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NOTE

DYNAMIC MODULI CHANGE OF ALUMINUM HYDROXIDE SOL DURING SOL-GEL TRANSITION

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Abstract—Aluminum hydroxide sol was prepared by the sol-gel method. Dynamic moduli such as storage and loss moduli were measured to investigate the relative dominance of elastic and viscous contributions to the viscoelastic response of aluminum hydroxide sol during sol-gel transition. The loss modulus, a measure of viscous response, is larger than the storage modulus, a measure of elastic response, for the sols of low particle concentrations. But at a high particle concentration above 15.3 wt%, the storage modulus is found to be larger than the loss modulus. This inversion from a viscous to an elastic response with the increase of particle concentration is attributable to the formation of gel structures by the aggregation of dispersed particles.

INTRODUCTION

Sol-gel method for making glasses, ceramics, and composites has become an attractive and intensive research area in recent years due to its many advantages over the conventional method [1]. Aluminum hydroxide sol prepared from aluminum alkoxide mainly consists of platelet boehmite particles dispersed in water [2-3]. As it becomes concentrated by the evaporation of water, it shows progressively less fluidity because of the aggregation of primary boehmite particles and is finally transformed into a rigid solid known as "gel". During the sol-gel transition, the concentration of boehmite particles has a profound influence on the rheological properties.

The steady shear flow properties such as viscosity and yield stress have already been reported [1]. However, not much information has been reported on such viscoelastic properties (dynamic moduli) as storage and loss moduli. In this paper, the viscoelastic (dynamic) shear flow properties of the aluminum hydroxide sols are reported.

EXPERIMENTAL

Aluminum hydroxide sols were prepared from aluminum isopropoxide following the procedure of Yoldas [4]. Hydrolysis was carried out with distilled water in excess (100 moles of water per mole of alkoxide) in a round bottom flask with reflux condenser and stirrer. After 1 hr hydrolysis at 90℃ under vigorous stirring, the solution was peptized by adding required amount of HCl and then maintained at 90℃ for 1 hr under stirring. After peptizing, it was transferred to a glass beaker, which was uncovered and placed in an oven at 70℃ until a solid-like wet gel was formed.

Viscoelastic properties were measured with a Rheometrics Dynamic Spectrometer (RDS) [Model RDS-7700, Rheometrics, Inc., U.S.A.]. In the RDS measurements the parallel plate (diameter 35 mm) geometry was used. The storage (G′) and loss (G″) moduli were measured over the frequency range of 0.1-100 Hz. For the determination of particle weight, samples were dried at 70℃ in vacuum for 24-48 hrs until the constant weight was maintained.

RESULTS AND DISCUSSION

The aluminum hydroxide sol is a colloidal system which mainly consists of platelet boehmite particles dispersed in water. As the sol becomes concentrated through the evaporation of water, the particles in it tend to aggregate one another by van der Waals attraction force [3]. On further concentration, the aggregates come into frequent contact and adhesion with

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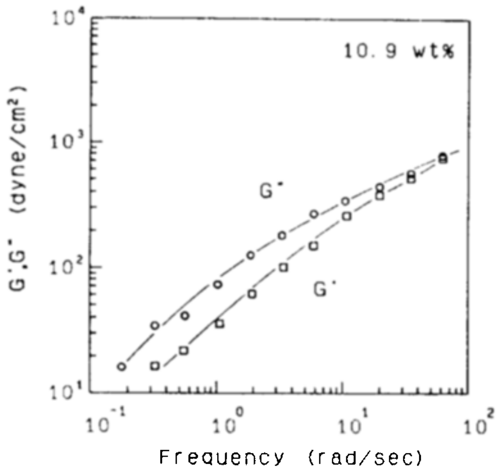

Fig. 1. The storage (G′) and loss (G″) moduli of the solution prepared with 0.10 HCl/alkoxide at particle concentration of 10.9 wt%.

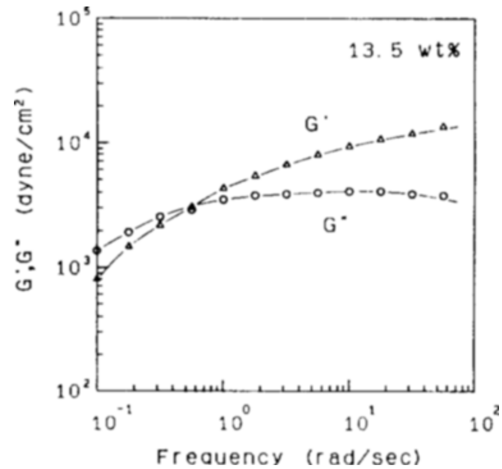

Fig. 2. The relation between storage (G′) and loss (G″) moduli at particle concentration of 13.5 wt%.

one another. The adhesion of aggregates extends out through the whole suspension and can give a solid-like gel structure which exhibits viscoelastic properties in the suspension. The viscoelastic properties of the boehmite particles, covering the range of high concentrations above 10 wt%, have been measured under oscillatory shear with the RDS.

Figs. 1-3 show the storage (G′) and loss (G″) moduli of the solutions, prepared with 0.10 HCl/alkoxide, at several boehmite concentrations plotted as a function of the angular frequency (ω). It is seen in the sol of 10.9 wt% of Fig. 1 that both G′ (ω) and G″ (ω), as the measure of the elastic and viscous response of

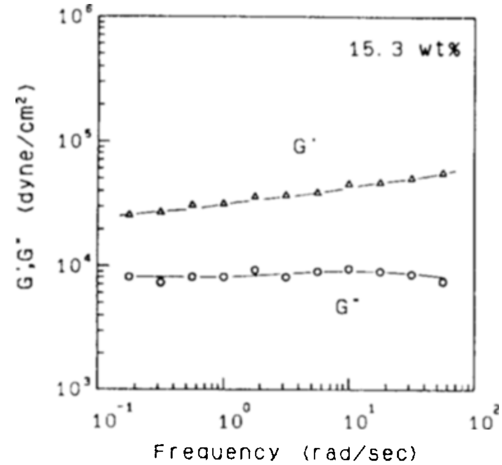

Fig. 3. The relation between storage (G′) and loss (G″) moduli at particle concentration of 15.3 wt%.

the system respectively, increase almost linearly with the increase of the frequency. Over the almostly entire frequency range, it is found that G″ (ω)>G′ (ω) indicating that the viscous response is more significant than the elastic response at this particle concentration.

The effect of the increase of the particle concentration is considered in Fig. 2. For the sol of 13.5 wt%, G′ (ω) crosses over the G″ (ω) at the frequency of around 5×10^{-1} rad/sec, and further increase of particle concentration makes G′ (ω) exceed G″ (ω) as shown in Fig. 3. At the boehmite concentration of 15.3 wt% both G′ (ω) and G″ (ω) moduli remain nearly constant with frequency in Fig. 3, which indicates the feature of a gel-like system [5].

According to the explanation of Sacks [6] for silica systems, the increase in the loss modulus reflects the rapid increase in effective solid loading by the microgel formation, whereas the increase in the storage modulus means an extensive particle-particle interaction and network structure development. The same principle can be applied to the boehmite system: At the low concentration of 10.9 wt% the particle interactions and gel structure formation in the sol are less dominant than the energy dissipation during flow due to the solid loading and hence G″ (ω)>G′ (ω). At the high particle concentration of 15.3 wt% the storage modulus G′ has higher values than the loss modulus G″ because of the development of particle interactions and gel structure.

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