Digest Journal of Nanomaterials and Biostructures Vol. 8, No. 4, October – December 2013, p. 1699 - 1710

FABRICATION AND PROPERTIES OF PAPER COATINGS WITH THE INCORPORATION OF NANOPARTICLE PIGMENTS: RHEOLOGICAL BEHAVIOR

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In the present work, TiO_2 nanoparticles and $CaCO_3$ nanoparticles were employed as additive pigments to modify the properties of conventional paper coatings. A series of paper coating formulations were designed and fabricated, with a focus on the effect of nanoparticle pigments on the rheological behavior of paper coatings. Furthermore, the role of corresponding factors involving solid content, binder, co-binder and dispersant addition levels was also systematically studied. Steady shear rheological measurements revealed that the as-obtained coating samples exhibited an obvious shear-thinning behavior over a range of shear rates from 0 to 500 s^{-1}. At a given shear rate lower than 10 s^{-1}, the coating viscosity had a tendency to go on rising as the addition level of nanoparticle pigments increased. Solid content, starch latex and hydroxyethyl cellulose addition levels were found to exert a significant influence on the coating viscosity. Herschel-Buckley model was employed to fit the experimental rheological data of various paper coating formulations. Dynamic oscillatory rheological measurements gave a clear indication that the storage modulus was much higher than the loss modulus, indicating the solid-like elastic behavior of the obtained coatings. Moreover, it was apparent that the paper coatings underwent a marked increase in viscoelasticity with the increased nanoparticle pigments.

(Received September 3, 2013; Accepted November 19, 2013)

Keywords: Nanoparticle pigments; Pigmented coatings; Rheological behavior; Model fitting; Dynamic viscoelasticity

1. Introduction

Pigmented coatings have been used extensively to improve surface properties in a great variety of industrial processes such as paints [1, 2] and nanocomposite films [3, 4]. Paper and paperboard as the most widely used mediums of cultural transmission, are typically pigment coated with the goal of improving their optical properties and surface properties [5, 6]. Correspondingly, some other performances, such as ink absorption, surface strength, opacity, brightness and gloss are all improved through a desirable spatial arrangement of pigment coating components in the coating layer. Paper coating is a fairly well mixed colloidal suspension of mineral pigments, binder and thickener as well as additives for control of pH, brightness and pigment dispersion, among which the most important component is the pigments [7]. Pigments are usually derived from various minerals, such as kaolin clay, calcium carbonate and titanium dioxide

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pigment, and these pigments may be used singly or in combination. Recently, great efforts have been made to better understand the impacts of pigment blend on the properties of paper coatings [8, 9]. Al-Turaif [10] displayed that the properties of paper coatings were affected by pigment shape and size in the different extents. It was found that pigment blend with coarse precipitated $CaCO_3$ particles may cause lower gloss and higher roughness of coated paper than pigment blend with fine precipitated $CaCO_3$ particles, and the coating gloss was strongly associated with pigment size, while the coating roughness was largely controlled by substrate absorbency. Also, Lee et al. [11] reported that the particle size in pigment mixture played an important role in the optical properties and printing qualities of coated paper. Owing to the advantages of small size particles in paper coatings, the incorporation of nanoparticle pigments in paper coatings has received great attention.

Nanoparticle pigments with the size range from 1 to 100 nm [12] have been widely used in the manufacture of plastics [13], rubber [14], oil paint [15], ink [16], and coatings [17] due to their unique physicochemical properties such as morphology, crystallite structure, particle size, surface area, porosity and thermal stability. Actually, in recent years, an increasing effort has been devoted to the application of nanoparticle pigments in paper coatings. The influence of common precipitated $CaCO_3$, ZnS (80 nm) and Al-Mg-silicate (30 nm) on the light scattering of coated paper has been investigated by Juuti et al. [18]. Gong's study confirmed that two nano-sized pigments, such as clay and ground $CaCO_3$ can improve the optical and print performance of coated paper significantly [19]. Ghule et al. [20] reported that the paper coatings with ZnO nanoparticles without the aid of binder exhibited an antibacterial activity against the bacterium Escherichia coli. In a recent work, Prasad et al. [21] investigated the functional behavior of paper coated with zinc oxide-soluble starch nanocomposites, and found that the brightness, smoothness, print density, print uniformity, picking velocity and oil absorbency of the paper underwent a significant improvement compared to the bulk-ZnO coated paper. In our previous studies [22, 23], surface modification of nanosized $CaCO_3$ with silane coupling agent and palmitic acid was carried out, the microstructure and interfacial properties of the as-obtained samples were characterized in detail, and their application effect on the coated paper was also preliminarily discussed.

Paper coating rheological behavior is generally known to be quite complicated. The role of the rheological behavior on the structure and surface properties of coated paper has long been emphasized by the paper coating industry. Initially, the rheological behavior of paper coatings is a key factor for runnability and water retention, determining their potential application in coated paper. Furthermore, rheological behavior measurement is an effective approach to characterize the interactions between different components of paper coatings. Moreover, it also acts as a quality control method of paper coatings. As a consequence, it is very essential to acquire various kinds of information from rheological measurements in an effort to improve the design of coating formulations [24].

However, there are limited numbers of studies regarding the rheological behavior of paper coatings in the presence of nanoparticle pigments in the previous literature. In the present work, we focused on the influence of TiO_2 nanoparticles and $CaCO_3$ nanoparticles on the steady shear rheological behavior and the dynamic oscillatory rheological behavior of paper coatings, and the role of several major factors involving solid content, binder, co-binder and dispersant addition levels was also stressed. Moreover, the classical rheological model was employed to fit the experimental rheological data of various coating formulations. The work would be helpful to understand the function mechanism of the nanoparticle pigments in paper coatings and offer guidance for the improvement of coated paper properties.

2. Experimental

2.1 Materials

The main components of the paper coatings studied in this work were: the refined kaolin with high brightness and desired viscosity purchased from China International Medicine Co. Ltd.,

$CaCO_3$ nanoparticles and TiO_2 nanoparticles supplied by Asia Pulp & Paper Co. Ltd. A commercially available modified starch latex replacing styrene-butadiene latex was used here as the main binder in paper coatings. Hydroxyethyl cellulose (HEC), one of the most important cellulose derivatives, was obtained from Aladdin and acted as a very versatile co-binder in this work. In addition, sodium hexametaphosphate (SHMP), tert-butyl alcohol (TBA) and sodium hydroxide (NaOH) serving as dispersant, foam control agent and pH control agent, respectively, were appropriately added into paper coatings. The various components of model paper coatings were specifically illustrated in Table 1.

Table 1 Model paper coating formulations

Raw material name	***Purpose of addition***	***Addition level***
Kaolin	Main pigment	80 wt%-96 wt%
$CaCO_3$ nanoparticle	Additive pigment	2 wt%-10 wt%
TiO_2 nanoparticle	Additive pigment	2 wt%-10 wt%
Modified starch latex	Binder	10-18 pph of pigment
HEC	Co-binder	0.2-1.0 pph of pigment
SHMP	Dispersant	0.2-1.0 pph of pigment
TBA	Foam control agent	0.2 pph of pigment
NaOH	pH control agent	About 0.5 pph of pigment

2.2 Preparation of paper coatings

In a model process of coating preparation, the required pigments were initially dispersed in water using a laboratory high-speed disperser (GFJ-0.4, Shanghai Xiandai Environmental Engineering Technique Co. Ltd., China). In order to achieve the best possible coating quality, corresponding chemicals, namely dispersing agent, pH control agent and certain other additives, were added into the suspensions to improve the dispersion stability of paper coatings. More specifically, various pigments were firstly dispersed in water in the presence of SHMP to form pigment suspensions. The well-dispersed suspensions were then mixed with TBA, starch latex, HEC as well as other components at high shear stress to achieve the targeted paper coatings. The pH of paper coatings was adjusted to about 8.5 with NaOH. These additives such as TBA and NaOH were normally diluted or dispersed, depending on the material. Finally, the obtained paper coatings were screened using a 100-mesh screen for further analysis. Various paper coating formulations are listed in Table 1. For the paper coating formulations, the same procedure was carried out, except that the addition levels of each component were varied.

2.3 Rheological behavior measurement

The steady shear rheological measurements of various coating samples were measured by using a Physica MCR301 advanced cylinder rotary rheometer (Anton Paar, Austria). The dynamic oscillatory rheological measurements were performed on a commercially available AR 550 rheometer (TA Instruments, America). All rheological measurements in the laboratory were done at 25 °C.

3. Results and discussion

3.1 Effect of solid content on the steady shear rheological behavior of paper coatings

Various coating formulations were made at a solid content of 40 wt%, 45 wt%, 50 wt%, 55 wt% and 60 wt%, and the rheological curves are presented in Fig. 1(a) and 1(b). As can be seen, at low shear rate range, these coatings have a non-linear flow curve and exhibit shear-thinning behavior. Meanwhile, an apparent extrapolated yield stress can be found for these coatings, indicating that these suspensions behave like Bingham pseudoplastic fluid. The viscosity of the paper coatings versus shear rate is shown in Fig. 1(b). It can be observed that the viscosity appears to undergo a gradual decrease as the shear rate increases from 0 to 50 s^{-1}, which has been previously observed in similar paper coatings [25]. This shear-thinning behavior can be explained by virtue of the fact that a better orientation of the pigment particles can be achieved with the increase of shear rate, thus leading to a reduction of the viscosity of the suspensions [26]. With further increasing the shear rate, however, these coatings maintain a constant viscosity regardless of the applied shear rate, evidencing that an almost Newtonian behavior occurs at a high shear rate range. It has been generally recognized that the state of Newtonian behavior is mainly due to the presence of thixotropic agents in the suspension and the disagglomeration of clusters with increasing shear stress [27, 28].

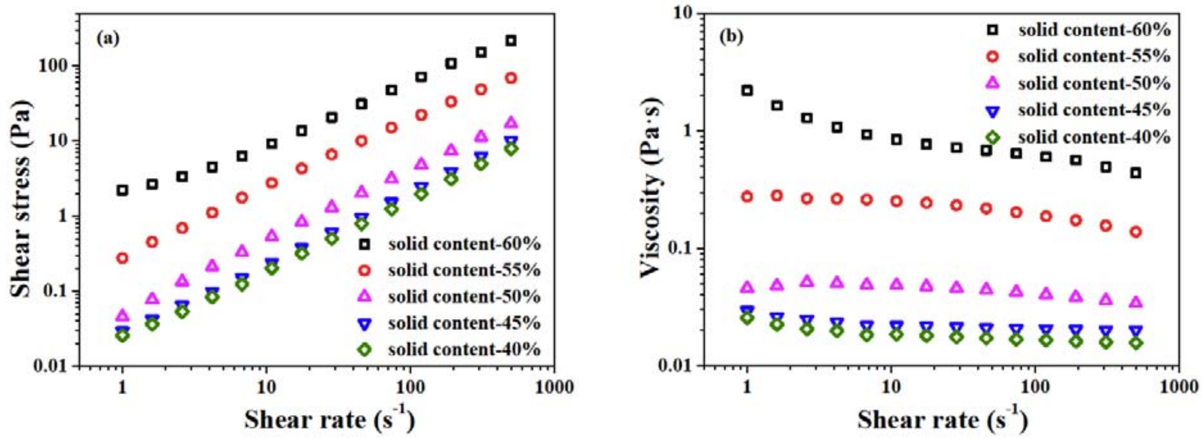

Fig. 1. (a) stress and (b) viscosity as a function of the shear rate for the paper coatings, with various solid content.（Paper coating formulation: kaolin level of 96 wt%, TiO_2 level of 2 wt%, $CaCO_3$ level of 2 wt%. Based on the total weight of pigment, starch level of 14 pph, HEC level of 0.4 pph, SHMP level of 0.6 pph.）

Fig. 1(b) further presents that the viscosity of the above coatings shows a strong dependence on the solid content. At a given shear rate, the coating viscosity shows an upward trend as the solid content increases. Obviously, paper coating sample with 60 wt% solid content possesses the highest viscosity, and appears to be most sensitive to shear rate. This result may be largely attributed to the decrease of water content in coatings. For lower solid content coatings, the particles have a random arrangement due to Brownian motion and would exhibit near Newtonian flow behavior with a low shear viscosity. For higher solid content coatings, inter-particle interactions play an important role in the rheological properties of paper coatings, and the volume packing fraction of paper coatings can reach the maximum value. In such coatings, particles of different sizes are present and the smaller particles are interposed between larger particles, leading to a steady rise in the inter-particle impact, and thus resulting in a considerable increase in viscosity [28].

Various models have been well-known to be employed to reflect the rheological behavior of paper coatings, including Bingham plastic model, Herschel-Bulkley (HB) model, Carreau model and many others. Actually, HB model, commonly considered as a refinement of the classical Bingham model of visco-plastic behavior, can offer a good fit to the shear-rate dependence of plastic viscosity [29]. Hence, HB model was employed here to fit the rheological data of various paper coating formulations, as demonstrated in Eq.(1) [22]:

$$\tau = \tau_y + k\gamma^n \quad (1)$$

In Eq. (1), τ and τ_y refer to the shear stress and yield stress, respectively, the latter of which is a critical value of shear stress, below which a plastic material behaves like a solid. k and n represent the fluid consistency and fluid behavior index, respectively, and γ signifies the applied shear rate. The fitted rheological parameters of various coating formulations are listed in Table 2.

Table 2 Rheological parameters of the HB model fitted to the rheological curves of the paper coating formulations with various solid content.

Solid content (wt%)	***τ_y (Pa)***	***k (Pa s ^{n})***	***n***
40	0.0092	0.0189	0.9704
45	0.0141	0.0210	0.9929
50	0.0527	0.0730	0.8776
55	0.6767	0.5688	0.7748
60	1.2593	1.7471	0.7794

Table 2 reveals that, similar to the viscosity, the yield stress is also defined by the solid content of paper coatings, which is in good agreement with the results in Fig 1(a). The paper coating samples with a solid content of 40 wt%, 45 wt %, 50 wt%, 55 wt%, 60 wt% possess a yield stress at 0.0092 Pa, 0.0141 Pa, 0.0527 Pa, 0.6767 Pa, 1.2593 Pa, respectively. Such high discrepancy might be explained that increasing the solid content of paper coatings reinforces the interaction between particles and the inner structure of the suspensions, in turn resulting in a considerable increase in yield stress, thus making the coatings difficult to flow [30]. The k and n values are commonly employed to predict the pseudoplastic behavior, in particular in pigmented coatings [26, 31]. In Table 2, evidently, k value tends to go on rising with the increased solid content, implying the visible rise of coating consistency. Furthermore, the obtained paper coatings can be found to show a flow behavior index less than 1.0, providing direct evidence that these coating suspensions exhibit pseudoplastic (shear-thinning) behavior.

3.2 Effect of HEC on the steady shear rheological behavior of paper coatings

Fig. 2(a) gives the steady shear stress curves representing the paper coatings with different level of HEC.

Correspondingly, the rheological parameters derived from HB model are presented in Table 3. As shown in Table 3, *n* value signifies that all the coatings display pseudoplastic behavior, exhibiting a significant decline in viscosity with increasing the shear rate from 1 to 500 s^{-1}. The predicted result with HB model is virtually supported by the shear viscosity curves, as indicated in Fig. 2(b). For the paper coating sample with 0.6 pph of HEC, when the shear rate increases from 1 to 500 s^{-1}, the viscosity decreases from 0.78 to 0.27 Pa·s. This decline trend is more apparent for the coating sample with a higher addition level of HEC. This can be attributed to the deformation and breakdown of the structured pigments and the flocs, which is partly related to the addition of starch and HEC. Besides, the thickening effect of HEC on the paper coatings is visible in Fig. 2(b), showing an obvious increase in the viscosity of coatings with increasing the level of HEC. This result is in conformity with the rheological parameters supplied by HB model in Table 3. This finding could be due to the fact that HEC is a water soluble polymer which adsorbs water and forms a specified network in solution through simple entanglements of their hydrated chains [32]. It should also be pointed out that the strength of the network is considered to be associated with the molecular weight and the concentration of HEC in water [33].

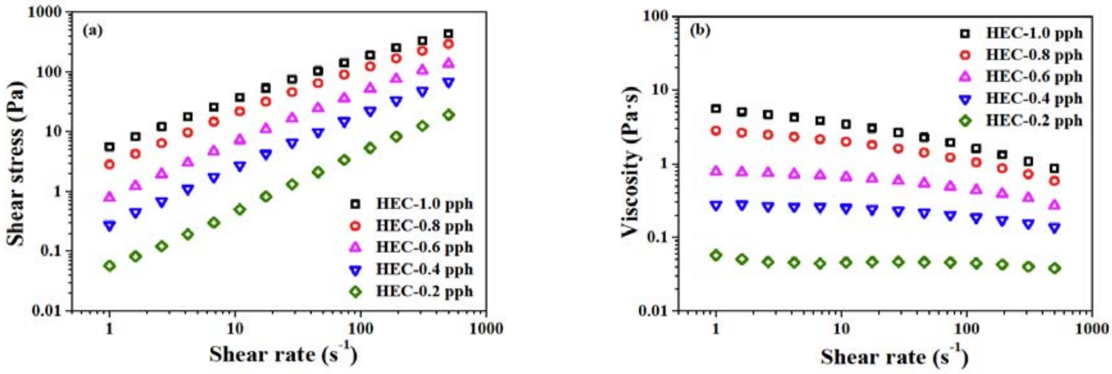

Fig. 2. (a) stress and (b) viscosity as a function of the shear rate for the paper coatings, with various levels of HEC. (Paper coating formulation: kaolin level of 96 wt%, TiO_2 level of 2 wt%, $CaCO_3$ level of 2 wt%. Based on the total weight of pigment, starch level of 14 pph, SHMP level of 0.6 pph. Solid content of paper coatings 55 wt%.)

Table 3 Rheological parameters of the HB model fitted to the rheological curves of the paper coating formulations with various levels of HEC

HEC level (pph)	τ_y *(Pa)*	*k (Pa s* ^{n}*)*	***n***
0.2	0.1021	0.0773	0.8878
0.4	0.6767	0.5688	0.7748
0.6	3.4852	2.4575	0.6526
0.8	8.1253	7.8338	0.5880
1.0	12.7102	14.0827	0.5563

3.3 Effect of starch on the steady shear rheological behavior of paper coatings

Effect of starch addition on the rheological properties of paper coatings was investigated, and the rheological curves are shown in Fig. 3. Similarly, HB model was employed to predict the rheological properties of paper coatings, and the corresponding rheological parameters are listed in Table 4. In general, the effect of latex on the low-shear viscosity of paper coatings is strongly associated with the interactions of the latex with other coating components. Fig. 3 shows a marked viscosity change with increasing the starch latex addition level, and the low-shear viscosity increases simultaneously. This is further verified by the rheological parameters in Table 4. It can be derived that the bigger value of τ_y can be achieved with the increase of the starch addition in the formulation. The result is consistent with the assumption that all polymers containing a substantial amount of hydroxyl groups can undergo hydrogen bonding with hydrated surface species on kaolin [25]. The latex used in this work contains abundant hydroxyl groups, and these hydroxyl groups can contribute to the interactions between latex and pigment component. This may be responsible for the significant increase in the low-shear viscosity with increasing the starch addition from 10 to 18 pph based on the weight of total pigment.

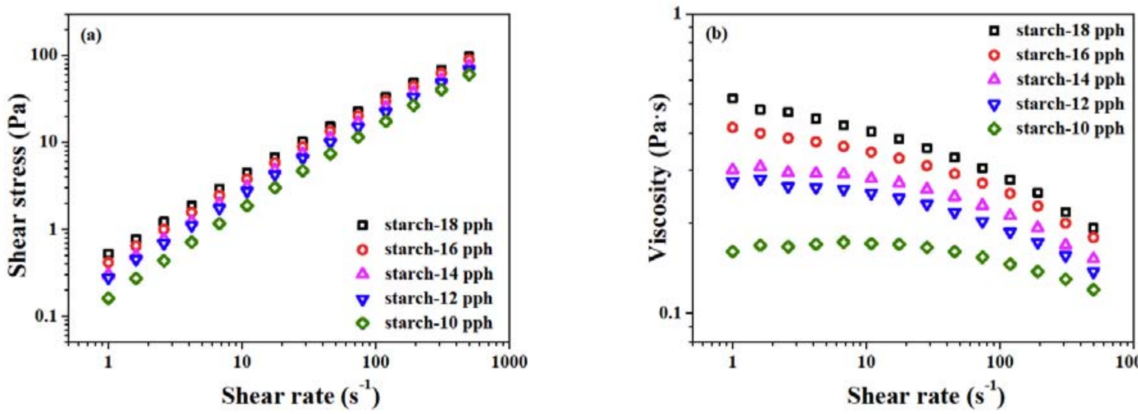

Fig. 3. (a) stress and (b) viscosity as a function of the shear rate for the paper coatings, with various levels of starch. (Paper coating formulation: kaolin level of 96 wt%, TiO_2 level of 2 wt%, $CaCO_3$ level of 2 wt%. Based on the total weight of pigment, HEC level of 0.4 pph, SHMP level of 0.6 pph. Solid content of paper coatings 55 wt%.)

Table 4 Rheological parameters of the HB model fitted to the rheological curves of the paper coating formulations with various levels of starch

Starch level (pph)	τ_y *(Pa)*	*k (Pa s ^{n})*	*n*
10	0.3255	0.2971	0.8560
12	0.8427	0.6814	0.7603
14	0.6767	0.5688	0.7748
16	0.8779	0.8010	0.7612
18	1.0696	0.9987	0.7377

3.4. Effect of dispersant on the steady shear rheological behavior of paper coatings

As can be seen from Fig. 4, paper coating samples with 0.4 pph, 0.6 pph, 0.8 pph, and 1.0 pph dispersant exhibit almost the same stress and viscosity curves over the entire shear rate range. Table 5 gives the rheological parameters from HB model of paper coatings with different dispersant addition level, and it shows intrinsic consistence in the value of τ_y, k and n of paper coatings. In addition, Fig. 4(b) shows that at the lower shear rate of 0-10 s^{-1}, the coating viscosity decreases significantly as the dispersant addition increases from 0.2 to 1.0 pph. However, at the higher shear rate (over 100 s^{-1}), the viscosity of all coatings displays uniform distribution regardless of the dispersant addition level. This behavior can be explained by virtue of the fact that at lower shear rate, particle-particle interactions of coatings tend to be more decisive, so the dispersant added to optimize the particle-particle interactions can be advantageous to repulsive surface charges. Whereas, at higher shear rate, the repulsive effect of dispersant on particle–particle interactions can be overlooked, and the shear stress may be consider to be critical to redisperse the particles in paper coatings [34].

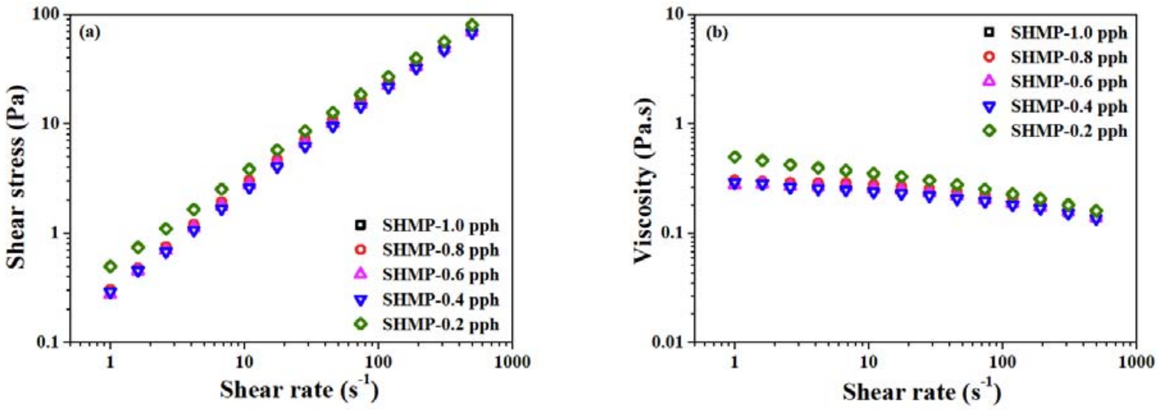

Fig. 4. (a) stress and (b) viscosity as a function of the shear rate for the paper coatings, with various levels of SHMP. (Paper coating formulation: kaolin level of 96 wt%, TiO_2 level of 2 wt%, $CaCO_3$ level of 2 wt%. Based on the total weight of pigment, starch level of 14 pph, HEC level of 0.4 pph. Solid content of paper coatings 55 wt%.）

Table 5 Rheological parameters of the HB model fitted to the rheological curves of the paper coating formulations with various levels of SHMP

SHMP level (pph)	**τ_y (Pa)**	**k (Pa s^n)**	**n**
0.2	0.6123	0.7651	0.7506
0.4	0.5733	0.6086	0.7902
0.6	0.6767	0.5688	0.7748
0.8	0.7534	0.6265	0.7739
1.0	0.6513	0.5964	0.7677

3.5. Effect of nanoparticle pigments on the steady shear rheological behavior of paper coatings

The role of nanoparticle pigment addition on the rheological behavior of paper coatings was emphasized. A series of coating formulations were performed by changing the addition level of $CaCO_3$ nanoparticles and TiO_2 nanoparticles, and the resulting rheological curves are given in Fig. 5. Accordingly, the rheological parameters are presented in Table 6. It is evident that the coatings containing nanoparticle pigments exhibit a rheological behavior of shear-thinning type over the entire shear rate range. Furthermore, when increasing the addition level of nanoparticle pigments, the as-obtained paper coatings appear to show a more apparent shear-thinning behavior. As shown in Fig. 5 (a), paper coatings show near Bingham plastic behavior having apparent extrapolated yield stress. The shear stress generally increases with increasing the addition level of nanoparticle pigments. Also, a yield stress τ_y can be observed for these coatings in Table 6. Moreover, Fig. 5(b) shows that, at the shear rate ranging from 0 to 10 s^{-1}, increasing the nanoparticle addition level leads to an obvious rise in the viscosity of paper coatings. This can be mainly due to the nano-scale particle size and high specific surface area of the added pigments, which would be favorable for the strong interaction between the nanoparticle pigments and other coating components. With further increasing the shear rate, the coating samples shows no indication of significant variation in the viscosity.

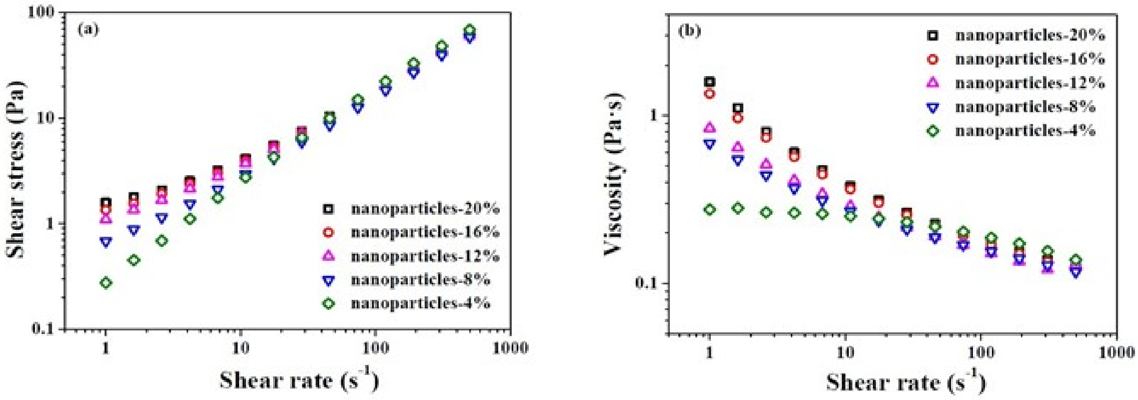

Fig. 5. (a) stress and (b) viscosity as a function of the shear rate for the paper coatings, with various levels of nanoparticles. (Paper coating formulation: kaolin level of 80-96 wt%, the proportion of TiO_2 to $CaCO_3$ is 1:1. Based on the total weight of pigment, starch level of 14 pph, HEC level of 0.4 pph, SHMP level of 0.6 pph. Solid content of paper coatings 55 wt%.）

Table 6 Rheological parameters of the HB model fitted to the rheological curves of the paper coating formulations with various levels of nanoparticles

Nanoparticle level (wt%)	*τ_y (Pa)*	*k (Pa s ^{n})*	*n*
4	0.6767	0.5688	0.7748
8	0.3474	0.3674	0.8152
12	0.6962	0.4625	0.7826
16	0.9344	0.4662	0.7788
20	1.1920	0.4366	0.7945

3.6. Effect of nanoparticle pigments on the dynamic viscoelasticity of paper coatings

It is well known that material viscoelasticity represents a combination of viscous and elastic behavior, which is also available in evaluating the flow behavior of paper coatings. The well-established parameters associated with dynamic viscoelasticity are storage modulus (G') and loss modulus (G''). Generally, oscillatory shear experiment, a frequency sweep where the amplitude of oscillation is constant while the frequency of oscillation is varied, can be considered as the preferable method to describe the viscoelastic behavior. Here, the storage modulus and loss modulus as a function of the frequency for paper coatings containing various levels of nanoparticle pigments were measured and are plotted in Fig. 6. It can be observed that the storage modulus G' is higher than the loss modulus G'' over an entire frequency range, providing direct evidence that the obtained paper coating samples containing nanoparticle pigments display a solid-like elastic behavior. More importantly, it is obvious that both the storage modulus and loss modulus show a visible increase with increasing the addition levels of nanoparticle pigments, which implies that the incorporation of nanoparticle pigments can impart a great amount of elastic and viscous energy to paper coatings. In particular, as the addition level of nanoparticle pigments increases from 12 to 16 wt%, a sharp increase in viscoelastic behavior occurs, as illustrated in Fig 6. These results strongly support the conclusion that the presence of nanoparticle pigments has a profound impact on the dynamic viscoelastic behavior of paper coatings. In general, the viscoelastic behavior of paper coatings is a function of the strength of bonds in the network formed. Actually, the nanoparticle pigments may be presumed to work similar to special fillers in the particle-particle interactions of coating components to form a network. Owing to its nano-scale size and high specific surface area, nanoparticle pigments would indeed exhibit a good filling effect and a selective filling feature, thus facilitating the strength of the obtained network structure. In addition, the storage modulus and loss modulus tend to go on rising as the frequency increases. When increasing the frequency up to 10 Hz, the storage modulus G' and loss modulus G'' of the paper coatings containing 16 wt% of nanoparticle pigments can reach 2628 Pa and 614 Pa, respectively, which is similar to the results reported in previous literature [25].

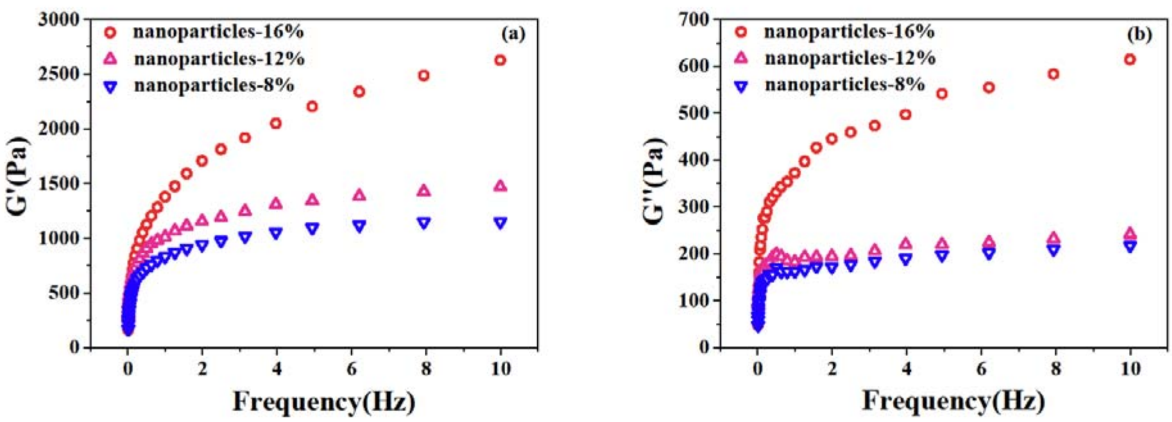

Fig. 6. (a) storage modulus G' and (b) loss modulus G'' as a function of the frequency for paper coatings, with various levels of nanoparticles.

4. Conclusions

A systematic study on the rheological behavior of paper coatings modified with inorganic nanoparticle pigments was conducted in this work. It was derived from steady shear rheological measurement results that almost all of the as-fabricated paper coatings have a non-linear flow curve and exhibit shear-thinning behavior. Also, an apparent extrapolated yield stress was found for these coatings. The influencing degree of all investigated factors toward steady shear rheological behavior can be ordered as follows: solid content > HEC addition > starch addition > nanoparticle addition > dispersant addition. Rheological parameters of Herschel-Buckley model was found to well describe the results obtained by shear rate dependent measurement. In another aspect, the storage modulus G' was higher than the loss modulus G'', demonstrating that the obtained paper coating samples displayed a solid-like elastic behavior. More importantly, both the storage modulus G' and loss modulus G'' showed a marked increase with increasing the addition level of nanoparticle pigments. All of the above results supported the conclusion that paper coating featured a complicated flow behavior, strongly associated with several main factors. The incorporation of nanoparticle pigments in paper coating indeed exerted an important influence on the rheological behavior, in particular on the dynamic viscoelastic behavior of the pigmented paper coatings.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (Grant No. 31100442), the Science and Technology Program of Zhejiang Province of China (Grant No. 2012C322080), the Science and Technology Program of Hangzhou City of China (Grant No. 20120433B63), the Science and Technology Program of Zhejiang Environmental Protection Bureau of China (Grant No. 2012B008), 521 Talent Cultivation Program of Zhejiang Sci-Tech University (Grant No. 11110132521310).

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