Morphology and Properties of Highly Talc- and CaCO₃-Filled Poly(vinyl alcohol) Composites Prepared by Melt Processing

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ABSTRACT: Highly filled poly(vinyl alcohol) (PVA) composites, PVA/talc and PVA/CaCO₃, were successfully prepared through melt-processing technology without use of coupling agents or compatibilizers. Morphology analysis showed that both of the two fillers well dispersed in PVA matrix. Differently, the flaky talc obviously oriented, while the particulate CaCO₃ was randomly scattered in PVA matrix. Rheological analysis showed that the two filled systems exhibited a common shear-thinning phenomenon, confirming their pseudoplastic nature. Compared with the particulate CaCO₃, the flaky talc exhibited a closer arrangement in the composite, resulting in a lower threshold filler content to form the network structure. Therefore, PVA/talc composite showed higher melt viscosity and lower extrudate-swell ratio. The orientation of the flaky talc particles in the system also resulted in the higher melting and heat-deflection temperatures, as well as a greater reinforcement effect on the composite, but a lower elongation at break. © 2013 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 130: 3050–3057, 2013

INTRODUCTION

Polymer/inorganic composites have attracted more and more attentions because of their low cost and good performance, e.g., high strength and stiffness, good heat resistance, and dimensional stability.[1–5] Poly(vinyl alcohol) (PVA) is a multi-hydroxyl polymer with high polarity, hence shows good compatibility with most inorganic/organic fillers, and PVA based composites with high performance are expected to be prepared without the need to introduce coupling agents and interfacial modifiers.[6] This largely avoids the performance loss of the composites, eliminates the unnecessary chemical pollution caused by small-molecule modifiers, and shortens the processing procedures.[7,8] Furthermore, PVA has good mechanical and barrier properties, excellent acid, alkali and organic solvents resistance, and biodegradability under certain conditions, thus has become one of the most popular environmentally friendly materials. Recently, there has been sustained growth in the number of the studies on PVA and its composites.[9]

Even though, high performance PVA based composites, especially highly filled PVA based composites, are quite difficult to be prepared, because the melt temperature of PVA is very close to its degradation temperature and almost no processing window is given to PVA. Up to now, most PVA products are based on and limited to wet-processing procedures, which tends to be complex with low efficiency and yield products, and cannot exhibit the excellent material properties of PVA. Consequently, melt processing technology with easy operation, low cost, high efficiency as well as environmental friendliness has always been the research focus for the development of the processing technology for PVA.

The key to realize the melt processing of PVA is to obtain a large processing window by increasing its degradation temperature or lowering its melting point.[10] So far, three main methods have been adopted to improve the processability of PVA: blending with other thermoplastic polymers, chemical modification, and plasticization.[11–14] Among them, plasticization is recognized as a simple, economic, and highly efficient technology.[15,16] However, it should be noted that in this technology, the partial evaporation and thermal degradation of small-molecule plasticizers at high processing temperature, which will result in the unstable flow behavior of the melt and poor appearance of the products, are key problems that must be solved.[17] Moreover, the introduction of inorganic filler will surely lead to an increase in the viscosity of the filled system, especially at high filler content, resulting in the significant deterioration of the processing properties. Therefore, the preparation of PVA/inorganic composites by melt processing, especially highly filled composites, is still a challenge. Fortunately, in our previous work, by adopting molecular complexation and plasticization, we have realized the stable thermal

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processing of PVA[10] and obtained a number of high-performance thermalplastic PVA products, such as melt-spinning fibers, extrusion-blowing films, hollow containers, and injection samples.[10,18] This provided the solid base for the preparation of PVA/inorganic filler composites with high performance and low cost.

As known, the properties of polymer/inorganic filler composites are not only related to the chemical structure of fillers, but also depend on their particle shape and size.[19,20] For example, fibrous, tubular, and flaky fillers with a large ratio of length to diameter can reinforce a polymer matrix more effectively. In this study, two frequently used inorganic particles with different shapes, flaky talc and particulate $CaCO_3$, were chosen as the fillers for PVA. By adjusting the processing conditions, PVA/talc and PVA/$CaCO_3$ composites with more than 50wt% filler content could be successfully prepared by extrusion and injection molding, which is a breakthrough for the PVA industry. The morphological, rheological, thermal, and mechanical properties of these PVA-based composites were also investigated using techniques including scanning electron microscopy (SEM), small-amplitude dynamic oscilatory measurement, capillary rheology, and differential scanning calorimetry (DSC) to gain a better understanding of the effects of these inorganic fillers on the properties of the composites.

EXPERIMENTAL

Materials
PVA1799 (degree of polymerization 1750, hydrolysis degree 99%, density 1.26–1.30 g/cm^3) was provided by the Sichuan Vinylon Factory, SINOPEC (China).

Talc (average diameter: $\sim$9 μm; density: $\sim$2.8 g/cm^3) and $CaCO_3$ (average diameter: $\sim$8 μm; density: $\sim$2.7 g/cm^3) were supplied by Shenzhen Hayond powder Technology Co. (Guangdong, China).

Preparation of Samples
Thermoplastic PVA (TPVA, MFI 10.5 g/10 min) was obtained by adding 75 wt % dried PVA into 25 wt % prepared aqueous solution of polyol, and then letting the solutions completely seep into PVA at ambient temperature in a sealed vessel.

PVA based composites were prepared by the following steps. The different amount of filler powders were firstly mixed with TPVA at the speed of 1200 rpm for 10 min in a FW-400 high-speed mixer (Zhongxing Instrument Co., China) to obtain the mixtures containing different filler contents (10, 20, 30, and 50 wt %, respectively). Then the collected mixture was further mixed in a SHJ-20 twin-screw extruder (Nianjing Giant Machinery Co., China) at 175°C to obtain the PVA composites, and subsequently injected by a single screw injection molding machine (Haitian Plastic Machinery Co., China) at 185°C to obtained the test specimens. The samples with talc and $CaCO_3$ were abbreviated as T - x and Ca - x, respectively; x was the weight percentage of the filler in the composite.

Characterization
Particle Size Analysis. The particle size distribution of the raw talc and $CaCO_3$ powders was characterized by laser diffraction, using a Malvern Master Sizer 2000, as shown in Figure 1.
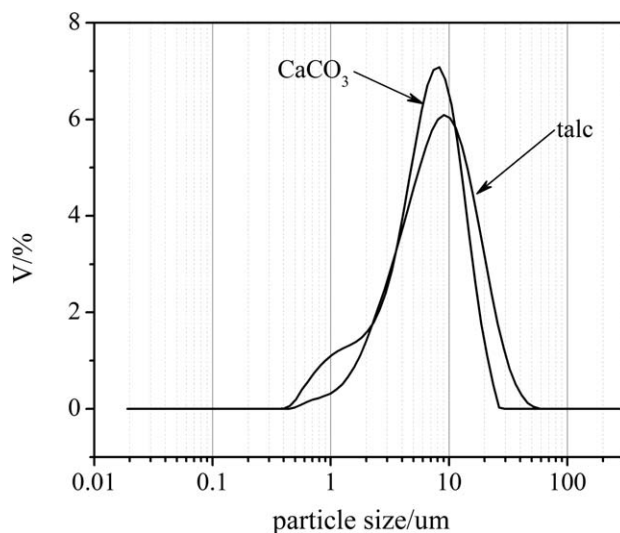

Figure 1. Particle size distribution of the used $CaCO_3$ and talc.

Scanning Electron Microscopy (SEM) Observation. The dispersion behavior of the filler in the composite was observed by using an Inspect F SEM instrument (FEI Instrument, Co.) at 0.5 Torr and 10 kV. Specimens were fractured at liquid-nitrogen temperature, and sputtered with gold for taking micrographs.

Dynamic Rheological Measurement. The dynamic rheological properties of TPVA and its composites were measured on a TA R2000 rotational rheometer (TA Instrument, Co.) with a parallel plate with 25 mm diameter and 1.5 mm thickness geometry. In order to avoid the evaporation of the plasticizers, a seal of silicon grease was used at the outer surface of the gap between the two plates. The tested samples with the diameter of 25 mm and the thickness of 2 mm were obtained by heat pressing at 180°C.

The oscillatory measurements were carried out to investigate the rheological behaviors of the melts of TPVA and its composites. During the measurement, the strain was set at 1% to ensure the linear viscoelastic region and the measured frequency range was 0.01–100 Hz.

Capillary Rheology Measurement. The capillary rheology measurements of TPVA and its composites were carried out on a RHEOGRAPH 2002 high-pressure capillary rheometer (Gottfert, Germany) with a diameter of 1mm and a length-to-diameter ratio of 16. The sample was firstly fed in the barrel of the rheometer and pre-heated for 3 min at 180°C. Subsequently, the measurement was performed in the shear rate range of 50–3000 s^{-1} for all samples. The extrudate-swell ratio (W) was calculated as the following formula: $W = (\phi - \phi_o)/\phi_o$. here, ϕ and ϕ_o were the extrudate diameter and the die diameter, respectively.

Differential Scanning Calorimeter (DSC) Analysis. The melting and crystallization behaviors of samples were studied on TA Q20 (TA Instrument, Co.) thermal analyzer under a nitrogen flow with a sample weight of approximately 6 mg. The melt curves of samples were obtained at a heating of 10°C/min from
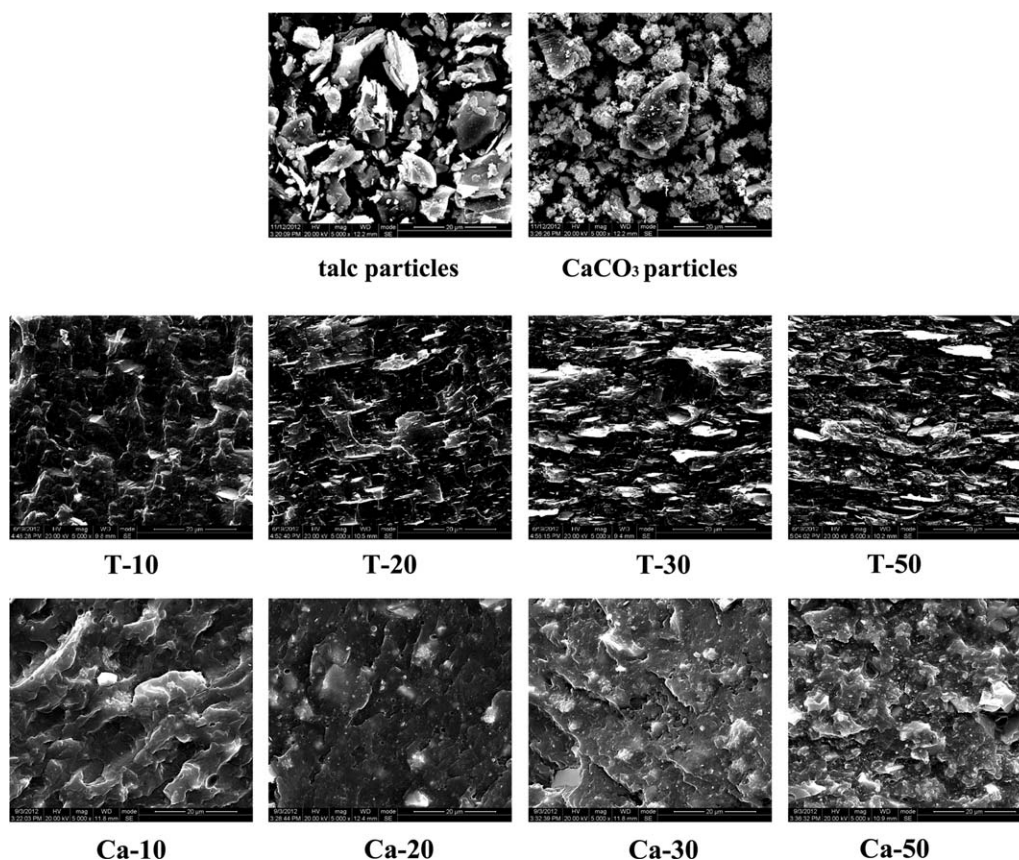
Figure 2. SEM images of original talc and CaCO$_3$ particles, PVA/talc and PVA/CaCO$_3$ composites.

20 to 250°C, while the crystallization curves were obtained by first heating the samples at the rate of 10°C/min from 20 to 220°C, then holding for 2 min, and last cooling the samples to the ambient temperature at the rate of 10°C/min. The samples were firstly heat-treated at 100°C for 12 h under vacuum before testing.

Heat Distortion Temperature (HDT) Measurement. HDT values of all composites were tested on a HDV2 (Atlas Instrument, Co.) apparatus at a heating rate of 2°C/min and a load of 0.455 MPa according to ASTM D648. All the samples were placed at 100°C for 12 h in an oven to remove the residual water in them before testing. The HDT value for each sample was the average of at least three successful tests.

Mechanical Measurement. The samples were heat-treated at 100°C for 12 h under vacuum to remove the residual water before testing. The tensile tests of the dumbbell-shaped samples were carried out on a RG-L-10 (Reger Instrument, Co., China) universal testing machine at 50 mm/min at room temperature according to the ASTM D638 test method. The values of the parameters were average values by calculation over fives specimens for each group.

RESULTS AND DISCUSSION

Dispersion of Fillers in PVA

Figure 2 shows the SEM images of PVA/talc and PVA/CaCO$_3$ composites with different filler contents. It could be seen that

both talc and CaCO$_3$ were well dispersed in the PVA matrix, suggesting the good compatibility of these two fillers with PVA. As well known, PVA is a multi-hydroxyl polymer with high polarity, the hydroxyl groups in it can form hydrogen bonds with the Mg–OH and Si–OH groups on the edges of the talc layers, and the highly polar points on the surfaces of the talc layers formed as a result of long-term exposure to air and water.[21] In the same way, these hydroxyl groups can form relative strong interfacial adhesion through the –OH and Ca^{2+} on the surfaces of CaCO$_3$ particles.[22] Therefore, the homogeneous dispersion of both fillers in the PVA matrix occurred without adding any coupling agents or other surficial modifiers. The strong interfacial interactions between the fillers and PVA were beneficial to exert the enhancing effects of the fillers on PVA and obtain the composites with designed properties.

An obvious difference in the dispersion morphologies of talc and CaCO$_3$ in PVA matrix could be seen by further observation, i.e., the regular orientation of talc flakes and the random dispersion of CaCO$_3$ particles. This might be caused by the different shapes or specific surface area of talc and CaCO$_3$. Compared with the original talc particles (Figure 2), the talc particles in PVA/talc composites were obviously smaller and thinner. The reason might be that when suffered the strong shearing and mixing force of the screw during melt processing, the asymmetric lamellar structure of talc was liable to dissociate owing to the weak interlayer van der Waals' forces, forming smaller,
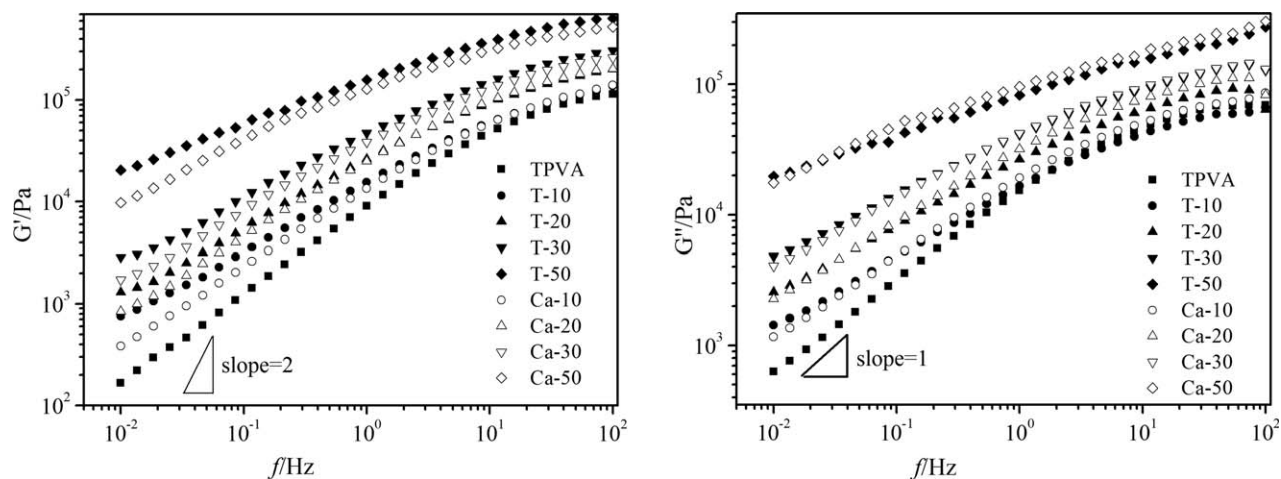
Figure 3. Plots of G' and G'' versus f of TPVA, PVA/talc and PVA/ CaCO$_3$ composites.

thinner slices that oriented along the melt-flow direction. While for CaCO$_3$ particles with regular symmetrical shapes and fewer degrees of freedom (DOF) than talc, no obvious orientation was observed. The different dispersion morphologies of talc and CaCO$_3$ resulted in their different effective contact areas with PVA, and might further influence the processing and service properties of their composites.

It could also be seen from Figure 2 that with the increase of filler content, the distance between filler particles became narrower and some particles even contacted each other, resulting in the increased interparticle interactions, especially at high content. The talc particles still retained the close orientation state and homogeneous dispersion in the PVA matrix. In contrast, the dispersion of CaCO$_3$ particles became more disordered and appeared stacks, which was liable to produce stress concentrations and defects, and was detrimental to the improvement of the properties of the obtained composites.

Rheological Behaviors of PVA Composites

Dynamic Rheology. Small-amplitude dynamic oscillatory measurement is an effective method for the analysis of the rheological behavior of a polymer composite and can reveal the microstructure and interactions in the composite. Figure 3 shows the plots of G' and G'' versus f for TPVA, PVA/talc, and PVA/CaCO$_3$ composites. Obviously, the values of G' and G'' of all the studied systems increased with the frequency and filler content, but the trend for each sample was different, especially at low frequencies. As well known, the high frequency region in the modulus curve represents the short time scale corresponding to outside stimulus, which only allows the small mobile units in the system to relax. The low frequency corresponds to the long time scale, which allows relatively large units such as polymer chain segments and molecular chains to move and relax, and can better reflect the different relaxation behaviors of materials with different microstructures.[23,24] As a result, the low frequency region is also called the fingerprint region. According to linear viscoelastic theory, for polymer melts without crosslinking and special interactions, the slopes of the plots for log G' and log G'' versus log f are 2 and 1, respectively. However, it was found from Figure 3 that although the linear relationship was still maintained, the slopes of log G' and log G'' versus log f for TPVA were smaller than 2 and 1, respectively, especially in the low frequency region. These deviations from the normal values might originate from the presence of dynamic hydrogen bonds between PVA molecules, which acted as the dynamic physical crosslinking points in the system, limiting the relaxation of PVA molecules. Compared with TPVA, the slopes of the plots for PVA/talc and PVA/CaCO$_3$ composites were further reduced, especially in the low frequency region. Nonterminal behaviors were observed in the G' curves when talc content was above 20 wt % and CaCO$_3$ content was above 30 wt %, indicating the reduction and even disappearance of the dependence of G' of the composite on f. This phenomenon was known as the "solid-like behavior,"[25] and should be attributed to the formation of the three-dimensional particle network in the composite caused by increased filler–filler and filler–polymer interactions. This behavior was also attributed to the emergence of the heterogeneous structure in the composite,[3] which further inhibited the movement of molecular chains or chain segments, causing them to show incomplete relaxation behavior. The "solid-like behavior" of the PVA-based composite exhibited a dependence on filler content and has been observed in other polymer-based composites.[26,27]

By comparing the relationships between the modulus and f of the composites, it was also found that the values of G' and G'' of the composites were higher than those of TPVA, and the amplification was enlarged with the increase of filler content. The increase in the modulus of the composite indicated the restrictive effect of the rigid particles and their network on the movement and relaxation of PVA molecular chains. Noticeably, PVA/talc composite showed higher G' and G'' values than PVA/CaCO$_3$ composite and its non-terminal behavior appeared at lower filler content, suggesting that PVA/talc composite melt had higher elasticity and more obvious "solid-like behavior" than PVA/CaCO$_3$ composite, which was perhaps attributed to the anisotropic nature of talc platelets. Compared with the regularly particulate CaCO$_3$, the asymmetric lamellar structure of talc endowed it with larger specific surficial area, leading to stronger interactions with PVA molecules and more contact among filler particles. Therefore, talc could more easily form a
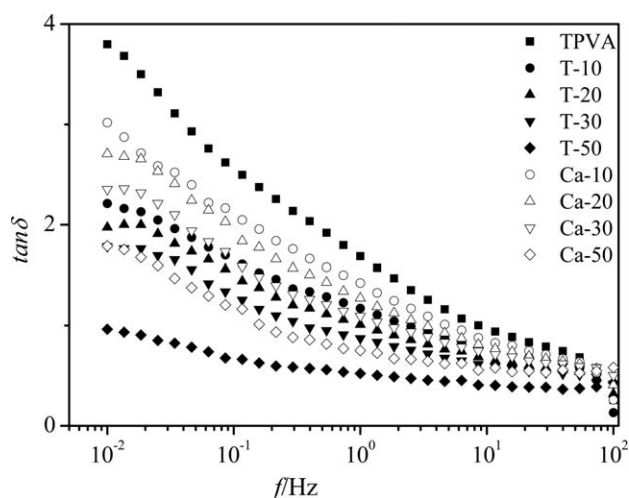
Figure 4. Tan δ plots of TPVA, PVA/talc, and PVA/CaCO$_3$ composites.

Figure 5. η of TPVA, PVA/talc, and PVA/CaCO$_3$ composites.

dense filler network and confine the movement and relaxation of more PVA molecular chains. The stronger interactions between talc particles also resulted in the greater inter-particle friction and collision during melt-flow processing of the composite, especially the highly filled composite, further restricting the relaxation behavior of PVA molecules. All of the above observations implied that talc might have more negative effects on the flow behavior of the PVA melt.

The value of tan δ (G''/G') can be used as an indicator of the elasticity of the melt in the studied system. Figure 4 shows the values of tan δ for TPVA, PVA/talc, and PVA/CaCO$_3$. Generally, a tan δ value above 1 implies that the polymer melt has a predominant viscous component, while values less than 1 indicate that the amount of the elastic component is higher. As shown in Figure 4, tan δ decreased with increasing frequency and filler content, which could be attributed to the confinement effect of the fillers on the movement of PVA molecular chains.[20] The value of tan δ of the talc-filled sample was lower than that of the CaCO$_3$-filled composite, indicating that the elasticity of PVA/talc composite melt was higher and the resistance of talc particles to the melt flow was greater during the processing. This might be due to the greater interactions between PVA and the flaky talc particles, and the denser filler network formed by the asymmetric talc.

Capillary Rheology. The rheological behavior of a polymer melt, which can not only affect the quality of the final products, but also provide information that can be used to optimize the practical processing conditions, is very important for the stable melt processing of polymeric material. Differing from the pure polymer, the viscosities of the filler filled polymer composites depend not only on the properties of the polymer, but also on the physical and chemical properties, size, shape, and content of the fillers.[3]

Figure 5 shows the dependence of shear viscosity of TPVA, PVA/talc, and PVA/CaCO$_3$ on the shear rate and filler content. The decrease in the viscosities of all samples with shear rate was indicative of their pseudoplastic nature. After adding fillers, the shear viscosity exhibited the obvious increase, especially at low
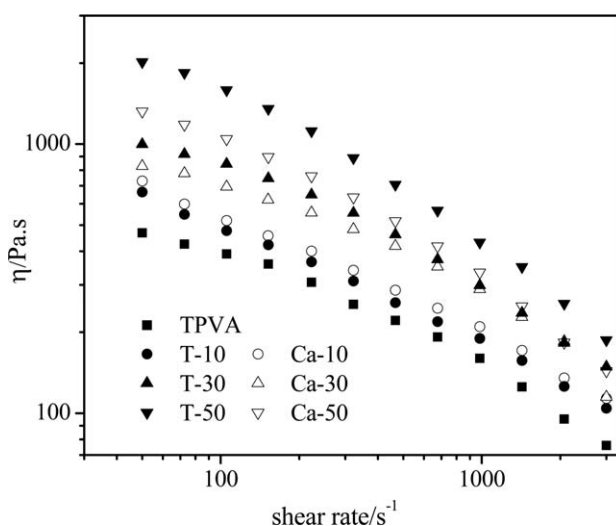
shear rate, owing to the spatial confinement of the fillers on the motions of the PVA molecular chains. It was found that the shear viscosity of PVA/talc composite was evidently higher than that of PVA/CaCO$_3$ composite, especially at high filler content. As known, the factors determining the rheological behavior of the filled system vary with filler content. At low filler content, there were not enough filler particles to form the network, so the flow behavior of the composite was mainly influenced by the hydrodynamic effects of the filler particles. As a result, the two composites showed similar shear viscosity at low filler content. As filler content increased, the filler particles gradually became closer and even contacted with each other, leading to the increase of the interactions between particles and the formation of the particle network in the system. This played a major role in controlling the rheological behavior of the composite at high filler content. The anisotropic talc flakes had larger specific surface area and length-to-diameter ratio than the regular CaCO$_3$ particles. So there were greater interactions between talc particles and easier formation of the filler network, which could confine the motion and relaxation of more PVA chains, and
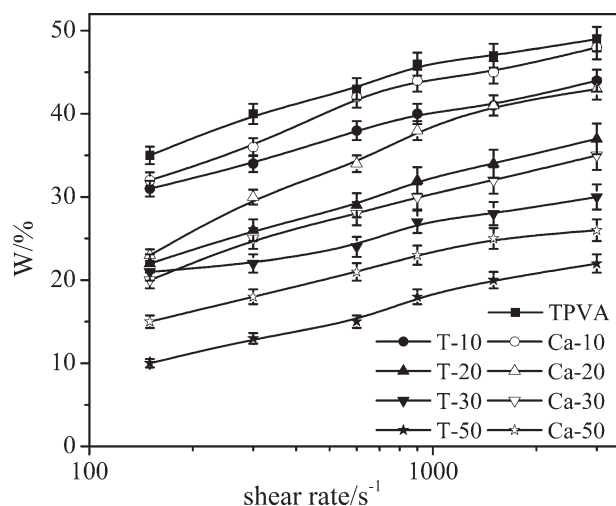

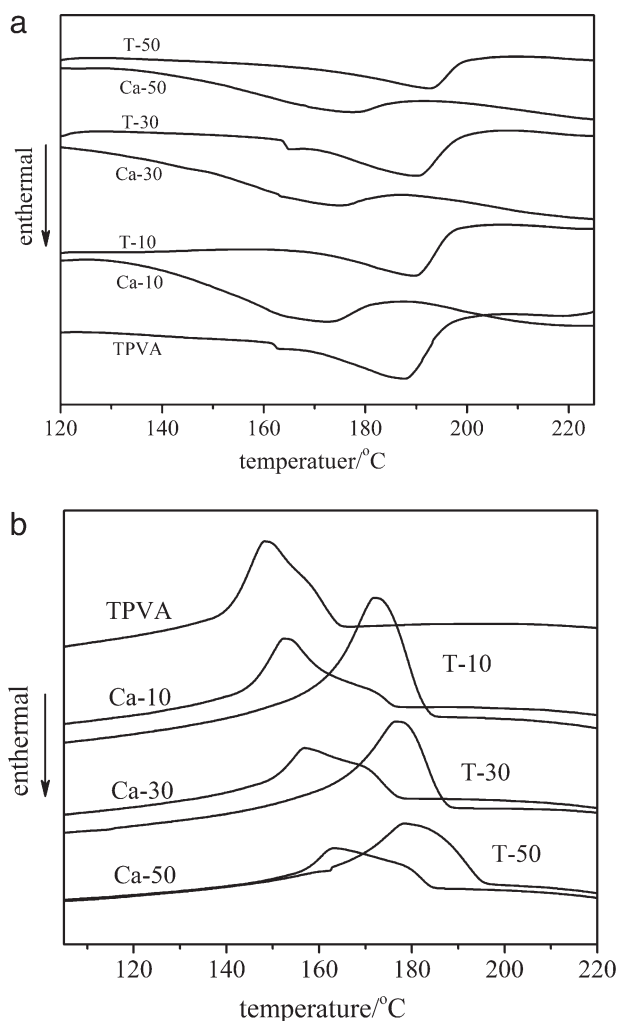**Figure 6.** Extrude-swell ratios of TPVA and PVA composites.

Figure 8. HDT of PVA/talc and PVA/CaCO$_3$ composites.

energy stored in the polymer melt flowing at higher shear rates. Apparently, the addition of inorganic fillers into TPVA lowered its extrudate-swell ratio. As well known, the extrudate-swell of polymer mainly depends on the recovery of elastic deformation imposed in the capillary. The presence of filler could increase the energy dissipation of the composite by the movement of the filler and limited the movement of polymer chains by its confinement effect, thus reducing the elasticity recovery and led to the lower extrudate-swell ratio of the composite. Moreover, with the increase of filler content, the effects of filler on matrix became more and more obvious, resulting in the decrease of elasticity recovery and extrudate-swell of the composite. Owing to the stronger confinement effect of the talc particles with large specific surface area, PVA/talc composites showed the smaller extrudate-swell ratios.

Thermal Behavior

Figure 7(a) showed the heating DSC curves of TPVA, PVA/talc and PVA/CaCO$_3$ composites. The thermal parameters obtained from the DSC curves were listed in Table I. Obviously, the melt enthalpies of the composites were lower than that of TPVA (ΔH_m: 38.5 J/g), indicating that the addition of the inorganic fillers suppressed the crystallization of PVA, which might be attributed to the confinement effect of the inorganic fillers on the movement of PVA molecular chains. However, this confinement effect of fillers on PVA molecules was quite different, leading to the higher enthalpy and shaper melting peak of PVA/talc composite compared with PVA/ CaCO$_3$ composite at the same filler content, also the opposite changes of melting temperatures of the composites compared with TPVA (T_m: 187.6°C). The

Figure 7. Heating (a) and cooling (b) DSC curves of TPVA, PVA/talc and PVA/CaCO$_3$ composites.

thus increasing the shear viscosity of the composite to a greater extent. The larger specific surface area of talc could also extend the diffusion path of the modifiers and enhanced the interactions between talc layers and modifiers, thus weakening the plasticization effect of the modifiers on PVA molecular chains. This resulted in the higher viscosity and lower fluidity of PVA/talc composite relative to PVA/CaCO$_3$ composite.

The extrudate-swell ratios of PVA/talc and PVA/CaCO$_3$ composites were also studied, as shown in Figure 6. The extrudate-swell ratio (W) of each sample increased with increasing shear rate ascribing to the enhancement of the recoverable elastic
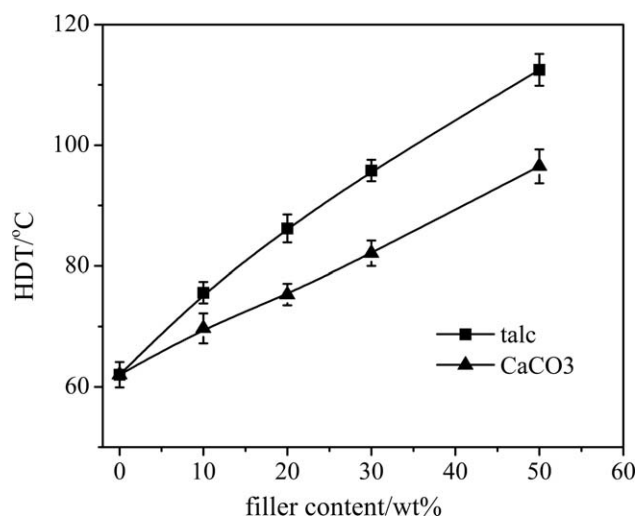
Table I. T_m and ΔH_m of PVA/Talc and PVA/CaCO$_3$ Composites

Filler content/wt %	T_m (°C)		ΔH_m (J/g)		T_c (°C)		T_0 (°C)	
	Talc	CaCO$_3$	Talc	CaCO$_3$	Talc	CaCO$_3$	Talc	CaCO$_3$
10	188.7	172.3	30.2	26.7	171.7	153.6	182.3	174.6
30	190.1	175.1	27.8	22.3	177.3	157.1	187.5	176.8
50	192.5	177.9	17.7	14.5	180.5	164.9	195.6	182.9

addition of talc enhanced the melting temperature of the system, while CaCO$_3$ resulted in the lower melting temperature. This could be ascribed to the larger surface area of talc relative to CaCO$_3$ and the orientation of the talc flakes, as shown in the SEM images. The orderly arrangement of talc might further promote the orientation of more PVA molecules, and therefore more nucleation effects were present during the crystallization of PVA/talc composites, resulting in the more ordered morphology and perfect crystals of PVA, as well as the higher melting temperature. The nucleation effects of fillers on PVA could be confirmed from the cooling DSC curves of TPVA, PVA/talc and PVA/CaCO$_3$ composites [Figure 7(b)]. Obviously, the onset crystallization temperature and the crystallization temperatures (T_c) of the composites were higher than that of TPVA, identifying the nucleation effect of inorganic fillers on the crystallization of PVA. Furthermore, T_c of PVA/talc composites was evidently higher than that of PVA/CaCO$_3$ composites at the same filler content, showing the stronger nucleation effect of talc on PVA. This might be related to the larger specific surface area and the greater orientation degree of talc flakes.

The heat-deflection temperature (HDT), as defined by ASTM D648, is the temperature at which a specimen deflects 0.25 mm under a designed stress, and is usually considered as the highest service temperature of thermoplastics. For polymer-based composites, the addition of inorganic filler usually improves the stiffness and HDT of the polymer matrix.[28,29] It can be seen from Figure 8 that the HDT of TPVA was approximately 62°C, and the HDT of the composites showed an approximately linear increase with respect to the filler content. This was due to the strong interactions between PVA–filler and the formation of the filler network, which limited the movement of the deformable PVA molecules. It can also be seen from Figure 8 that PVA/talc composite showed a higher HDT compared with CaCO$_3$-filled composite, and this difference increased as the filler content increased. This phenomenon was related to the dispersion states of the two different fillers and their anisotropy. The highly oriented talc particles in PVA matrix (Figure 2) were capable of confining the mobility of more PVA molecules, thus resulting in the higher HDT. Moreover, at high filler content, the network consisting of flaky talc particles was denser and the strength of the network was higher than the network consisting of granular CaCO$_3$ particles, thus largely increasing the HDT of PVA/talc composite. In addition, the lower melting point with the addition of CaCO$_3$ might be one of the reasons to cause the lower HDT of PVA/CaCO$_3$ composite compared with PVA/talc composite.

Mechanical Behavior

Figure 9 showed the dependence of the mechanical properties of PVA/talc and PVA/CaCO$_3$ composites. Apparently, the tensile strength and modulus of the two composites showed an obvious increase at low filler content, indicating that both fillers had reinforcement effects on PVA matrix owing to the strong interactions between PVA and fillers. However, after filler content reached a certain value, about 15 wt % for talc and 10 wt % for CaCO$_3$, the tensile strength of the composites decreased with further increasing filler content. This might be due to the stress concentration and defects caused by the increase of filler content in the composites. The modulus of both composites
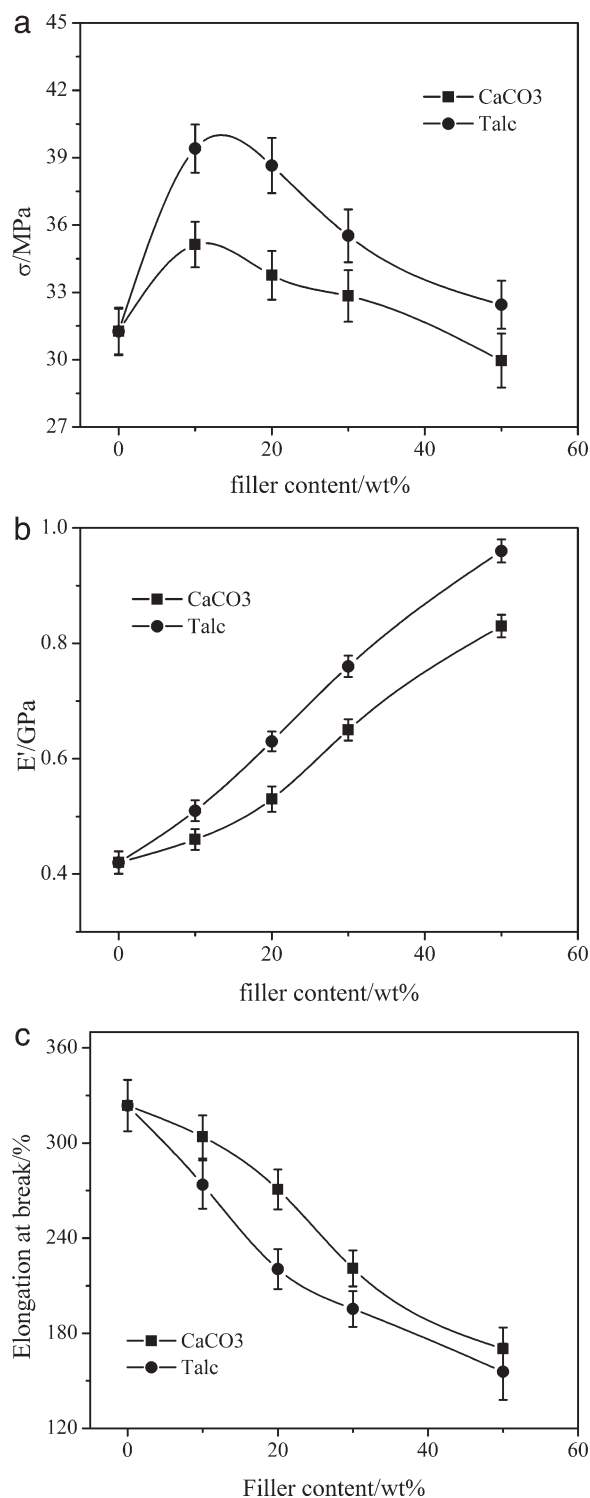

Figure 9. Mechanical properties of PVA/talc and PVA/CaCO$_3$ composites (a: tensile strength, b: modulus, c: elongation at break).

seemed to be mainly related to the content of the rigid filler rather than the interactions between filler and PVA or the dispersion quality of the filler in the composite, thus showing an approximately linear increase with the increase of filler content. Compared with CaCO$_3$-filled composite, PVA/talc composite had higher tensile strength and modulus at the same filler

content. This could be attributed to the stronger interactions between PVA and talc with larger surface area, which resulted in the less defects in the system and greater reinforcement effect of talc on the composite than that of $CaCO_3$.

The elongation at break of both composites continuously decreased with the increase of filler content ascribing to the confinement of the rigid fillers and their network on PVA chains. Compared with $CaCO_3$, the confinement effect of the anisotropic talc particles and talc network on the mobility of PVA molecules was stronger, resulting in the larger decrease of the elongation at break of PVA/talc composite.

CONCLUSIONS

Two highly filled PVA-based composites, PVA/talc, and PVA/$CaCO_3$, were successfully prepared through thermal processing without using any coupling agents or compatibilizers. The flaky talc particles produced a strong interfacial adhesion and were oriented along the melt-flow direction, while $CaCO_3$ particles were randomly scattered in PVA with no regular orientation. Compared with the $CaCO_3$-filled sample, PVA/talc composite exhibited a lower threshold filler content to form a three-dimensional network with higher melt elasticity because of the higher anisotropy and lager specific area of talc layers. The melts of TPVA and PVA-based composites exhibited a shear-thinning phenomenon, confirming their pseudoplastic nature. Compared with $CaCO_3$, talc resulted in a higher shear viscosity and shear stress, lower extrudate-swell ratio and higher melting and heat-deflection temperatures. Furthermore, the talc composite exhibited a higher tensile strength but a lower elongation at break owing to the greater confinement effect of the highly oriented talc layers on the motion of PVA molecules. The realization of the melt processing of the highly filled PVA/inorganic composites is the breakthrough in the processing technology of PVA, and will further enrich and extend the applications of PVA products in the fields of films, plates, tubes, containers, etc. However, the flow ability of PVA composites with more than 70 wt % fillers and the use of the composites in the relatively high humidity are still the challenge and should be resolved in our further work.

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