a medium of identical radioactivity will receive different energies depending upon the size and shape of the container. In this case, if the intensity of radiation received by the elements in geometry  $A$  is  $I_A$  and that in geometry  $B$  is  $I_B$  then,

$$\frac{I_B}{I_A} = \frac{V_A}{V_A + V_V} \cong \frac{\pi r^2 2r_c}{\frac{4}{3} \pi r_c^3} = \frac{3}{2} \left(\frac{r}{r_c}\right)^2 \quad r \ll r_c \tag{2}$$

A qualitative verification of such a conclusion can be obtained by running an experiment in which the reactants are sensitive to radiation and the products formed are related to the amount of radiation received.

The radiation induced polymerization of acrylamide:



has been amply studied.<sup>16-25</sup> The monomer polymerizes through free radical mechanism and is known to be one of the most easily polymerizable monomers. The waterlike solution of the monomer increases in viscosity as the reaction proceeds and the polymer forms. The rate of increase of viscosity depends upon the initial concentration of the monomer and the total dose of radiation received. For this study, a 3% (w/v) solution of acrylamide in water containing 10% (w/v) sodium chloride was used. The reactions were run at 80°C. High temperature and the presence of salt was necessary to simulate the conditions existing in typical oil reservoirs.

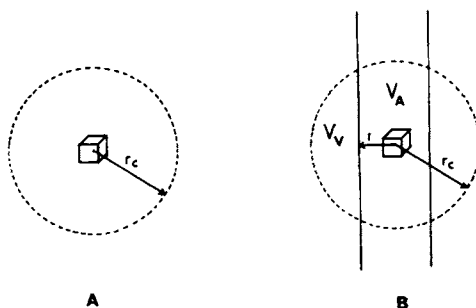


Fig. 1. Hypothetical volume element in radioactive solution contained in vessel. (A) Size of vessel much larger than maximum depth of penetration of radiation. (B) Size of vessel smaller than depth of penetration of radiation.

## EXPERIMENTAL

The activity of the solution used in the internal irradiation experiments was 1 mCi/ml at the time the samples were prepared. This activity will decrease exponentially according to the law of radioactive decay. Phosphorus-32 in the form of orthophosphoric acid in 0.02*N* hydrochloric acid was diluted with 10% saline water and neutralized to pH 7.0 with 0.1*N* sodium hydroxide. Separate solutions of 10% sodium chloride and 6% acrylamide in 10% salt solution were prepared and filtered. Calculated quantities of each were mixed together to obtain the desired level of concentration of each component. The acrylamide was used as received.

Four different diameter tubes were chosen for the internal irradiation experiments: 0.38 mm (Teflon), 0.86 mm (Teflon), 2.29 mm (Tygon), and 6 mm (glass). The length of the capillary tubes was calculated so it could contain approximately 1.2 ml of the solution. The problem of handling very long tubes was overcome by wrapping them on a 6 × 6-in. wire screen such that the parallel segments of a tube were at least 6.5 mm apart (Fig. 2). Thus, the probability of a segment of a tube being affected by high energy electrons from another segment nearby would be negligible.

Experiments were run in a substantially oxygen-free atmosphere. Distilled water used in the experiment was further deoxygenated by boiling for about 10 min. The ends of the Tygon or Teflon tubes were sealed by melting and the vapor space above the sample in the vials used in <sup>60</sup>Co irradiation was replaced with nitrogen. Only the 6-mm diam tube sample was used repeatedly for viscosity measurements. Final viscosity measurements were made on all other samples (including those irradiated externally with <sup>60</sup>Co) after uninterrupted exposure for the desired period. Once opened, these samples were not used again.

A Brookfield cone-and-plate viscometer was used to take viscosity readings at 25°C. The viscosity of the sample in the 6-mm diam tube was measured in a rolling ball viscometer (0.25-in. diam steel ball in a 7-mm glass tube inclined at an angle of 30°, distance traveled by the ball 10 cm). This viscometer was standardized by comparing the roll time of the ball to the viscosity obtained on the Brookfield viscometer.

The external irradiation experiments were conducted in a cobalt irradiation source seated in the bottom of a 15-ft deep pool of water. The temperature bath containing the sample vials could be kept inside a cylindrical vessel which could

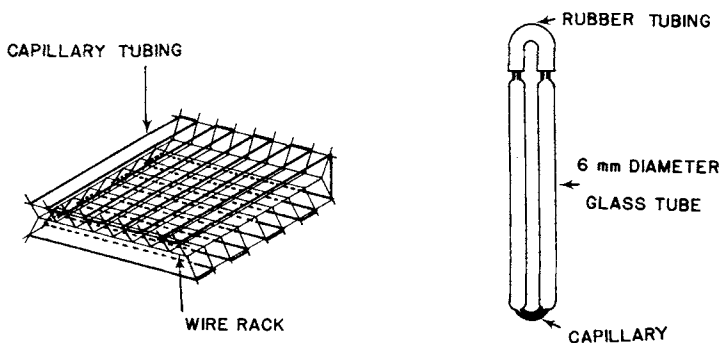


Fig. 2. Sample tubes.

be lowered into the pool to sit snugly into the hollow of the annular casing (Fig. 3). The temperature bath was partially filled with water which was heated to provide the desired temperature in the vapor space where the samples were kept. The electricity to the heater was regulated by a rheostat and the temperature of the vapor space was monitored by a thermocouple. The temperature was controlled within  $\pm 2^\circ\text{C}$ . Dosimetry studies were done in this setup by the Fricke procedure,<sup>26</sup> and the samples were irradiated in the identical geometric setup. The dose rate for the  $^{60}\text{Co}$  source was 350 rad/min.

## RESULTS AND DISCUSSIONS

The relative viscosities of the polymer solutions irradiated externally at 28 and  $80^\circ\text{C}$ , and of those using internal radiation at  $80^\circ\text{C}$  in different diameter tubes are presented in Table I. The relative viscosity is the actual viscosity of the solution divided by the viscosity of the monomer solution (3% acrylamide, 10% salt). The total apparent (i.e., maximum possible) dose  $D$  received by the samples in  $^{32}\text{P}$  irradiation experiments can be calculated from<sup>14(c)</sup>:

$$D = 0.593 \frac{\bar{E} C_o}{\lambda \rho} (1 - e^{-\lambda t}) \text{rad} \quad (3)$$

where  $\bar{E}$  is the average energy of the  $\beta$  particle = 0.70 MeV;  $\lambda$  is the radioactive decay constant =  $0.693/t_{1/2}$ ;  $t_{1/2}$  is the half life equal to  $3.413 \times 10^2$  hr;  $t$  is the time elapsed in hr;  $C_o$  is the initial activity in mCi/ml = 1.0; and  $\rho$  is the density of the solution = 1.13 gm/ml.

This expression does not take the size of the container into account. It is clear from the preceding arguments that different containers will intercept different

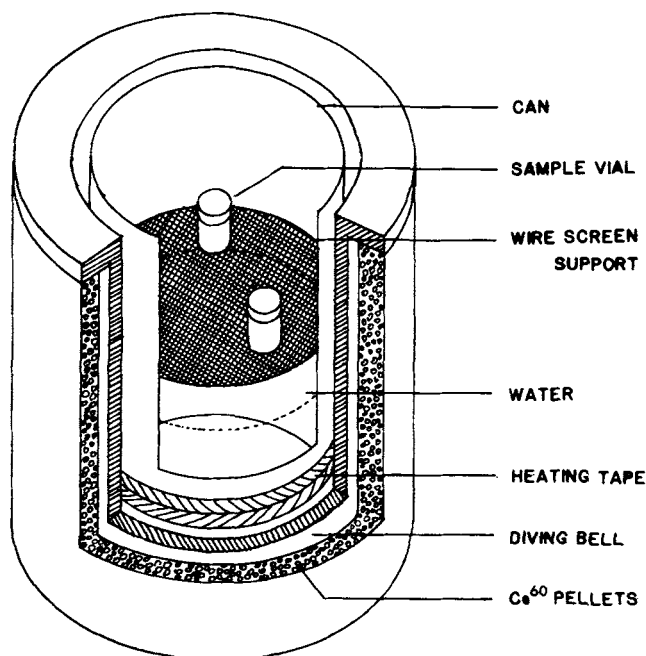


Fig. 3.  $^{60}\text{Co}$  irradiation facility.

TABLE I  
Relative Viscosities of Irradiated Solutions of 3% (w/v) Acrylamide and 10% (w/v) Salt

Dose rads $\times 10^{-4}$	Irradiation in $^{60}\text{Co}$ pit		Internal irradiation with $^{32}\text{P}$ at $80^\circ\text{C}$ diam. of capillary, mm			
	28°C	80°C	6	2.29	0.86	0.38
	1.70	—	—	5.7		
2.1	16.8	21.1				
3.7	—	—	20.8			
4.3	54.0	—				
4.4	—	27.5				
6.4	63.1	28.1				
8.5	—	—		19.5	4.9	1.6
10.7	65.5	23.4				
15.0	67.1	17.7	19.0			
18.9	—	—	15.8	11.0	5.8	2.6
19.7	96.6	16.2				
23.4	209.5	—				
23.9	—	—	11.1			
27.7	—	8.8				
30.7	gel	—				
31.3		—	7.8			
41.1		—	5.7			
51.1		—				
51.6		9.1	4.2			
59.4		—		2.4		

fractions of emitted radiant energy depending in a complex manner upon the ratio of the characteristic size and the maximum depth of penetration, [see Fig. 1(A), (B)], and therefore the true dose will be a complex function of this ratio. Equation (3) yields a number which assumes total absorption of all average-energy  $\beta$  particles.

In the case of irradiation with  $^{60}\text{C}$ , the expression used was

$$D = I_0 t \text{ rad} \quad (4)$$

where  $I_0$  is the intensity of radiation =  $2.13 \times 10^4$  rad/hr and  $t$  is the exposure time in hr.

A plot of the relative viscosities of the samples in different diameter tubes against the total apparent radiation dose absorbed appears in Figure 4. It is clear from the figure that the same solution contained in different diameter pores will increase in viscosity at characteristically different rates and that this simply results from different diameter tubes intercepting different amounts of radiation. The dependence of the efficiency of energy transfer on diameter becomes more evident from Figure 5, in which the relative viscosities of solutions in different tubes is plotted against their diameters after they have received total apparent doses of  $5 \times 10^4$  and  $8.5 \times 10^4$  rad. As anticipated, little or no change in the efficiency of energy absorption is seen once the diameter of the container is larger than the maximum depth of penetration of the  $\beta$  particles, for which the fraction of intercepted energy is unity. The asymptotic curve can be predicted from mathematical considerations also, which will be published separately. However, two very interesting observations can be made at this point. First, concerning the viscosity data obtained from samples irradiated externally by  $^{60}\text{Co}$  at  $80^\circ\text{C}$ , plotted as curve A in Figure 6, only slightly higher sample viscosities were ob-

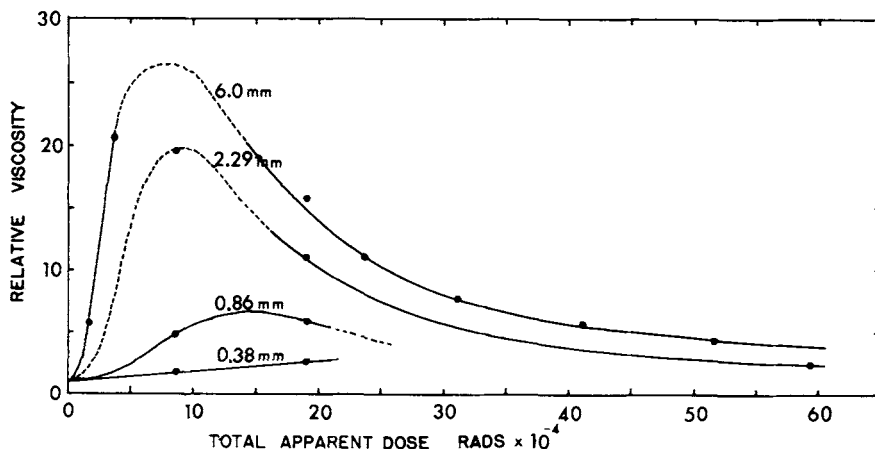


Fig. 4. Relative viscosities of samples in different diam tubes containing dissolved <sup>32</sup>P vs. total apparent dose received (initial intensity 1 mCi/ml).

tained in the case of irradiation externally by <sup>60</sup>Co when compared at the same total dose with the viscosities of the sample in the 6-mm diam tube. The maximum viscosity attained in the <sup>60</sup>Co irradiation at 80°C was about 27 cP, and this is also the value which viscosity-versus-diameter lines in Figure 5 approached asymptotically. Since the  $\gamma$  rays from radioactive <sup>60</sup>Co have a half-thickness value (depth needed to attenuate the intensity of the original beam to half) of 11 cm in water and the radius of the vial was about 1.25 cm, the system could be imagined to be equivalent to an internally irradiated solution contained in an infinitely large vessel.

Second, concerning the rate of increase of viscosity of the samples in different diameter tubes: If in the early stages of reaction the increase in viscosity could be assumed to be proportional to the rate of reaction,<sup>27</sup> the initial slopes of these curves will be proportional to the initial rates of reaction in different diameter tubes. These slopes are calculated and presented in Table II. Again, at low concentrations and low viscosities when the polymer molecules are not very large,

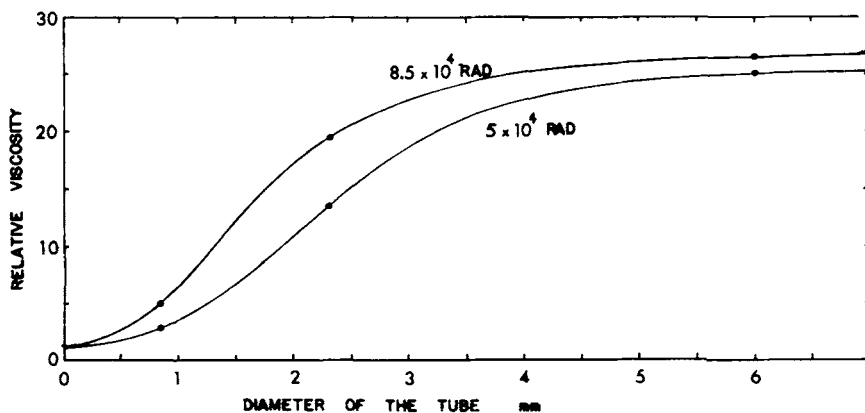


Fig. 5. Relative viscosities attained as function of tube diam at two total apparent doses.

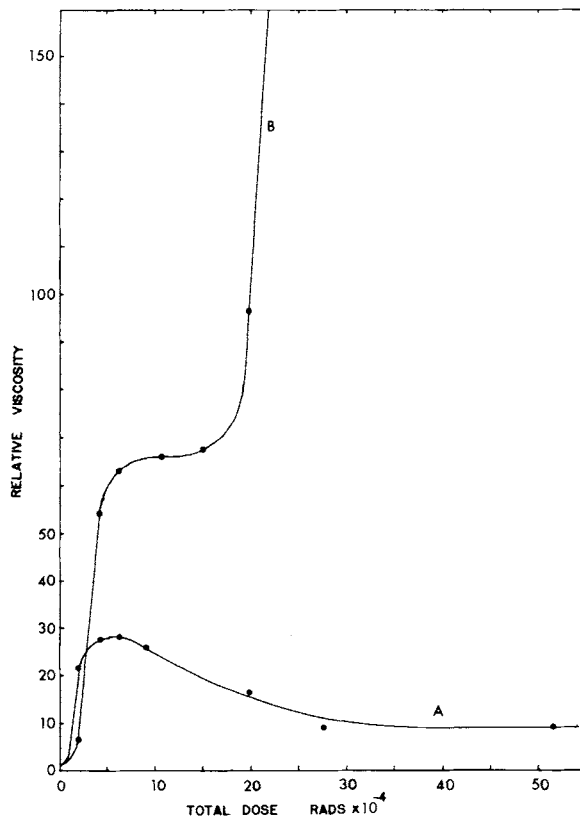


Fig. 6. Relative viscosities of samples irradiated with  $^{60}\text{Co}$  vs. total dose received at two temperatures. Curve A =  $80^\circ\text{C}$ ; curve B,  $28^\circ\text{C}$ .

the rate of reaction is proportional to the square root of the intensity of irradiation.<sup>28</sup> Substituting this into eq. (2),

$$\frac{R_B}{R_C} = \left(\frac{I_B}{I_C}\right)^{1/2} = \left(\frac{3}{2}\right)^{1/2} \frac{r_B}{r_C} \quad (5)$$

i.e., the ratio of the slopes of the curves in Figure 6 should be proportional to the ratios of their diameters. The results of the calculations are presented in Table

TABLE II  
Comparison of Ratio of Slopes of Relative Viscosity Curves with Ratio of Diameters

diam. mm	Slope of rel. visc., curve $R$	Ratio of slopes ( $R/R_C$ )	Ratio of diam. ( $r/r_c$ )	$R/R_C$ from eq. (5)
15.8 <sup>a</sup>	15.59	1.0	1.0	1.0
6.0	7.52	0.484	0.380	0.465
2.29 <sup>b</sup>	—	—	—	—
0.89	0.82	0.053	0.054	0.066
0.38	0.10	0.006	0.024	0.030

<sup>a</sup> Critical depth of penetration  $r_c = 7.9$  mm.

<sup>b</sup> Calculations based on results from 2.29-mm diam tube are not included because of lack of suitable data point to determine the slope.



II. The agreement seems to be quite satisfactory considering the nature of assumptions involved. The rate of reaction in a hypothetical tube of critical radius (corresponding to a maximum depth of penetration  $r_C = 7.9$  mm) was calculated as the slope of the viscosity-versus-dose curve in the  $^{60}\text{Co}$  irradiation experiment. The large difference in the two calculated values at the smallest diameter may result from the large induction period at very low dose rate, which when considered would give a higher rate of reaction than the one used in Table II. In addition, the fact that many  $\beta$  particles could simply escape from a very small tube before they start interacting, would also decrease the rate of reaction.

Since the purpose of the investigation was to polymerize the monomer with the eventual formation of gel, it was somewhat disconcerting to find that the viscosities of the samples decreased after certain maxima were reached depending upon the diameter. One possible interpretation is that the polymer was undergoing some sort of degradation. That this is not the result of some peculiarity in the nature of internal irradiation is apparent from the fact that samples irradiated externally with  $^{60}\text{Co}$  exhibited similar behavior. To ascertain whether this depolymerization resulted from the high reaction temperature or the radiation itself, another set of experiments using solutions of the same monomer and salt concentrations were irradiated at room temperature ( $28^\circ\text{C}$ ) in the  $^{60}\text{Co}$  pool. An interesting phenomena was observed in addition to the expected behavior. Initially the viscosity of the solution (see curve B, Fig. 6) rapidly increased, reached a plateau (perhaps even decreased somewhat), and then increased rapidly to form a clear crosslinked gel; on further irradiation the gel shrank in volume, expelling water. Similar results on crosslinking and shrinkage of gel upon irradiation have been reported,<sup>23</sup> starting from poly(acrylamide), poly(acrylic acid), poly(vinyl alcohol), and poly(vinyl pyrrolidone). The fact that the same monomer and salt solution crosslink at lower temperature but degrade at higher temperature, even at a total dose well beyond that needed to crosslink, generally obviates an assumption of possible oxidative degradation unless the formation of  $\text{H}_2\text{O}_2$  in irradiated water increases significantly at higher temperatures. Prolonged irradiation of aqueous solution of acrylamide at room temperature well beyond the formation of gel, does not appear to have been reported previously.

Next, a polymer solution was prepared from commercial poly(acrylamide) (obtained from Polyscience Inc., MW  $5 \times 10^6$ ). The viscosity of this solution was the same as the maximum viscosity ( $\sim 30$  cps) obtained in the samples irradiated at  $80^\circ\text{C}$ . This solution was kept in a temperature bath at  $80^\circ\text{C}$  for a prolonged period of time. No significant decrease in viscosity was observed. Similar results were obtained with polymer solutions prepared from the monomer under  $^{60}\text{Co}$  irradiation and then kept at  $80^\circ\text{C}$  for prolonged period without irradiation. The conclusion to be drawn from these experiments is that irradiation at high temperature is responsible for the deleterious effect on the solution viscosity.

Further experimental work to elucidate the phenomenon of decreasing viscosity and means to inhibit it is in progress. Since little or no control can be exercised on the temperature of the oil reservoirs, a more feasible and attractive alternative is to use crosslinking agents in solution with the monomer. Again, because of the small number of data points and their location on the graph, it was not possible to quantitate the phenomenon of dependence of energy transfer

efficiency on the container diameter. The validity of the experimental points can be reinforced by mathematical considerations. Such a treatment will be considered elsewhere.

### CONCLUSION

It has been demonstrated that selective rates of radiation-induced reactions can be achieved by combining the information on the depth of penetration of  $\beta$  particles with an appropriate choice of the diameter of the vessel containing the dissolved radioactive substance. Similar considerations could be applied to  $\alpha$  particles and to a much lesser extent to  $\gamma$  radiation. Practical applications of such a result are as yet unexplored and pose interesting possibilities. Further experimental, feasibility, and safety studies must be done before the process could be applied to achieve selective plugging of larger holes in the oil reservoirs, in order to improve sweep efficiency while at the same time leaving the smaller pores open to the flow of the flooding fluid.

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