SHORT COMMUNICATIONS

Lubrication Enhancement by Surface Gelation of Thin Liquid Films of Methylcellulose and Hydroxypropyl Methylcellulose Solutions

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In a previous paper,[1] we demonstrated that thin liquid films of aqueous dilute polymer solutions between two glass surfaces considerably reduce the static friction coefficients μ_s. Even at a polymer concentration as low as 0.01%, μ_s was about 30% lower than that with a liquid water film. This reduction may be attributed to polymer adsorption onto glass surfaces, which reduces direct contact areas as well as intermolecular repulsive forces between solid surfaces. Moreover, we observed that β-1,4-polysaccharide derivatives (*i.e.*, cellulose derivatives) are more effective in the reduction of μ_s than vinylic polymers. As methylcellulose and hydroxypropyl methylcellulose are used as additives for extrusion moulding of fine ceramics, it is quite important to study the lubricity effect of these polymer solutions in the process. In this paper, we measured the temperature dependence of static friction coefficient μ_s of thin liquid films of aqueous MC and HPMC solutions to see effects of gelation that occurs when these polymer solutions are heated in relevant temperature ranges.[2,3]

The effects of surface gelation on surface activities and surface elastic properties of polymer solutions have been studied by

Sarkar[4] and Kim, Peiffer, and Pincus.[5] However, no studies have been reported on the lubrication properties of surface-geled thin films of polymer solutions. Interestingly enough, we have first discovered that the gelation in thin liquid films of methyl cellulose and hydroxypropyl methylcellulose aqueous solutions between two glass surfaces reduces effectively the static friction coefficient μ_s at the gelation temperature. This discovery is quite important not only in understanding the lubrication behavior for thin liquid films of polymer solutions but also in technological applications for the fine ceramic industry. We report here the experimental results leading to this discovery.

Methylcellulose (MC) and hydroxypropyl methylcellulose (HPMC) used as samples were described in the previous paper.[1] Their molecular characteristics are listed again in Table I. Twice distilled water was used as the solvent.

The apparatus described in the previous paper[1] was used to measure μ_s according to the measuring procedure established. Watch glasses and optically flat glass plates were cleaned as described previously.

Gels of MC were prepared at 70°C for 20 h and gels of HPMC were prepared at 80°C for 20 h in U-shaped tubes as described pre-

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Table I. Molecular characteristics of samples[a]

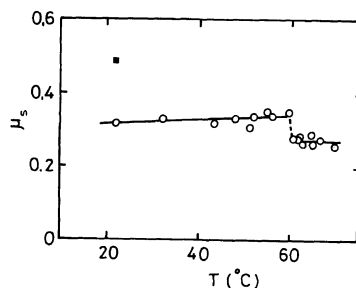| Samples	$M_w/10^4$	DS	MS
MC	5.73	1.76	
HPMC	86.3	1.89	0.27

[a] The abbreviation used are as follows: MC, methylcellulose; HPMC, hydroxypropyl methyl cellulose; DS, degree of substitution made in an anhydroglucose unit; MS, degree of molar substitution of propylene oxide reacted per anhydroglucose unit.
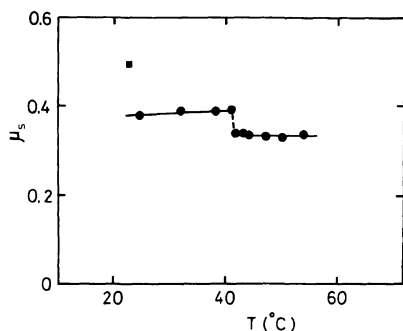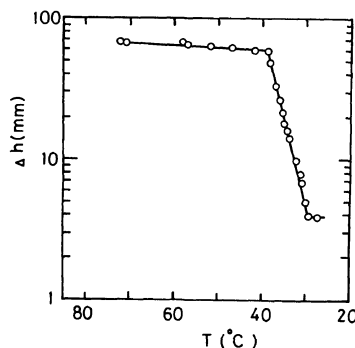

Figure 1. Static friction coefficient μ_s vs. temperature plots for methylcellulose (MC). $C_p = 0.1$ g/100 ml. ■, the μ_s values measured on the thin liquid film of pure water.

Figure 2. Static friction coefficient μ_s vs. T plots for hydroxypropyl methylcellulose (HPMC). $C_p = 0.1$ g/100 ml. ■, the same as in Figure 1.

Figure 3. Gel melting temperature of MC. $C_p = 2.0$ g/100 ml. The values of Δh denote the differences of gel column height prepared at the relevant temperature in U-shape tubes.[2]

Figure 4. Gel melting temperature of HPMC. ●, $C_p = 1.0$ g/100 ml and ○, $C_p = 2.0$ g/100 ml. The implication of Δh is the same as in Figure 3.

viously.[2,6] The U-tubes were then cooled at the rate of 1 K/5 min and the temperature at which pressure head caused flow through a fine capillary was measured as the melting temperature of gels.
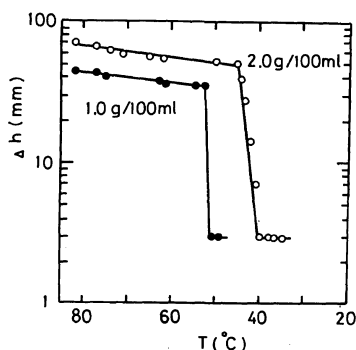
As shown previously,[1] the observed values of μ_s became constant irrespective of load in accordance with Coulomb's friction law, when the load W is more than 120 g. The temperature dependence of static friction coefficient μ_s for thin liquid films of MC and HPMC aqueous solutions at $C_p = 0.1$ g/100 ml is shown in Figures 1 and 2. The friction coefficient μ_s^0 of thin water film is about 0.5, and hence, it is clear from these figures that these polymer solutions are effective in reducing μ_s. With increasing temperature, μ_s only slightly increases first but suddenly drops to lower values at 40°C for MC and at 60°C for HPMC. Figures 3 and 4 show plots of gel column

height Δh vs. temperature for MC and HPMC of the specified concentrations. It is clear that gels are converted to sols by cooling. Although

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the gelation temperatures, that is, sol-gel transition temperatures, were somewhat higher than the gel melting temperatures, the correlation between the transition temperature for μ_s and the sol-gel transition temperature is evident. As described in the previous paper, polymer adsorption onto glass surfaces is responsible for the lubrication action of thin polymer solution films. In general, the polymer concentration is much higher in the adsorbed layer than in the bulk solution.[7] Hence for a bulk polymer concentration of 0.1 g/100 ml, the adsorbed polymer concentration becomes 1 to 2 g/100 ml. Therefore, the correlation between the drop in μ_s and the sol-gel transition temperature may be attributed to the surface gelation of MC and HPMC in the adsorbed layers.

It is concluded that liquid films of hydrophilic polymers are more effective for lubricity in the gel state than in the sol state.

The molecular mechanism of this lubricity effect is now being studied by measuring thickness of liquid film as well as adsorbed amounts of polymers on glass surfaces above gelation temperature and the results will be reported separately.

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