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VISCOSITY AND STABILITY STUDIES OF LIQUID PARAFFIN EMULSIONS PREPARED BY HYDROXYPROPYL METHYLCELLULOSE

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ABSTRACT

Liquid paraffin-water emulsions were prepared by three viscosity grades of hydroxypropyl methylcellulose (HPMC) polymer which showed pseudoplastic behavior. The viscosity of emulsions increased, but the mean droplet diameter of globules decreased on increasing the concentration of the polymer. On storage at 25°C, the viscosity of emulsions stabilized by low viscosity grade polymers increased. For higher viscosity polymer, the emulsion viscosity initially increased and then decreased slightly on aging. Lower viscosity grade HPMC polymers have higher emulsifying efficiency, whereas higher viscosity grade polymer emulsion is much more stable towards centrifugation.

INTRODUCTION

Derivatives of cellulose polymers are used in emulsion preparations [1-3]. The effect of different techniques on the preparation of various emulsions using methylcellulose as emulsifying agent have been reported[4]. Polymers form complex with some surfactants resulting in a synergistic effect on emulsion stability[5]. Interfacial properties of aqueous solutions of methyl and hydroxypropyl methylcellulose were studied by Sarkar[6]. The surface shear viscosities of aqueous HPMC solutions and its relationship with various parameters have been reported[7]. In the present study the effect of polymer concentration on droplet diameter and viscosity of emulsions on storage have been reported. Centrifugation studies were performed to determine the stability of emulsions.

MATERIALS AND METHODS

Hydroxypropyl methylcellulose (Colorcon) of three viscosity grades (E15, E50 and E4M) was used. A 2% solution of these polymers has a viscosity of 15, 50 and 4000 cps respectively. Other materials were of reagent grade.

HPMC polymer solution was prepared in hot distilled water. To it light liquid paraffin (50% w/w) was added and the mixture was homogenized with a Silverson mixer emulsifier for five minutes. Viscosities were measured by a Contraves Rheomet 135 at shear rate 50/s and at 25°C. Samples of emulsion were stored at 25°C and their viscosities were measured periodically and examined visually to check any phase separation. Droplet size analysis was done by a microscope fitted with a calibrated graticule in the eyepiece. A minimum of 500 droplets was measured for each sample. Accelerated stability of each emulsion was measured by centrifuging at 5000 rpm for 30 minutes for four times and the amount of aqueous layer separated was measured. Each experiment was done in triplicate and the average value was recorded.

RESULTS AND DISCUSSION

Emulsions (o/w) were prepared with HPMC polymers and the mean droplet diameter (D_{av}) of each emulsion was measured. In Fig. 1a the D_{av} is plotted as a function of polymer concentration. The droplet diameter of E15 and E50 polymer emulsions decreased with the increase in the concentration of the polymer. But above 1.5% there was less decrease in D_{av}. At higher polymer concentration, the interfacial layer become saturated with the polymer and droplet diameter did not decreased significantly with the increase of the concentration. E4M polymer emulsion showed different droplet size distribution pattern. Up to 0.75% (w/w) of polymer, D_{av} decreased linearly. Above that concentration, the mean droplet diameter increased significantly. This increase in the mean droplet diameter is due to the high viscosity of the E4M polymer which offers greater resistance to dispersion of the oil and prevents in decreasing the globule size during preparation of emulsion.

The lower viscosity grade HPMC (E15 and E50) polymer emulsion had smaller droplet diameter than the higher viscosity grade polymer (E4M) emulsion. This is in agreement with the previous findings[2,3]. Low molecular weight polymers diffuse more rapidly to the interface than the high molecular weight agents, although the later is strongly adsorbed [8]. For this reason lower molecular weight HPMC polymer (E15 and E50) emulsions had smaller droplet size than those of higher molecular weight polymer (E4M) emulsion.

All emulsions showed pseudoplastic behaviour, i.e. the viscosity decreased with increased rate of shear. Fig. 1b shows the viscosity change of emulsions as a function of concentration. The viscosity of emulsions increased with molecular weight and concentration of the polymer. Increasing the concentration of the polymer decreased the mean droplet diameter of emulsion (with the exception of 1% E4M emulsion) as evident from Fig. 1a. This increased viscosity with increased concentration of the polymer is due to decreased droplet diameter. The interfacial layer becomes thicker with increasing molecular weight[9]. So the higher molecular weight polymer (E4M) emulsion showed higher viscosity than the lower molecular weight polymer emulsion.

The emulsions were stored at 25°C and their viscosities were measured periodically. Fig.2a shows the apparent viscosity change of emulsions on aging. There was four to five fold increase in viscosity of E15 and E50 emulsions and the fluid like emulsions were semi-solid after twelve weeks of aging. This increase in viscosity is due to unfolding and increased adsorption of polymer on the oil globules during storage. E4M polymer emulsion also showed initial increase in viscosity. But after three weeks of storage, the viscosity gradually decreased and after twelve weeks of storage,

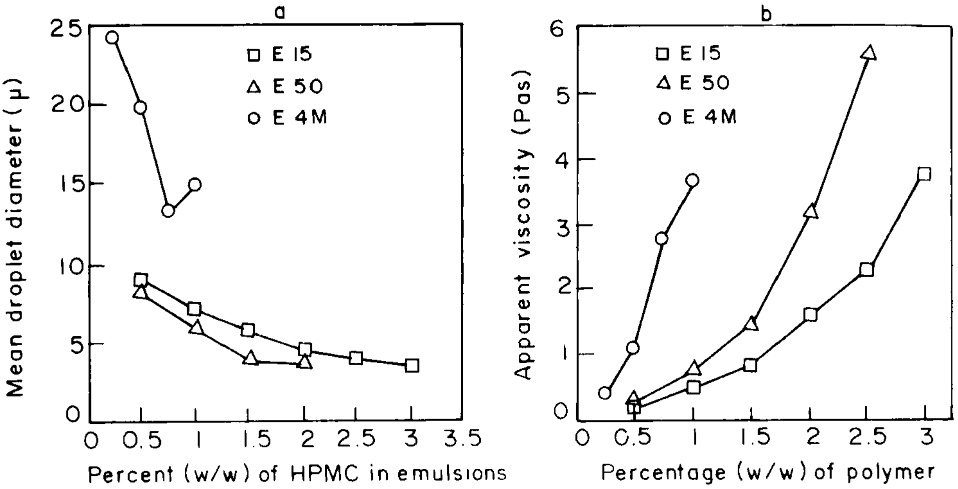

FIGURE 1

(a) Mean droplet diameter (D_{av}) of emulsions plotted against HPMC concentration. (b) Viscosity of emulsions as a function of polymer concentration. Polymers E15 □ E50 △ ; E4M ○ . Shear rate 50/s, temperature 25°C.

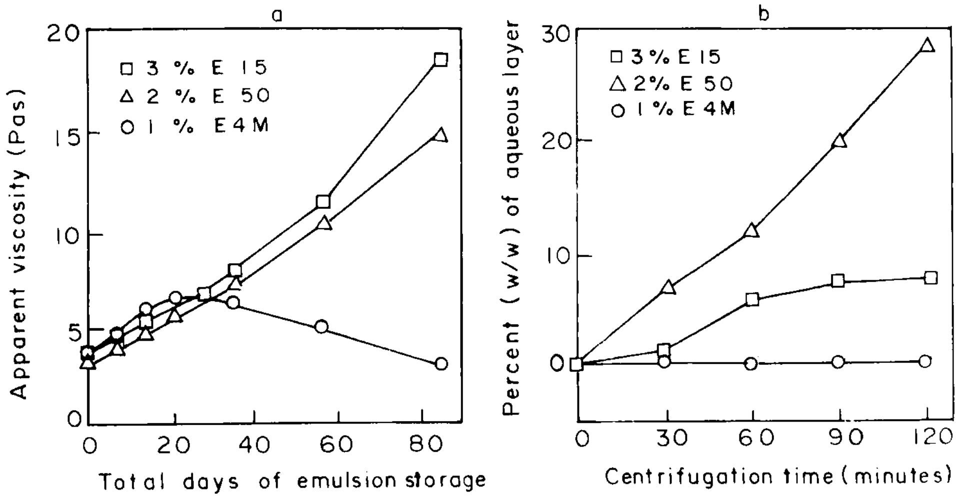

FIGURE 2

(a) Viscosity change of HPMC emulsions on storage at 25°C. (b) Percentage of lower layer separated on centrifugation of emulsions at 5000 rpm (1200 g) for two hours. Polymers : 3% E15 □; 2% E50 △ ; 1% E4M ○ .

the viscosity decreased by 20% of the initial viscosity. On prolonged storage, the long chain high molecular weight polymers (E4M) become dehydrated resulting in the decrease of viscosity.

Accelerated stability tests on emulsions were performed by centrifugation. Fig. 2b shows the percentage (w/w) of aqueous layer separated from emulsions on centrifugation at 5000 rpm (1200 g) for 2 hours. For first 30 minutes there was little phase separation in 3% E15 emulsion. During the next one hour of centrifugation there was gradual separation of lower layer. The last 30 minutes of centrifugation did not increase the percentage of lower layer. In case of 2% E50 emulsion there was gradual increase in aqueous layer, but there was no phase separation in 1% E4M emulsion after two hours of centrifugtion.

The mean droplet diameter of 3% E15 emulsion is less than 2% E50 emulsion (Fig. 1a). Also, the viscosity of 3% E15 emulsion is more than that of 2% E50 emulsion (Fig. 1b). So these two factors combined together make 3% E15 emulsion more stable than 2% E50 emulsion during centrifugation. The droplet diameter of 1% E4M emulsion is larger than the other two polymer emulsions. Still 1% E4M emulsion is more stable due to the high viscosity which prevents phase separation during centrifugation.

The minimum concentration of HPMC required to produce stable emulsion was also determined. This was found to be 3, 2 and 1% for E15, E50 and E4M polymer respectively. At these concentratons and above, the emulsions were stable and showed no visible phase separation for four months under investigation.

CONCLUSIONS

The low viscosity grade HPMC polymers have higher emulsifying efficiency than the high viscosity grade polymer. Low molecular weight polymers diffuse more rapidly to the interface than high molecular weight agents. E4M emulsion is very stable and did not show any phase separation after centrifugation. The viscosity of the E4M emulsion slightly decreased on aging, but the viscosity of E15 and E50 polymer emulsions increased significantly on storage. So it is suggested that a combination of low and high viscosity grades of HPMC polymers would produce better and more stable emulsion which can be used as pharmaceutical ointment base.

REFERENCES

1. G. F. Biehn and M. L. Ernsberger, Ind. Eng. Chem., 40 (8), 1449 (1948).
2. R. E. M. Davies and J. M. Rowson, J. Pharm. Pharmacol 12, 237 (1960).
3. A. A. Kassem, E. N. El-din, A. E. Fouli and Y. E. Hamza, Bull. Fac. Pharm. Cairo University., 10 (9), 199, (1971) (Pub. 1973).
4. Z. Krowczynsky and B. Kluczykowska, Proc. 2nd. Conf. Appl. Phys. Chem., 2, 711 (1971).
5. R. Buscall, Prog. Colloid. Poly, Sci., 63, 15 (1975).
6. N. Sarkar, Polymer, 25, 481 (1984).
7. R. D. Klodt, R. Wuestneck, R. Ruhle, and W. Thuemmler, Abh. Akad, Wiss, DDR, Abt. Math., Naturwiss. Tech., 257 (1986) (Pub. 1987).
8. F. D. Snell and I. Reich, Am. Perfumer Essent. Oil Rev., 58, 171 (1951).
9. J. M. G. Lankveld and J. Lyklema, J. Colloid Inter. Sci., 41(3), 475 (1972).