Journal of Alloys and Compounds 676 (2016) 513–520

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Study on the surface properties of colored talc filler (CTF) and mechanical performance of CTF/acrylonitrile-butadiene-styrene composite

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A R T I C L E I N F O

Article history:
Received 16 December 2015
Received in revised form
13 March 2016
Accepted 23 March 2016
Available online 25 March 2016

Keywords:
Talc
Filler
Inverse gas chromatography
Contact angle
Acrylonitrile-butadiene-styrene

A B S T R A C T

In this work, a novel colored talc filler (CTF) was prepared, and its surface properties were subsequently studied and compared to those of talc filler (TF) using inverse gas chromatography (IGC) and contact angle measurement. The mechanical properties of acrylonitrile-butadiene-styrene (ABS) composites filled with CTF and TF were investigated as well. The results indicated that the dispersive component (γ_S^D) for both samples contributed the major part of the total surface energy (γ_S^T). The values determined by the contact angle methods were consistent, although lower than those using IGC analysis. Compared to γ_S^D, the polar component (γ_S^{SP}) contributed less to γ_S^T, implying a lower polarity for both samples. The γ_S^{SP} values calculated by the contact angle methods were also consistent and lower than those calculated using IGC. The lower γ_S^T value for CTF could reduce filler particle-particle interactions, allowing a better dispersion in ABS matrix, and thus leading to an increase in ABS/CTF composite performance.

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1. Introduction

Talc is a plate-like layered structure magnesium silicate mineral, in which the octahedral brucite layer is sandwiched between two tetrahedral silica sheets. Many reports have indicated an improvement in performance after a polymer matrix was reinforced with talc filler [1–5]. However, it has been widely recognized that the filler nature of talc influences its reinforcement ability, depending on the surface activity, particle size, surface area, and surface functional groups [6,7]. Among these properties, surface activity affects the reinforcement ability of filler the most, because the chemical nature of a particle's surface determines filler–filler and filler-matrix interactions. These interactions in turn affect the filler's dispersion in the polymer matrix and thus the final performance of the composite. Therefore, a better understanding of a filler's surface properties is critical for determining the most effective polymer reinforcement fillers.

However, to the best of our knowledge, the surface characterization of talc-derived fillers has been poorly reported and is poorly understood. In this work, a colored talc filler (CTF) was prepared, and its surface properties were subsequently studied and compared to those of talc filler (TF), using IGC and contact angle measurement. The mechanical properties of ABS composites filled with CTF and TF were investigated as well.

2. Basic theory

2.1. IGC method

2.1.1. Surface energy

In IGC, the solids to be characterized are packed into columns, and different gases are injected. The injection of known polar and nonpolar gases enables the determination of the surface properties of the packed materials. Molecular probes are injected at infinite

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http://dx.doi.org/10.1016/j.jallcom.2016.03.189
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dilution to rule out lateral probe-probe interactions and to favor probe-stationary phase interactions only. Stationary phase characterization is achieved by partitioning the samples between the mobile and stationary phases.

According to Fowkes [8–10], the total surface energy (γ_S^T) is often divided into dispersive (γ_S^D) and specific (γ_S^{SP}) surface energy components. The dispersive interactions consist of London, Keesom and Debye interactions, while the specific interactions include acid-base interactions, H-bonding and π-bonding.

$$\gamma_S^T = \gamma_S^D + \gamma_S^{SP} \tag{1}$$

A standard procedure of solid surface characterization is that the γ_S^D is first calculated using a series of *n*-alkanes as probes (in this case, Hexane, Heptane, Octane, Nonane and Decane); then the acid-base parameters can be determined from the dispersive parameters with acid-base probe molecules (in this case, Dichloromethane (DCM), Toluene, Chloroform). For the calculation of γ_S^D, the Dorris-Gray method [11] is commonly used and thus it was applied in this work. The contribution of acid-base properties is often obtained by first measuring the specific Gibbs free energies of adsorption (ΔG^{sp}) for the various polar probes. From the ΔG^{sp}, it can be calculated the acid-base numbers related to the specific surface energy. Based on the van-Oss-Chaudhury-Good (vOCG) approach [12] and applying the Della Volpe scale [13] for surface tension components of polar solvents, the γ_S^{SP} is subdivided into Lewis acid (γ_S^+) and Lewis base (γ_S^-) components.

$$\gamma_S^{SP} = 2\sqrt{\gamma_S^+ \gamma_S^-} \tag{2}$$

2.1.2. Deconvolution of surface energetic sites

Surface energy heterogeneity, manifesting with a distribution of various surface sites with different energy levels, constitutes an energetic map of the solid surface and thus allows a prediction of the materials properties, in particular the formulation of composites, adhesives or blends. Such a heterogeneity profile can be represented by an energy distribution function. IGC is not the only means of measuring surface energy heterogeneity and alternative techniques are considered for measuring surface free energy. However, the contact angle measurement requires macroscopic crystals in order to determine specific energy contributions. Such measurements on specific crystalline facets may neglect edge effects and defects, and reflect a bulk average surface energy for solids. Another approach is adhesion force measurement by atomic force microscopy, although there are theoretical and technical challenges to this methodology. IGC provides data over a wide range of probe surface coverages, yielding information about the relative heterogeneity of the surface energy distribution of a material. However, existing methods for interpreting IGC data are based on inappropriate assumptions, giving purview to the development of new approaches to the analysis of surface-energy heterogeneity.

A more robust method for computing the surface energy distribution of heterogeneous powders has recently been proposed. Based on sounder thermodynamic assumptions, Smith et al. [14] developed a new numerical model for energy calculation, expanding on initial efforts from the model proposed by Jefferson et al. [15]. Using this approach, the surface energy distribution is determined by fitting simulation parameters to experimental IGC data. A computer algorithm is used that varies the model parameters and compares the resulting simulation with the experimental data. The process is repeated until small changes in the model parameters do not improve the fit of the simulation to the experimental data. It is worth noting that, the use of thermodynamic principles for the filling of energetic sites is an important step in characterizing the effect of differing energetic contributions. This is a nontrivial problem, as can be seen by the nonlinear relationship displayed by mixtures of materials at different energies. In this instance, it would seem appropriate to use multiple single energy values at different weights, to simulate the different states. To account for slight defects around these energy sites, the use of Gaussian distribution for each was used:

$$f\left(\gamma_S^D\right) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\gamma_S^D - \mu}{\sigma}\right)^2\right] \tag{3a}$$

where σ represents the standard deviation and μ is the center of the energy. The standard deviations of the Gaussian functions are fixed at one. The distributions for all the energy states are then combined to form a single distribution, whose sum is normalized to the value 1 (as this sets the total energy sites to 100% of the surface).

2.2. Contact angle methods

There are numerous methods for calculating the solid surface energy from equilibrium liquid contact angles. Among these methods, the Zisman plot [16–18], Fowkes [19,20], Owens-Wendt-Kaelble (OWK) [21,22], vOCG [23] and Wu [24,25] are commonly used and thus were applied in this work.

The key equation used to determine the solid surface energies by contact angle measurement is the Young's equation [26], which was derived from the equilibrium condition of forces representing surface tensions at the contact point of three phases: solid, liquid and gas.

$$\gamma_L \cos\theta = \gamma_S - \gamma_{SL} \tag{3b}$$

where γ_S, γ_L and γ_{SL} are the surface free energies of solid, liquid and solid–liquid (mJ/m^2), respectively; and θ is the contact angle between the solid surface and the test liquid (°).

In the Young's equation, both θ and γ_L are measurable. In order to obtain γ_S and γ_{SL} by solving this equation, an additional relationship between these quantities has to be made. An understanding of the different methods requires an explanation of the term “work of adhesion (W_A)”. Thermodynamic adhesion is the *de facto* energy spent to restructure the bonded interface due to the atomistic interaction between two materials. The equation for W_A can be written as:

$$W_A = \gamma_A + \gamma_B - \gamma_{AB} \tag{4}$$

where γ_A and γ_B represent the surface tensions of phases A and B; and γ_{AB} represents the interfacial tension between the two phases. For the solid–liquid system, the equation can be written as:

$$W_A = \gamma_S + \gamma_L - \gamma_{SL} \tag{5}$$

Combining it with the Young's equation yields:

$$W_A = \gamma_L(1 + \cos\theta) \tag{6}$$

In a similar way, the work of cohesion (W_C) of one substance (e.g., A) can be defined as:

$$W_C = \gamma_A + \gamma_A - 0 = 2\gamma_A \tag{7}$$

Berthelot [27] stabilized the direction to surface energy calculations and assumed that W_A between the solid and the liquid equals the geometric mean of individual cohesion work.

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