# Swellability of Acrylate Methacrylate–Cellulose Matrix Systems and the Effect on Solute Diffusion Rates

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## INTRODUCTION

The spongy network structure formed when a polymeric material is compacted is referred to as a matrix.[1] A recent study[2] showed that a matrix consisting of an acrylatemethacrylate copolymer and a test solute, salicylic acid (ratio, 1 : 4), displayed a retarded diffusion of the solute. This finding was attributed to poor swellability of the polymer. A study in this laboratory also showed that certain celluloses extracted from locally available agricultural wastes were readily swelled in water. Mixing of these two types of polymers, which have opposite swelling characteristics, may therefore be used to obtain control of solute diffusion rates from their matrices.

## EXPERIMENTAL

To carry out the investigation, an acrylatemethacrylate copolymer (trade name: Eudragit RS100) was obtained from Rhom Pharma, Darmstadt. The polymer has a low content of hydrophilic quaternary ammonium groups and therefore poorly swelled in water.[3] Salicylic acid (reagent grade, BDH) was used as test solute. Cellulose was extracted from certain agricultural wastes (sugar-exhausted sugar cane fibres and maize cob). In the procedure, a sample of the crude in coarse powder (400 g) was added to an acidic mixture (400 mL) consisting of nitric acid (3.5% w/v) and sodium nitrite (0.001% w/v) and heated at 90°C for 3 h to remove soluble nitrolignins. The residue was collected by filteration and then digested (50°C, 2 h) with 4 L alkaline mixture of sodium hydroxide and sodium sulphite, each 2% w/v. After filteration, the residue was bleached successively at 50°C first with sodium hypochloride (0.3% w/v) and later with a solution of hydrogen peroxide (6% w/v). The residue was washed several times with water, dried at 50°C for 3 d, and then pulverised.

Granular coacervates of the copolymer and the solute were formed by dissolution of the polymer (1 g) and the solute (4 g) in ethanol (15 mL), followed by addition of sodium chloride solution (85 mL, 0.1 *M*, aq). The precipitate was washed three times with water, dried, and then pressed through a sieve (aperture size, 710 μm).

Coacervate microstructure was studied by photomicroscopy. A drop of ferric chloride solution (5% w/v) was added to a sample of the coacervate granules thinly spread on a microscope slide. Photo micrographs were taken at ×16 (the magnification that gave a clear resolution).

The coacervates with or without cellulose or maize starch BP were compressed to hard, poorly disintegrating matrices using a single punch machine (Manesty, type F3). Their mean weight was 119 ± 4 mg and hardness 8.7 ± 1.2 kg. The additives (cellulose or starch) did not affect matrix hardness remarkably in the test concentrations 0–20%.

To assess swellability, matrix dimensions (thickness and diameter) were determined before and after equilibration of the matrices in water at 37°C for 3 h. Surface water in wet matrices was blotted out, their wet weight determined, and then they were dried to a constant weight. From the data, the percent increase in matrix volume and the water uptake were calculated and taken as the swellability index.

Solute diffusion from the matrix was determined as follows: A sample was placed in a cylindrical basket (aperture size, 420 μm) and immersed in a leaching fluid (800 mL water at 37°C). The fluid was stirred 100 rev/min$^{-1}$ with a Gallenkamp single-blade stirrer. After 3 h, a sample (5 mL) was withdrawn from the leaching fluid to analyse for content of salicylic acid following a procedure described earlier.[2,4] Results of three replicate experiments were reproducible to ±15% (SD) of the mean.

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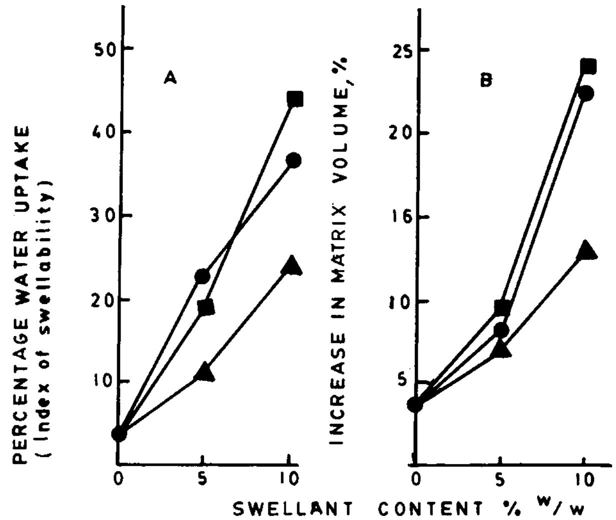


**Figure 1** Effect of the various additives—sugar cane cellulose (■), maize cob cellulose (●), and maize starch (▲)—on matrix swellability as measured by fluid uptake (A) and increase in matrix volume (B).

## RESULTS AND DISCUSSION

The results (Fig. 1) showed that cellulose imparted a high swellability to the matrixes to a greater extent than was achieved with starch. High swelling forces

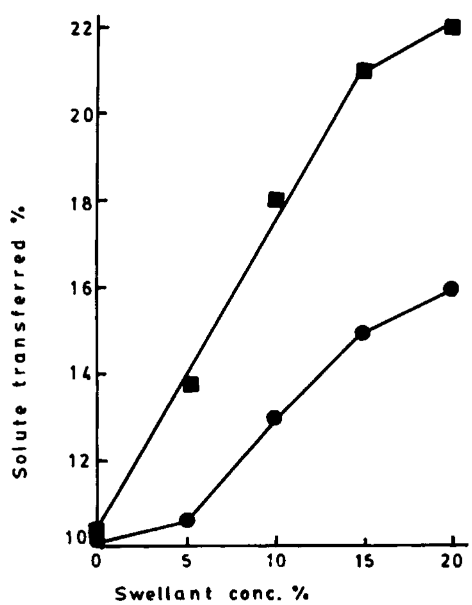


**Figure 2** Effect of swellant sugar cane cellulose (■) or maize starch (●) concentrations on solute diffusion from the matrices. Note that maize cob cellulose and sugar cane cellulose had a similar effect on solute diffusion.

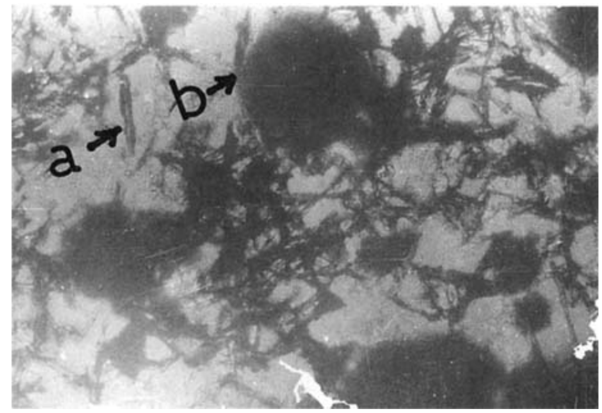


**Figure 3** Photomicrograph of the granular coacervates showing the free (uncoated) salicylic acid crystals (a) and the polymer coated salicylic acid particles (b).

caused rupture of matrices containing cellulose 15% during equilibration in water. The increased swellability led to a corresponding increase in solute diffusion rates (Fig. 2). Photomicrography (Fig. 3) of the granular coacervates (without swellants) revealed the presence of free (uncoated) crystals of the solute (designated a) and polymeric coated particles (designated b). The polymeric particles were stained blue, indicating the presence of salicylic acid.

It is expected that during compression the otherwise free solute crystals will become embedded in a network of the polymeric matrix. Solute diffusion from such a system takes place via aqueous-filled channels. Hence, increase in the matrix porosity obtained by leaching of a water-soluble additive[5] or swelling as obtained in this study is associated with increased matrix diffusivity.

Thus, inclusion of the readily swelled celluloses together with the poorly swelled acrylatemethacrylate copolymer in the matrix composition is an approach for obtaining control of solute diffusion rates from the systems studied.

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