Chemical and physical properties of poly (vinyl alcohol) hydrogel films formed by irradiation*

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Abstract Chemical and physical properties of poly(vinyl alcohol) (PVA) hydrogel films were investigated as a function of production factors. The experimental results show that the gel fraction depends strongly on the radiation dose, the degree of swelling is inversely dependent on the concentration of PVA solution, the tensile strength depends mainly on the PVA blending ratio and the elongation at break is inversely dependent on the radiation dose.
Keywords Poly(vinyl alcohol), Radiation, Hydrogels, Properties

1 Introduction

Hydrogel is a polymer material which exhibits the ability to swell in water and retains a significant fraction of water within its structure, but which will not be dissolved in water. Radiation-crosslinked hydrophilic polymers-hydrogels have been proved to be specially useful for medical and pharmaceutical purpose. They show usually good biocompatibility and are widely applied as artificial cornea, contact lenses, cardiovascular implants, drug delivery systems, separators for hybrid-type artificial organ, etc.[1]. Poly(vinyl alcohol) is a hydrophilic polymer that has excellent biocompatibility. It is widely used in many fields, specially in the biomedical field. For example, it is used in prosthetics, it also is used as a hemodialyzer membrane and as suture material, and its applicability to artificial microvessels or an artificial hyaloid body is being investigated.[2] Although numerous works have been reported on productions and properties of hydrogels, clinical applications of the hydrogels still are quite few. One of the major reasons may be that the mechanical strength of the hydrogels is too low for the clinical use as wound dressing. In order to improve the mechanical strengths of the PVA hydrogels, the present paper investigated the influence of production factors on the chemical and physical properties of the PVA hydrogel films. The properties include gel fraction, degree of swelling, tensile strength and elongation at break.

The production factors investigated were the concentration in PVA aqueous solution, PVA blending ratio in its blend solution with poly(ethylene oxide) (PEO), together with the radiation dose applied to the systems. The investigation method involved the factorial design of experiments in which four levels of PVA concentration and four levels of the PVA blending ratio were taken into account, together with five levels of applied radiation dose.

2 Materials and methods

All the materials used in this study were commercially available: PVA was supplied by Kuraray Co. Ltd, Japan. PEO was produced by Ichimaru Pharcos Co. Ltd., Japan. The concentration of PVA solution investigated was 5%, 10%, 15% and 20% W/V, the concentration of PVA solution blending with PEO was 10% and 20% W/V in which the PVA blending ratio were 20% and 40% and the level of radiation dose was 20, 40, 60, 80 and 100 kGy. To prepare the aqueous solutions, the polymers were added to distilled and deionized water, then stirred and heated to the boiling point at normal pressure. After obtaining a completely homogeneous solution it was poured into glass-covered 12 cm diameter tissue-culture dishes to a depth of 2 mm. Then they were irradiated using electron beams at current of 10 mA and acceleration energy of 1.7 MeV generated from a KFFG-1 Electric Beam Accelerator made by Shanghai Institute of Nuclear Research to yield crosslinked hydrogel films, at a dose rate of 20 kGy/pass and

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room temperature of 25°C. After irradiation the mechanical properties such as tensile strength and elongation at break were measured using an XLD-A tensile testing machine (Changchun Second Testing Machine Factory, China) with a traction velocity of 150 mm/min. The samples of dumbbell shape were 35 mm in length, with a central cross section of 4 mm. The thickness of samples was measured using a thickness tester (China). The gel fraction and the degree of swelling for the samples were obtained following our previously described methods.[3]

Table 1 Gel fraction (%)

Dose/kGy	PVA concentration/%			
	5	10	15	20
20	85.1	68.4	52.3	35.0
40	93.0	92.7	80.6	74.1
60	91.4	96.7	87.9	87.2
80	98.5	99.2	92.6	90.5
100	96.8	96.5	92.1	90.5

3 Results and discussion

In order to evaluate the results of each combination of factors, the mean values of three samples were calculated and are reported in Tables 1, 2, 3 and 4. The first step in the interpretation of the results was to verify that the main and mutual effects of the various factors have been estimated at an acceptable significance level, α=0.01. To this end an analysis of variance was performed with the results as presented in Table 5. The columns report the theoretical and calculated F values.

Table 2 Degree of swelling (%)

Dose/kGy	PVA concentration/%			
	5	10	15	20
20	1062.2	601.2	296.1	297.6
40	1000.0	719.6	381.7	295.0
60	776.0	653.3	457.9	237.0
80	795.5	672.7	484.0	330.6
100	673.2	536.9	293.4	464.6

Table 3 Tensile strength (MPa)

Dose /kGy	Concentration of PVA blending with PEO			
	10%		20%	
	PVA blending ratio		PVA blending ratio	
	20%	40%	20%	40%
20	0.8	1.5	2.5	3.5
40	0.5	0.4	1.2	2.2
60	0.4	0.6	2.0	2.2
80	0.5	0.1	0.9	1.6
100	0.1	0.1	1.0	1.5

Table 4 Elongation at break (%)

Dose /kGy	Concentration of PVA blending with PEO			
	10%		20%	
	PVA blending ratio		PVA blending ratio	
	20%	40%	20%	40%
20	535	450	1500	600
40	140	125	475	525
60	60	130	425	475
80	75	85	155	250
100	60	55	180	200

Table 5 Analysis of variance

	F theory	F gel	F swelling	F ts	F elongation
PVA/%	4.98	260.34	424.18	39.18	40.40
Dose/kGy	4.43	647.12	7.95	13.77	78.98
Mutual	3.23	50.51	22.51	0.98	6.72

It can be observed by comparing the statistical F-values, all the effects except the mutual effects of F tensile strength (F_{ts}) are very significant, at levels of significance α <0.01.

Figs. 1, 2, 3 and 4 show the main effects of the various factors on the measured chemical and physical properties. The points of Figs. 1, 2, 3 and 4 are the means of the data of each level of the experimented factors of Tables 1, 2, 3 and 4 respectively.

These results show that the gel fractions increase strongly with the radiation dose, whereas the increase in PVA concentration produces a slight negative effect. Because reaction of transient species of water radiolysis to yield of radiation crosslinking is many times higher in aqueous solution than in solid polymer[4], the gel fraction decreases with the increase of concentration of PVA solutions. The degree of swelling is strongly influenced by the PVA concentration. The degree of swelling decreases as the PVA concentration increases. The result is contrary to Ikada's research on PVA hydrogel.[5] The degree of swelling is intimately related to the material strength and absorbency of a hydrogel.[6] A moderate degree of swelling

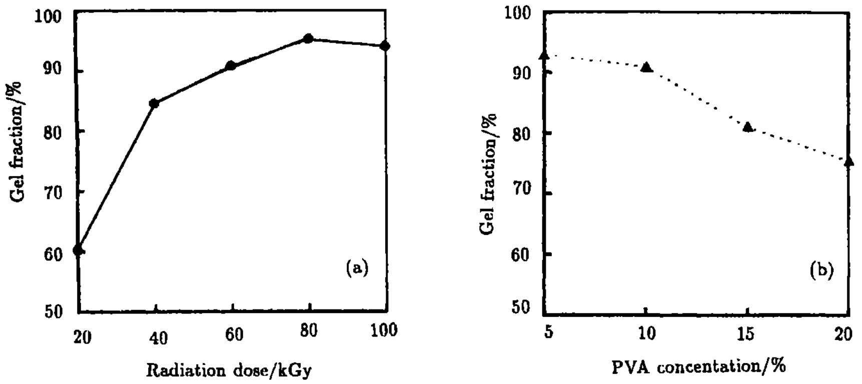

Fig.1 Main effects of radiation dose and PVA concentration on the gel fraction of PVA hydrogel films
(a) Radiation dose/kGy; (b) PVA concentration/%

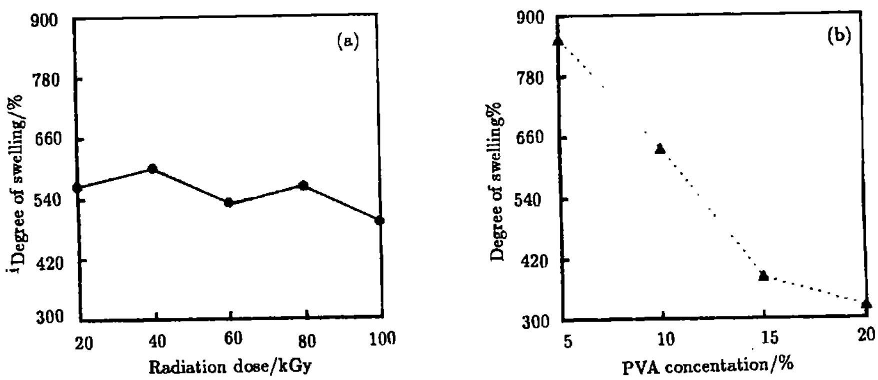

Fig.2 Main effects of radiation dose and PVA concentration on the degree of swelling of the PVA hydrogel films
(a) Radiation dose/kGy; (b) PVA concentration/%

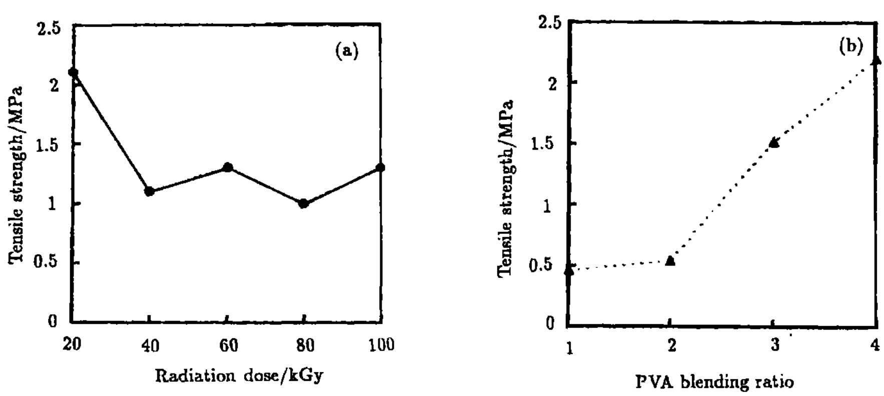

Fig.3 Main effects of radiation dose and PVA blending ratio on the tensile strength of PVA blends hydrogel films
(a) Radiation dose/kGy; (b) PVA blending ratio (1,2,3 and 4 indicate the PVA blending ratio of 20%, 40% in the blends concentration of 10% and 20% respenctively)

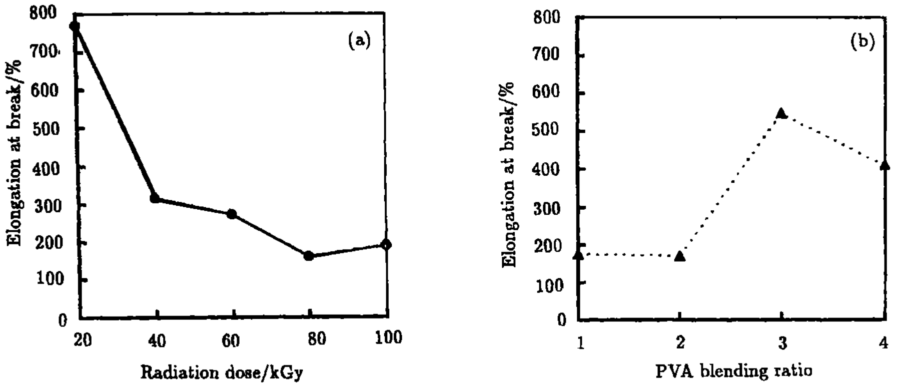

Fig.4 Main effects of radiation dose and PVA blending ratio on elongation at break of PVA blends hydrogel films

(a) Radiation dose/kGy; (b) PVA blending ratio (1,2,3 and 4 indicate the PVA blending ratio of 20%, 40% in the blends concentration of 10% and 20% respenctively)

is very necessary to the improvement of the mechanical strength of hydrogels. The PVA hydrogel films formed by irradiation of its aqueous solutions are somewhat fragile. To improve its mechanical strength, PVA blends with PEO at given concentration and ratio were prepared , the tensile strength of the PVA blend hydrogel films formed by radiation crosslinking is mainly influenced by the PVA blending ratio, the tensile strength shows a marked growth with increase of this variable. These results further indicated that PVA can improve the mechanical strength of the blends hydrogel films, this may be due to its reducing effect on the degree of swelling of the PEO hydrogels.[3,7] The elongation at break decreases significantly with increase of the radiation dose, but it increases significantly as the PVA blending ratio increase.

4 Conclusions

Our present study showed the influences of the experimented factors on the measured chemical and physical properties of the PVA hydrogels. The results suggest that radiation dose increase has little effect on the degree of swelling and tensile strength, but produces a positive effect on the gel fraction and a negative effect on the elongation at break. PVA concentration increase has a negative effect on the gel fraction and the degree of swelling, while PVA blending ratio increase has a positive effect on the tensile strength and elongation at break.

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