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Journal of Macromolecular Science, Part A

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713597274>

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To cite this Article Singh, Harpal and Tyagi, P. K.(1990) 'Note Radiation-Induced Grafting of 2-Hydroxyethyl Methacrylate Onto Silk for Biomedical Applications', *Journal of Macromolecular Science, Part A*, 27: 3, 385 – 389

To link to this Article: DOI: 10.1080/00222339009349562

URL: <http://dx.doi.org/10.1080/00222339009349562>

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NOTE

RADIATION-INDUCED GRAFTING OF 2-HYDROXYETHYL METHACRYLATE ONTO SILK FOR BIOMEDICAL APPLICATIONS

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INTRODUCTION

Silk is a natural protein fiber widely used as biostable suture in surgery. A number of properties of silk have been improved by grafting with vinyl monomers. Since greater hydrophilicity generally leads to better biocompatibility [1, 2], several workers have grafted hydrophilic monomers onto various substrates [3, 4]. This has led us to the work reported here.

EXPERIMENTAL

Medical-grade black braided silk suture (Size 4) obtained from Johnson & Johnson Company Ltd., Bombay, India, and original degummed pure mulberry silk twisted yarn (sericin content 2–3%) collected from the Central Silk Board of India, Bangalore, were used for all the grafting experiments. 2-Hydroxyethyl methacrylate (HEMA) monomer (E. Merck, Germany) was distilled at 70°C at 20 torr and stored at 4°C. Pure methanol (AR grade, BDH India) was used without any purification. Grafting was carried out using ^{60}Co gamma radiation by the procedure for grafting reported previously [5]. The present grafting was calculated by the equation $(W_2 - W_1)/W_1 \times 100$, where W_2 is the weight of the grafted sample and W_1 that of the ungrafted sample.

RESULTS AND DISCUSSION

It is evident from Fig. 1 that the percent grafting in pure water is low compared to the grafting in water-methanol mixtures. A possible reason is that any homopolymer of HEMA is precipitated in water and inhibits the diffusion of monomer to the growing chains as well as into the matrix of the sample.

The degree of grafting increases with increasing methanol content, and maximum grafting is obtained in a 1:1 water-methanol solvent mixture. The methanol has two major effects: 1) homopolymerization is reduced and 2) viscosity build-up in the surrounding solution is retarded. The grafting decreases at methanol levels above 50% and becomes negligible in 100% methanol.

At a constant total dose, grafting was found to be higher at lower dose rates for all the silk samples (Fig. 2). It was reported earlier that a number of (monomeric and polymeric) free radical species are formed during irradiation, and their number increases with increasing dose rate and that the rate of homopolymerization is greater than the rate of grafting at higher dose rates [6].

There is a linear increase in the percent grafting with increasing total dose of irradiation up to 0.302 Mrd, beyond which there is a marked fall in the rate of grafting (Fig. 3). Since the availability of monomer is higher in the initial stages, the monomer can diffuse easily to the grafting sites, but the rate of homopolymer formation is not

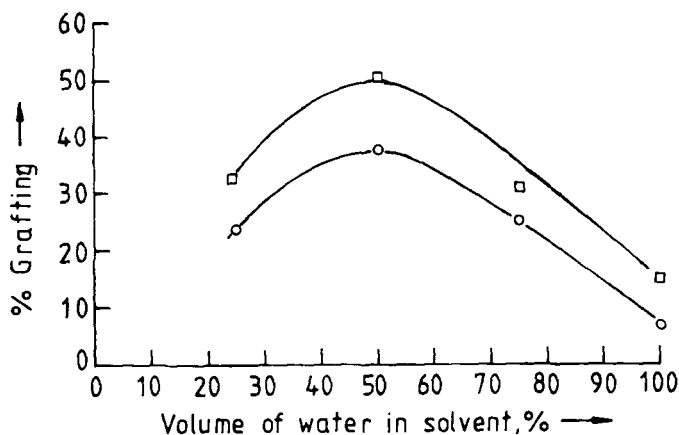


FIG. 1. Effect of solvent composition on percent grafting of silk. Solvent, methanol; monomer concentration, 20 vol%; total dose, 0.302 Mrd; dose rate, 56 rd/s. (□) Pure mulberry silk twisted yarn sample. (○) Medical grade silk suture sample.

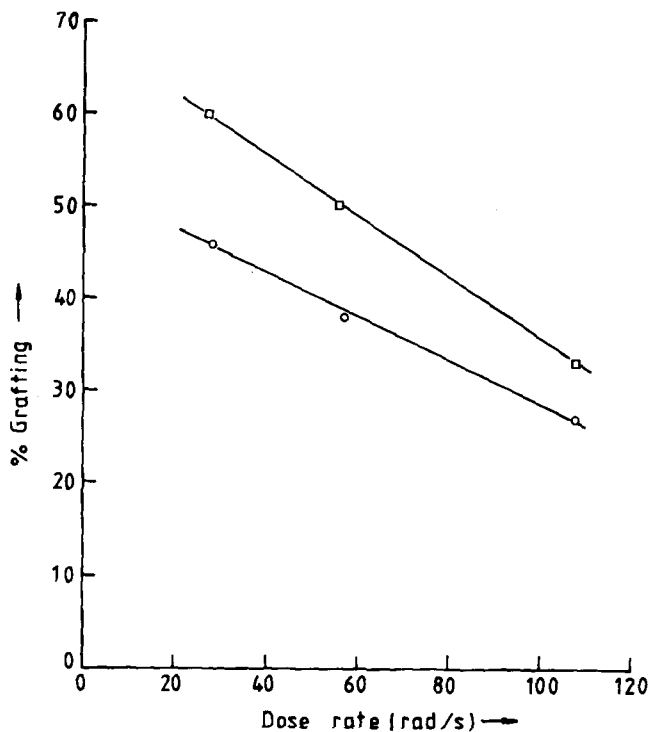


FIG. 2. Effect of dose rate on percent grafting of silk. Solvent, 50:50 methanol:water; monomer concentration, 20 vol%; total dose, 0.302 Mrd. (□) Pure mulberry silk twisted yard sample. (○) Medical grade silk suture sample.

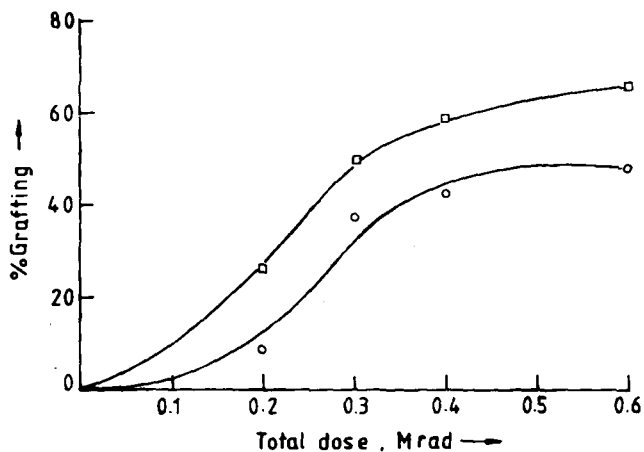


FIG. 3. Effect of total dose on percent grafting of silk. Solvent, 50:50 methanol:water; monomer concentration, 20 vol%; dose rate, 56 rd/s, (□) Pure mulberry silk twisted yarn sample. (○) Medical grade silk suture sample.

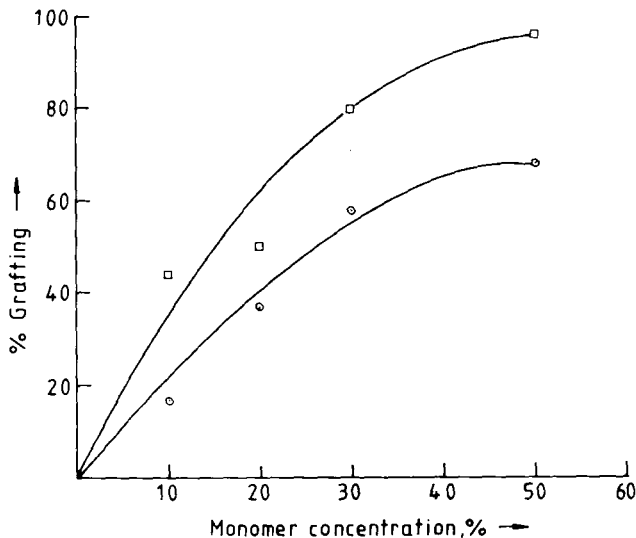


FIG. 4. Effect of monomer concentration on percent grafting of silk. Solvent, 50:50 methanol:water; total dose, 0.302 Mrd, dose rate, 56 rd/s. (□) Pure mulberry silk twisted yarn sample. (○) Medical grade silk suture sample.

much affected. Beyond a total dose of 0.302 Mrd, the slow decrease in grafting indicates that there is an increase in the rate of homopolymerization compared to the rate of grafting. Further, the increasing content of grafted polyHEMA may also act as a barrier to the diffusion of monomer into the polymer matrix.

Figure 4 shows that there is an increase in grafting with increasing monomer concentration, as has also been found in other grafting systems [7].

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Received November 1, 1988

Revision received April 12, 1989