

Effects of Polyvinylpyrrolidone and Carbon Nanotubes on Magnetorheological Properties of Iron-Based Magnetorheological Fluids

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Received 10 June 2005; accepted 22 December 2005

DOI 10.1002/app.24049

Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Magnetorheological (MR) fluids based on glycol, iron powder, polyvinylpyrrolidone (PVP), and carbon nanotubes (CNTs) were prepared. Effects of polyvinylpyrrolidone and carbon nanotubes on sedimentation stability and magnetorheological properties were studied. It is found that the synergetic effects of PVP and CNTs improve the sedimentation stability significantly, and the addition of CNTs reduces the sedimentation velocity and increases the equilibrium sedimentation ratio of the magnetizable particles in MR fluids remarkably. The addition of PVP can reduce the sedimentation velocity of the magnetizable particles, but cannot increase the equilibrium sedimentation ratio and will not change the up trend of apparent viscosity with the increasing intensity of the external magnetic field.

When the PVP content is lower, the increment of original apparent viscosity of the MR fluids at zero-intensity of magnetic field is inconspicuous, and their values of apparent viscosity under magnetic field are similar. However, the apparent viscosity of the MR fluids increases tremendously when the contents of PVP increase to certain degree. The results show that the synergetic effects of PVP and CNTs not only improve the sedimentation stability of the MR fluid but also promote its magnetorheological effect. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 1653–1657, 2006

Key words: magnetorheological; polyvinylpyrrolidone; carbon nanotubes; sedimentation stability; apparent viscosity

INTRODUCTION

Magnetorheological (MR) fluids are substances that exhibit an ability to change their flow characteristics by several orders of magnitude within several milliseconds under the influence of an applied magnetic field. These changes of the rheological properties of the fluids are completely reversible. For these unique properties, MR fluids have wide applications in vibration dampening devices, such as shock absorbers, vibration dampers, force/torque transfer (clutch) devices, and the like, especially in systems in which variable control of the applied dampening/force are adjustable.^{1,2} Therefore, MR fluids have bright future and can be used in fields such as smart structure, automobile, machine, architecture, medical treatment, and so on.^{3–7}

Regular MR fluids contain noncolloidal solid particles with at least five times density of that of the liquid

phase in which they are suspended. Appropriate dispersion of the particles in the liquid phase must be achieved so that the particles do not settle appreciably upon standing nor do they irreversibly coagulate to form aggregates. Without suitable stabilizing or suspending of the solid phase, sedimentation and/or flow induced separation of the solid phase from the liquid phase will occur.^{8,9} Such separation will have a drastic and detrimental influence on the properties of the MR fluid. Thus, the soft magnetic particles are kept in suspension by dispersing a thixotropic agent or thickener in the liquid vehicle.^{10,11} However, the addition of large amount of thixotropic agent or thickener will cause high original viscosity of the MR fluids at zero-intensity of magnetic field.

Magnetorheological fluids based on glycol, iron powder, polyvinylpyrrolidone, and carbon nanotubes (CNTs) were prepared in present work. Effects of polyvinylpyrrolidone and carbon nanotubes on viscosity, sedimentation stability, and magnetorheological performance of such kind of MR fluids will be discussed.

EXPERIMENTAL

The liquid-phase components of the MR fluids were prepared by adding polyvinylpyrrolidone ($M_n = 2500$,

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Contract grant sponsor: Open Fund of Shanghai Key Laboratory of Metal Function Materials Research and Application.

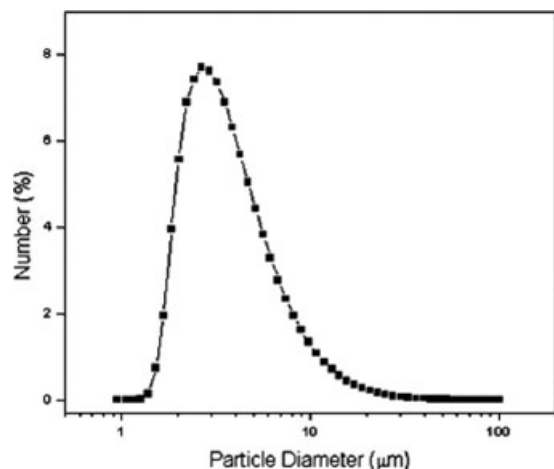


Figure 1 The particle size distribution of the iron powder.

Polyscience) or polyvinylpyrrolidone ($M_n = 30,000$, Shanghai Runjie Chemical Reagent Company) into glycol (analytical grade, Shanghai SSS Chemical Reagent Company) and stirring at 200–400 rpm until the PVP was completely dissolved in glycol. The CNTs with a diameter of 20 nm and a length of 50 μm (Chengdu Organic Chemistry Company) was then added into the liquid components and mixed for an additional 30 min. Following this mixing step, the soft magnetic particles of iron powder (Shanghai Chemical Reagent Company) with a mean diameter of 4.6 μm (as shown in Fig. 1, measured by LS230 laser particle analyzer, Beckman Coulter Company) were slowly and continuously added with agitation for 1 h thereafter. The MR fluid is then subjected to high shear mixing at about 2000–4000 rpm for 15–30 min.

The sedimentation ratio (R) was measured by placing the MR fluid in a vertical cylindrical container at 20°C. R can be calculated by,

$$R = b / (a + b) \times 100\% \quad (1)$$

where a is the length of the clear fluid and b is the length of turbid fluid. The morphology of the sediments of the MR fluids on the glass wall of the container was observed by the reflection mode of optical microscopy (Q600, Leica Co.). The apparent viscosity of the MR fluids versus the intensity of the magnetic field was measured by electromagnetic rheometer modified from NDJ-79 viscosimeter as shown in Figure 2 (Tongji Mechanical and Electrical Company). The equilibrium sedimentation ratio in the present work refers to the value of sedimentation ratio tested after 1 week. The electromagnet was fixed to produce magnetic field, which can be adjusted continuously by the current controller. The original metallic container of the rheometer was replaced by a vitreous container to reduce the influences on the distribution of mag-

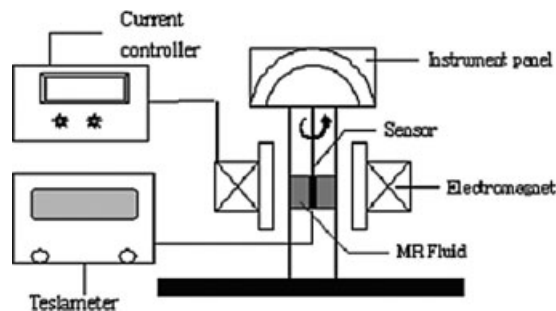


Figure 2 The experimental setup of modified NDJ-79 viscosimeter.

netic field. The apparent viscosity of the MR fluids was measured in shear pattern at the shear rate of 344 s^{-1} . Thus, the apparent viscosity of the MR fluids could be measured under a continuously changed magnetic field. The intensity of the magnetic field was measured by Teslameter (HT100G, Hengtong Magnetolectricity Co.).

RESULTS AND DISCUSSION

Effects of the addition of PVP and CNTs on sedimentation stability of the iron-based MR fluids

The sedimentation stability of the iron-based MR fluids after the addition of PVP and CNTs is shown comparatively in Figure 3. It can be found that the addition of either PVP or CNTs can also reduce the sedimentation velocity. The higher the M_n of PVP is, the lower is the sedimentation velocity. The synergetic effects of PVP and CNTs improve the sedimentation stability significantly. It can be found in Figure 4 that

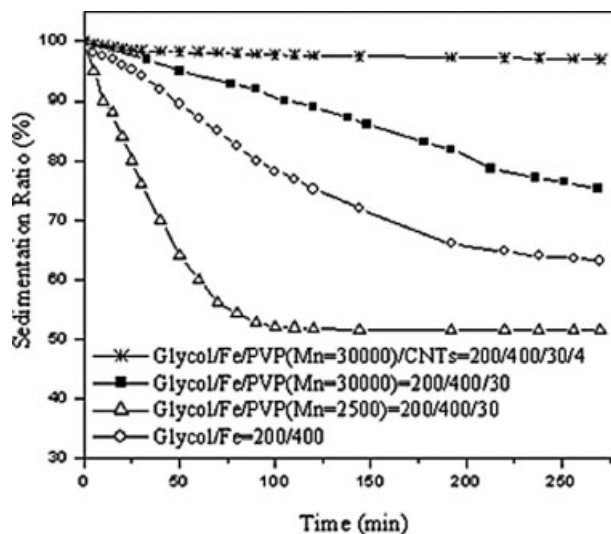


Figure 3 Effects of the addition of PVP and CNTs on sedimentation stability of the iron-based MR fluids.

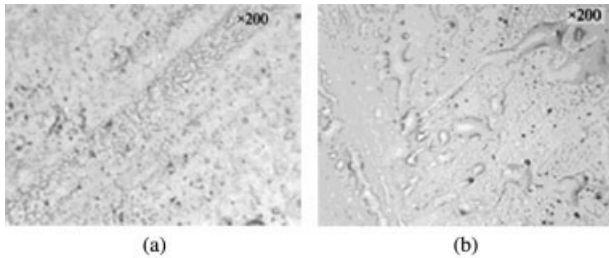


Figure 4 Morphology of the sediments of the MR fluids on the glass wall of the container, (a) without CNTs (Glycol/Fe/PVP ($M_n = 30,000$) = 200/400/30), (b) with CNTs (Glycol/Fe/PVP ($M_n = 30,000$)/CNTs = 200/400/30/4).

the wetting property of the fluid is changed and the structure of the sediment is incompact due to the existence of the liquid bubbles in it after the addition of CNTs in the MR fluids (glycol/Fe/PVP ($M_n = 30,000$)/CNTs = 200/400/30/4). The reason of the occurrence of the liquid bubbles after the addition of CNTs is not clear yet. One possible explanation is that PVP and CNTs in glycol form vesicles or giant vesicles, which are favorable for the higher sedimentation stability of the MR fluids.

Effects of PVP contents and CNTs contents on sedimentation kinetics and equilibrium sedimentation ratio of the iron-based MR fluids

In Figure 5, all the samples have an identical mass ratio of glycol/iron/PVP ($M_n = 30,000$)/CNTs = 200/400/30/ x , and the mass percentage of CNTs here refers to mass ratio of CNTs to the total weight of the system. The addition of CNTs not only reduces the sedimentation velocity but also increases the equilibrium sedimentation ratio of the magnetizable particles

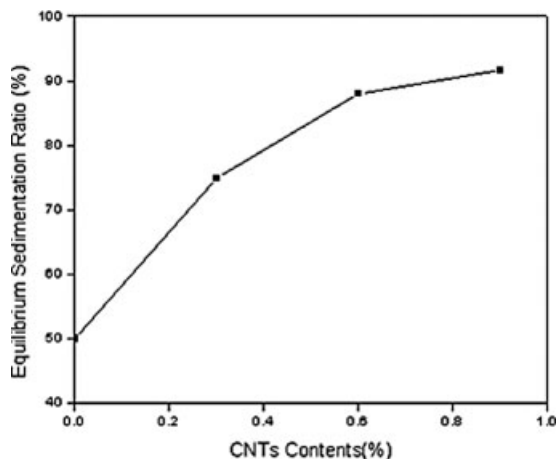


Figure 5 Effects of CNTs contents on the equilibrium sedimentation ratio of the iron-based MR fluids, the composition of the samples is Glycol/Fe/PVP ($M_n = 30,000$)/CNTs = 200/400/30/ x .

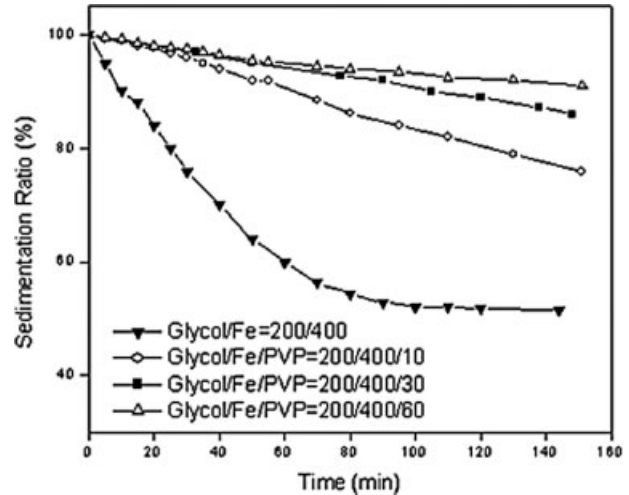


Figure 6 Effects of PVP contents on sedimentation kinetics and sedimentation ratio of the iron-based MR fluids (Glycol/Fe/PVP($M_n = 30,000$) = 200/400/ x).

in the MR fluid remarkably as shown in Figure 5. The equilibrium sedimentation ratio increases with increasing CNTs contents. It is mainly profited from the special character of CNTs. CNTs are rigid and have large length-to-diameter ratio in shape. The addition of CNTs in MR fluids will bring supporting effect to the magnetizable particles which can reduce the sedimentation velocity and increase the equilibrium sedimentation ratio of the MR fluids.

Figure 6 shows effects of PVP contents on sedimentation stability of the iron-based MR fluids. It is found that the addition of PVP can reduce the sedimentation velocity of the magnetizable particles in MR fluids, but cannot increase the equilibrium sedimentation ratio as shown in Table I. The sedimentation velocity decreases with increasing PVP contents. This is mainly attributed to the increment of the viscosity of the carrier fluid. According to Stokes equation (eq. (2)), higher viscosity of the carrier fluid will cause lower sedimentation velocity of the MR fluids.¹²

$$V_0 = 2(\rho - \sigma)g r^2 / 9\eta \quad (2)$$

In eq. (2) V_0 is sedimentation velocity of the particles in carrier fluid; r and ρ are the radius and apparent

TABLE I
Effects of PVP ($M_n = 30,000$) Contents on the Equilibrium Sedimentation Ratio of the MR Fluids

Samples	Equilibrium sedimentation ratio (%)
Glycol/Fe = 200/400	48.4
Glycol/Fe/PVP = 200/400/10	48.2
Glycol/Fe/PVP = 200/400/30	48.2
Glycol/Fe/PVP = 200/400/60	48

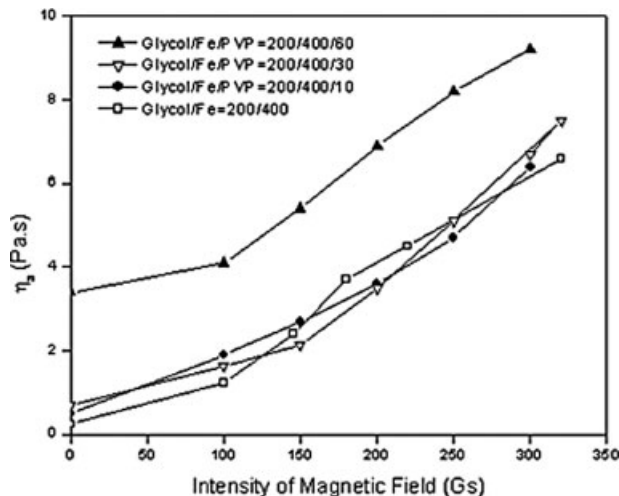


Figure 7 Effects of PVP contents on magnetorheological properties of the iron-based MR fluids (Glycol/Fe/PVP($M_n = 30,000$) = 200/400/ x).

density of the particles respectively; σ and η are the density and viscosity of the carrier fluid respectively, and g is the gravitational constant.

Effects of PVP contents and CNTs contents on magnetorheological properties of the iron-based MR fluids

The apparent viscosity of the MR fluids was measured by the electromagnetic rheometer shown in Figure 2. Figure 7 shows the effects of PVP contents on the magnetorheological properties of the MR fluids. It is found that the apparent viscosity of the MR fluids increases with increasing intensity of the external magnetic field rapidly. However, the addition of PVP will not change the up trend of the apparent viscosity of MR fluids with increasing intensity of the external magnetic field. The addition of PVP in the MR fluids increases the original apparent viscosity of MR fluids at zero-intensity of magnetic field. Furthermore, when the PVP content is lower, the increment of original apparent viscosity of the MR fluids at zero-intensity of magnetic field is inconspicuous, and their values of apparent viscosity under magnetic field are similar. The main reason is that the macromolecular chains just intertwist with each other slightly when the content of PVP is low. However, when the contents of PVP increase to certain degree, the molecular chains intertwist seriously, which results in tremendous increment of apparent viscosity (glycol/Fe/PVP ($M_n = 30,000$) = 200/400/60).

The effects of CNTs contents on magnetorheological properties of the iron-based MR fluids are shown in Figure 8. It can be found in Figure 8 that the apparent viscosity of the MR fluids increases with increasing CNTs contents. However, the increase of apparent

viscosity is more obvious as the magnetic field becomes stronger. The reason is that the magnetizable particles under the magnetic field will be connected as a cluster of chains paralleled to the magnetic field and the CNTs are fixed in different chains. Thus, the magnetorheological effect is promoted by the addition of CNTs.

CONCLUSIONS

MR fluids based on glycol, iron powder, polyvinylpyrrolidone, and carbon nanotubes were prepared. It is found that the synergetic effects of PVP and CNTs improve the sedimentation stability significantly. The addition of CNTs reduces the sedimentation velocity and also increases the equilibrium sedimentation ratio of the magnetizable particles in the MR fluids remarkably. The equilibrium sedimentation ratio increases with increasing CNTs contents. The addition of PVP can decrease the sedimentation velocity of magnetizable particles in the MR fluids, but cannot increase the equilibrium sedimentation ratio. The sedimentation velocity decreases with increasing PVP contents. The addition of PVP will not change the up trend of apparent viscosity of MR fluids with the increasing intensity of the external magnetic field. When the PVP content is lower, the increment of original apparent viscosity of the MR fluids at zero-intensity of magnetic field is inconspicuous, and their values of apparent viscosity under magnetic field are similar. However, the apparent viscosity of the MR fluids increases tremendously when the contents of PVP increase to certain degree. It is also found that the apparent viscosity of the MR fluids increases with increasing CNT contents. The increment of apparent viscosity is more obvious as the magnetic field becomes stronger.

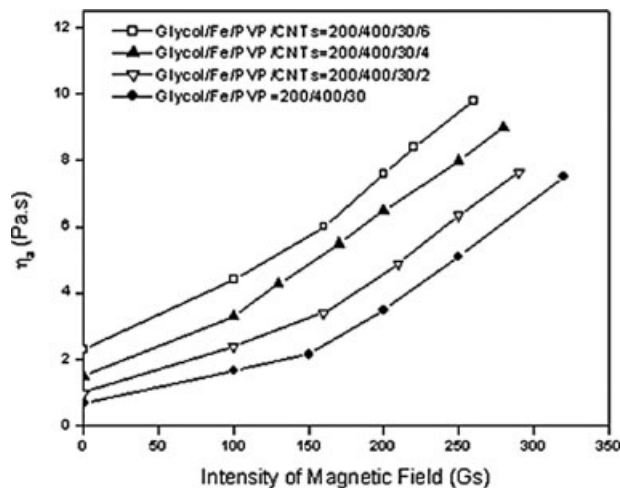


Figure 8 Effects of CNTs contents on magnetorheological properties of the iron-based MR fluids (Glycol/Fe/PVP($M_n = 30,000$)/CNTs = 200/400/30/ x).

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