# Radiation-Induced Sol-Gel Transition of Protein: Effects of Radiation on Flow and Melting Properties

Ionizing radiation strongly influences the chemical and conformational properties of biological macromolecules,<sup>1,2</sup> and also, some biological macromolecules, such as proteins, form a hydrogel.<sup>3-5</sup> Since the radiation-induced sol-gel transition of protein poses a problem of considerable interest, it was considered desirable to study the effects of radiation on the flow and melting properties. The protein used in this work was gelatin since it was described in a previous article.<sup>6</sup>

#### **EXPERIMENTAL**

Gelatin used as the model protein in this work was a commercial material produced by the Kanto Chemical Co., Ltd.

In irradiation, the solid gelatin was irradiated by  $^{60}$ Co gamma rays in air at room temperature at dose rates of  $6.0 \times 10^4$  to  $1.3 \times 10^5$  rad/h.

In viscometry, the irradiated solid gelatin was dissolved in distilled water at about 80°C, and cooled at 25°C for 24 h. Then the viscosity of the solution was measured.<sup>6</sup>

In thermometry, the irradiated solid gelatin was dissolved in distilled water at about  $80^{\circ}$ C and gelatinized at  $2^{\circ}$ C for 24 h. The gelatin hydrogel was then warmed at a rate of  $0.2^{\circ}$ C/min, and the melting point was measured by the method of Eldridge and Ferry.

In calculation of heat of reaction for cross-linking process, the heat energy required to dissociate cross-links of the gelatin hydrogel was calculated using the melting point given by the equation of Eldridge and Ferry.

$$\log_{10} C = \Delta H/2.303RT + \text{constant} \tag{1}$$

where C is the gelatin concentration (g/L),  $\Delta H$  is the heat of reaction for the cross-linking process of the gelatin hydrogel (kcal/mol of cross-links), R is the gas constant, and T is the melting point of the gelatin hydrogel (K). Equation (1) is converted to Eq. (2)

$$\Delta H = \left( K \log_{10} C_1 / C_2 \right) / \left( 1 / T_1 - 1 / T_2 \right) \tag{2}$$

 $k = 2.303 \times R$ 

#### **RESULTS AND DISCUSSION**

The changes in reduced viscosity of gelatin at various radiation doses were studied with 10% gelatin at 25°C. Figure 1 shows the relation between the values of the reduced viscosity and the radiation dose. The changes in reduced viscosity of gelatin at different times after irradiation  $(5 \times 10^6 \text{ rad})$  were studied with 10% gelatin at 25°C. Figure 2 shows the relation between the values of the reduced viscosity and the time after irradiation. From these results it is clear that the reduced viscosity is decreased depending upon the irradiation until a certain value. If such changes in reduced viscosity are due to destruction or restoration of the sol form of gelatin molecule, increased radiation dose or increased elapsed time after irradiation should result in further destruction or restoration, and the reduced viscosity should continue to decrease or recover until a certain value.

The changes in melting point of gelatin at various radiation doses were studied with 3-10% gelatin. Figure 3 shows the relation between the values of the melting point and the radiation dose. Also, the changes in heat energy required to dissociate cross-links of gelatin with irradiation were estimated by the equation of Eldridge and Ferry.<sup>1</sup> Figure 4 shows the relation between the values of the heat of reaction and the radiation dose. While, the changes in melting point of

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Fig. 1. Viscosity vs. radiation dose (10% gelatin hydrosol at 25°C).



Fig. 2. Viscosity vs. time after irradiation (10% gelatin hydrosol,  $3 \times 10^6$  rad and  $25^{\circ}$ C.



Fig. 3. Melting point vs. radiation dose (10% gelatin hydrogel).



Fig. 4. Heat of reaction vs. radiation dose.



Fig. 5. Melting point vs. time after irradiation (10% gelatin hydrogel and  $3 \times 10^6$  rad).



Fig. 6. Heat of reaction vs. time after irradiation  $(3 \times 10^6 \text{ rad})$ .

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gelatin at different times after irradiation  $(3 \times 10^6 \text{ rad})$  were studied with 3–10% gelatin. Figure 5 shows the relation between the values of melting point and the time after irradiation. Figure 6 shows the relation between the values of heat of reaction and the time after irradiation. From the results it is clear that the melting point and the heat of reaction are decreased depending upon the irradiation, while that of the melting point and the heat of reaction are increased depending upon the elapsed time after irradiation until certain values. If such changes in melting point and heat of reaction are due to destruction or restoration of cross-links of gelatin molecule, increased radiation dose or increased elapsed time after irradiation should result in further destruction or restoration, and the melting point and the heat of reaction should continue to decrease or recover until certain values.

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