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Relation between viscosity of carboxymethylcellulose aqueous solutions and charge of its particles in the presence of preservatives

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Summary

The properties of a commercial sodium carboxymethylcellulose, Blanose 7 MD, have been investigated by Rheometrics Mechanical Spectrometer RMS-605 and Zeta Meter. The influences of added preservatives on the viscosities and ζ-potential values, were variable, probably due to specific effect of anions on electrical double layer of sodium carboxymethylcellulose particles.

Introduction

The dispersions of sodium carboxymethylcellulose have been investigated from rheological (Salama, 1981; Primorac et al., 1986) and surface charge (Djurić and Jovanović, 1983, 1984, 1985) points of view. Few authors have considered the relation between the viscosity and ζ-potential (Jovanović and Djurić, 1985; Rambhau et al., 1977; Dawes and Groves, 1978), in spite of the fact that the presence of an electrical double layer exerts a profound influence on the flow behaviour, well known as the electroviscous effect (Hunter, 1981).

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Materials and Methods

Blanose 7 MD was supplied by Hercules; Nipagin A sodium and Nipasol M sodium were supplied by Nipa Laboratories Ltd. All other materials were official grade (Ph. Jug. IV), water being glass distilled.

A Rheometrics Mechanical Spectrometer RMS 605 (Rheometrics Inc. U.S.A.) equipped with parallel plates has been used; a Zeta Meter (Zeta Meter Inc., New York) has been employed for measuring electrophoretic mobility.

The Blanose dispersions from 6 to 10% were prepared by mechanical stirring in distilled water or in 0.2% of aqueous solutions of preservatives. The viscosities were measured 24 h after preparation of the Blanose solutions, and after 45 days of storage at room temperature. All measurements were made in dynamic mode at 16 different frequencies at a temperature of 25 °C. As the solutions were in gel form, they required to be diluted before determination of electrophoretic mobility, and 1 : 100 dilutions used to be well suited to the Zeta Meter with regard to the particle observation and counting. All measurements and calculations were carried out as described in the instrument manual. pH values were taken on a Beckman Zeromatic pH Meter.

Results

Fig. 1 presents the viscosities of Blanose gels depending on frequencies applied and time of storage. Figs. 2–4 show the effect of frequencies on complex dynamic viscosity of Blanose gels prepared with 0.2% of different preservatives. Table 1 shows the effect of preservatives on the pH values and ζ-potentials of Blanose 7 MD gels.

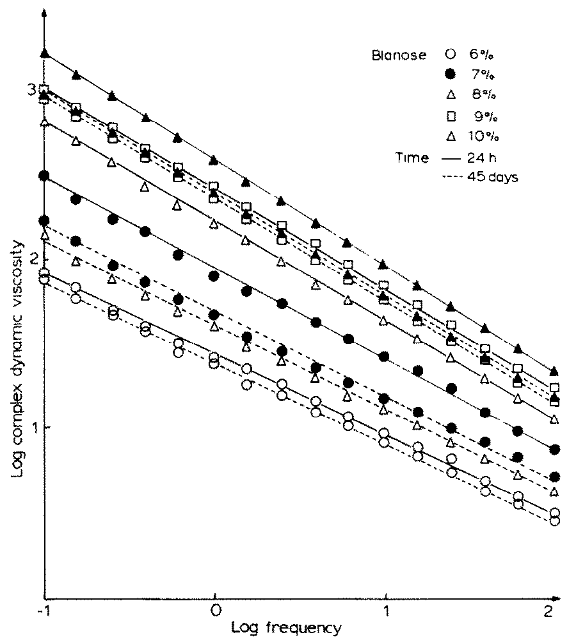

Fig. 2. The viscosities of Blanose gels depending on frequency in the presence of 0.2% of sodium-benzoate, measured after 24 h and after 45 days.

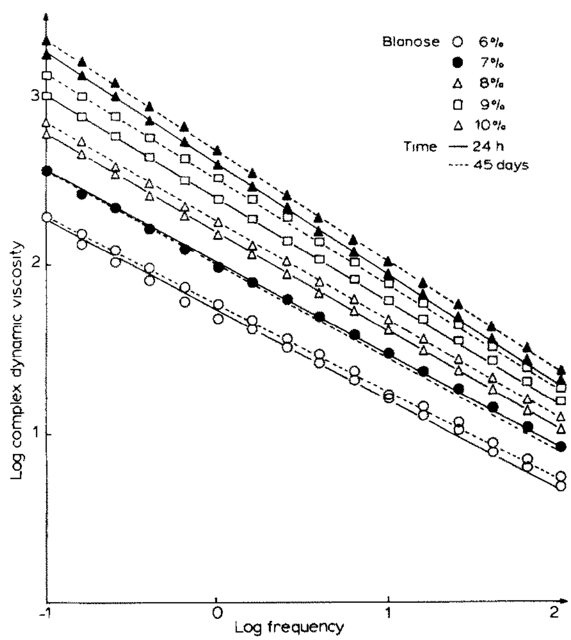

Fig. 1. The viscosities of Blanose gels depending on frequency measured after 24 h and after 45 days.

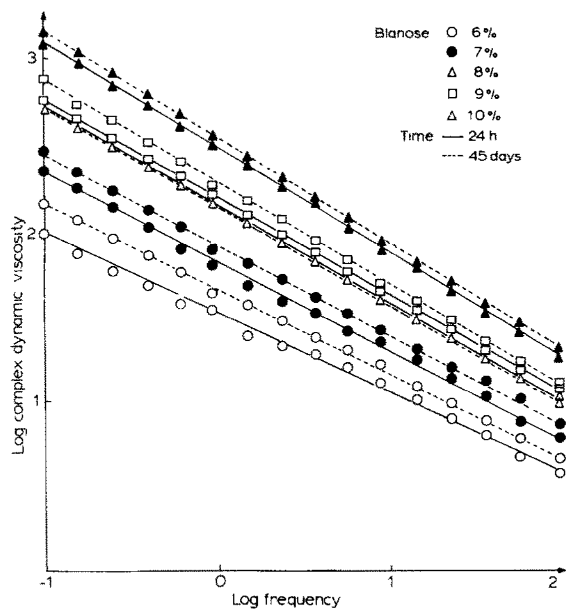

Fig. 3. The viscosities of Blanose gels depending on frequency in the presence of 0.2% of Nipagin A-sodium, measured after 24 h and after 45 days.

TABLE 1

The effect of preservatives on ζ-potential (mV) and pH value of Blanose gel

Sample	Blanose 10%		Blanose 9%		Blanose 8%		Blanose 7%		Blanose 6%	
	a	b	a	b	a	b	a	b	a	b
ζ-Potentials										
I	−56.47	−41.81	−34.69	−39.65	−32.07	−42.09	−32.71	−36.99	−26.94	−28.71
II	−72.92	−41.81	−37.47	−39.48	−40.38	−37.51	−30.53	−31.76	−25.71	−30.31
III	−49.18	−56.84	−41.17	−42.36	−40.78	−40.10	−27.53	−37.19	−25.54	−29.54
IV	−57.80	−55.61	−36.02	−54.60	−46.33	−42.40	−41.53	−36.14	−26.01	−27.81
pH values										
I	6.95	7.69	6.91	7.81	6.51	7.71	7.21	7.72	7.15	7.75
II	9.59	8.39	9.61	8.48	9.60	8.49	9.41	8.29	9.48	8.39
III	9.49	8.41	9.89	8.42	8.62	8.51	8.15	8.51	8.61	8.41
IV	7.44	7.05	7.33	7.12	6.89	7.12	6.93	7.05	6.46	7.02

I, Blanose gel; II, Blanose gel with 0.2% of Nipagin A-sodium; III, Blanose gel with 0.2% of Nipasol M-sodium; IV, Blanose gel with 0.2% of sodium benzoate; a, 24 h after preparation; b, after 45 days of storage.

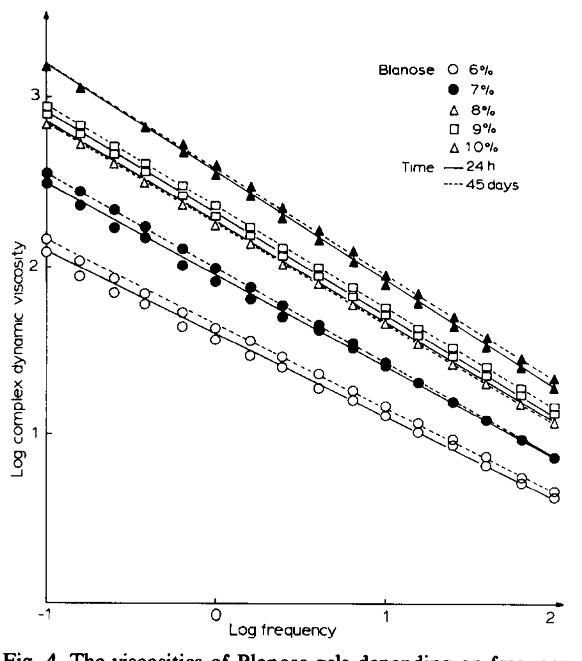

Fig. 4. The viscosities of Blanose gels depending on frequency in the presence of 0.2% of Nipasol M-sodium, measured after 24 h and after 45 days.

Discussion

The values of viscosity of samples without preservatives increased with time and were higher after 45 days, except for 7% of dispersion. Hydration of Blanose molecules after the preparation caused this phenomenon (Fig. 1). Sodium benzoate as preservative caused a slight decrease of the viscosities (Fig. 2). Nipagin A sodium influenced the viscosity of gels after the preparation and during the storage, as well (Fig. 3). But, observed changes are significant only for lower concentrations of preservative and become similar as the concentration of gels is increasing. With 10% of Blanose gel the viscosities in presence of Nipagin A sodium were even lower than with pure Blanose gel of the same concentration.

Comparing Figs. 4 and 1, it can be seen that Nipasol M sodium affects the viscosity of gels more significantly for lower (7%) and higher (10%) concentrations of Blanose, while determined values for medium concentrations, independently of storage, were very close.

Blanose gels without preservatives become less electronegative during storage, regardless of the concentration (Table 1). A very similar situation was observed with samples combined with Nipasol M sodium, the increase of ζ-potential being slighter. The charges of gel particles with Nipagin A sodium decreased, but complex viscosity increased. Sodium benzoate, affecting the decrease of the viscosities during storage, modifies the particle charge variably. As the specific interactions between carboxymethylcellulose sodium and preservatives have not been reported in literature, there is no evidence of association between compounds; it can only be supposed that the different electrolytes employed have specific influence on double electric layers of dispersed particles. This dependence is determined primarily by the anion present, as sodium is associated with each preservative. It was obvious that in all investigated samples viscosity changes were not very significant. This is probably due to the low concentration of preservative, which, as a salt, was not able to compete for the adsorbed water molecule surrounding the hydrated polymer, because of the affinity of the salt ions for water. Ordinarily, as the polymer molecules become dehydrated, their dispersion becomes less viscous. This slight tendency has been observed only with samples containing sodium benzoate. In other samples the rheological variability on aging was determined. The values of ζ-potentials were influenced very much by the presence of preservatives, with high concentrations of gels, the degree of these changes being decreased with decreasing concentrations.

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