Surface Modification of Talc Particles with Phthalimide: Study of Composite Structure and Consequences on Physical, Mechanical, and Optical Properties of Deinked Pulp

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Commercial talc was modified with phthalimide to produce a composite filler, which was characterized by Fourier transform infrared spectroscopy (FTIR), X-ray diffraction spectroscopy (XRD), X-ray fluorescence spectroscopy (XRF), and scanning electron microscope (SEM). It was used as a papermaking filler, and its efficiency was compared with that of plain commercial talc and when mixed with additives (*i.e.*, cationic polyacrylamide and alum-rosin size). The results showed that phthalimide could be linked appropriately to the surface of talc particles through forming ester bonds with the surface hydroxyl groups. The brightness of the composite filler was more than that of the commercial filler due to a reduction in the impurities and removal of the metal compounds. Unlike other treatments, the brightness of the papers filled with modified talc (MT) was enhanced with increases in filler loading. In spite of reducing the paper strength in all treatments, the reduction was significantly less in the MT treatment, which implies the enhancement in particle tendency to develop hydrogen bonds in the fiber network.

Keywords: Composite filler; Talc; Phthalimide; Brightness; Paper strength

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INTRODUCTION

The steady increase in population and cultural development has led to a rise in the consumption of paper. The increasing per capita consumption of paper along with the steady decrease in the wood resources that provide the cellulosic raw material for the paper industry has made it necessary to pay more attention to paper recycling. Office waste papers are an important and remarkable source of chemically treated fiber and have a significant capacity for recycling and reuse in the production of different types of papers such as printing and writing papers. In addition, recycling processes such as deinking allow for improvement in the performance of recycled fibers for reuse in paper production. However, there are some problems associated with recycling such as the decrease in paper quality, the cost of collection, and the lack of a continuous supply of office waste papers to recycling plants, which limit their use in combination with virgin fibers (Ghasemian *et al.* 2009).

A general goal of paper production is to use fillers that have a lower cost than the cellulosic raw material; however, the use of fillers, especially when used at a high loading level, can create paper defects, poor filler retention, and decreased paper strength, sizing performance, and bending stiffness. Fillers can also increase wire abrasion and dusting during the printing process. Several methods can overcome or decrease at least one of these defects by forcing filler into the cellular cavities and/or walls of the pulp fibers (Klungness *et al.* 2000), making a filler/fines complex by premixing the fillers with fines/fibrils or precipitating the filler particles on the fines/fibrils (Silenius 2000; Subramanian *et al.* 2005, 2006, 2007, 2008), replacing the traditional fillers with new ones (such as starch-based biodegradable organic fillers and inorganic fibrous fillers with a high aspect ratio and highly desirable properties) (Hu and Deng 2004; Mathur 2004; Mollaahmad 2008; Shen *et al.* 2010; Peltonen *et al.* 2013), using functional strengthening agents before formation of the paper wet web (Chen *et al.* 2005; Fahmy and Mobarak 2008, 2009), fiber modification (Bratskaya *et al.* 2006; Kang 2007), and filler modification (Zunker 1985; Gill 1992; Shen *et al.* 2009, 2010; Song *et al.* 2009a,b; Laleg *et al.* 2011).

Talc—a hydrated magnesium-covered silica with the chemical formula of $Mg_3Si_4O_{10}(OH)_2$—is a hydrophobic filler (Ciullo and Anderson 2002). Talc is considered an inorganic polymer based on its two basic “monomer” structures: the silica tetrahedral and the magnesia octahedral. These minerals contain a continuous octahedral layer with the joined octahedral tied on a triangular side. This layer is bound on both sides by a continuous silica layer (Papirer *et al.* 1993; Charnay *et al.* 2001). It is found as either a white, apple green, or gray powder with a luster, pearly, or greasy appearance and a Mohs hardness of 1 to 1.5. It has low electrical conductivity and high resistance to acid, alkali, and heat. Talc filler is a hydrophobic material that easily blends and disperses in organic media including polymers, but it is not easily dispersed in aqueous solvents (Anderson 1996; He and Dunn 2004). Talc is widely used as a fine powder in several industrial products such as paper, paints, rubbers, polymers, ceramics, and putties (Johnson 1997; Johnson and Virta 2000; Ferrage *et al.* 2003a,b; Okunlola *et al.* 2003; Chandra *et al.* 2004; Boghdady *et al.* 2005; Helmy *et al.* 2005; Lefebvre *et al.* 2011; Bacchin *et al.* 2012; Marzbani *et al.* 2013). Talc possesses many favorable properties such as being organophilic and chemically inert and having a plate-like structure, which are the reasons for its use as filler in many different type of papers. The large relative particle size distribution of talc filler compared with other fillers leads to better retention, smoothness, and printability and less negative effects on the mechanical properties of papers (Ciullo and Anderson 2002; Wilson 2006; Vipul *et al.* 2011).

Recently, more attention has been given to evaluating the possible changes of the main properties of talc during processing because specific treatments such as mechanical grinding (Aglietti and Porto Lopez 1992; Liao and Senna 1992; Aglietti 1994; Filio *et al.* 1994; Sanchez-Soto *et al.* 1997; Kano and Saito 1998; Godet-Morand *et al.* 2002; Christidis *et al.* 2004; Terada and Yonemochi 2004), sonication (Perez-Maqueda *et al.* 2005; Perez-Rodriguez *et al.* 2006), flotation, leaching, magnetic separation (Piga and Marruzzo 1992; Houot *et al.* 1995; Boulos 2004; Ahmed *et al.* 2007), and surface modification (Lasmarias *et al.* 2003; Zhang *et al.* 2004; Wang and Somasundaran 2006; Lefebvre *et al.* 2011; Bacchin *et al.* 2012) can be used to modify the intrinsic characteristics of talc.

Bacchin *et al.* (2012) used carboxyl methyl cellulose (CMC) for surface pre-coating of talc particles. The results showed that CMC adsorption increased the negative surface charge and the wettability of the filler. Particle aggregation is prevented by the adsorbed CMC layer inducing electrosteric repulsion between the talc particles, which drastically improves the dispersion of talc particles. In another study by Lefebvre *et al.* (2011), hydrophobic silica (Aerosil R972®) was used to modify the surface of talc particles *via* a dry coating method. The coating obtained was a discrete coating of silica on the surface of the talc particles, which makes the talc more hydrophobic.

Lasmarias *et al.* (2003) invented a new method for surface modification of talc to improve its wettability and increase its affinity to cellulosic fibers. Talc particles were coated with aluminum hydroxide and then with a special cationic polymer, Alcofix® 131 (DADMAC homopolymer). The positive effect of this modification was evident, as the modified talc-filled papers had the best tensile strength and porosity compared with other treatments.

Hydrated aluminum oxide ($Al_2O_3.3H_2O$) and a mixture of phosphorus pentoxide (P_2O_5) and hydrated aluminum oxide were used by Zhang *et al.* (2004) to modify talc particles and study its effects on filler retention in the papermaking system. The talc modification had great impact on filler retention. The greatest retention occurred in the papers filled with talc particles modified by the mixture of $Al_2O_3.3H_2O$ and P_2O_5.

The goal of the present study was to use phthalimide (1,3-dihydro-1,3-dioxoisoindole; $C_6H_4(CO)_2NH$) to modify a talc particle surface and investigate the effect of using the composite talc as a papermaking filler on the physical, mechanical, and optical properties of deinked pulp.

EXPERIMENTAL

Materials

The materials used in this research are listed in Table 1.

Table 1. Materials Used in the Papermaking

Material	Manufacturer	Role
Commercial deinked pulp	Latif Paper Company, Karaj, Iran	Cellulosic raw material
Commercial talc	Merck KGaA Chemicals Co. Darmstadt, Germany	Filler
Composite talc	Invented	Filler
Saponified rosin	Ecopower Chemical Co., Ltd., Yuexiu, China	Sizing agent
Alum	Yixing Cleanwater Chemicals Co., Ltd., Yixing, China	Mordant agent
Cationic polyacrylamide (cPAM)	Yixing Cleanwater Chemicals Co., Ltd., Yixing, China	Retention agent

Methods

Filler modification and modification procedure

A mixed sample was prepared from commercial talc and phthalimide powders at equivalent molar weights and transferred into a three-neck flask. Next, 250 mL of xylene was added to the mixture as the reaction substrate. Nitrogen gas was blown into the flask for 1 min to remove oxygen gas and prevent oxidation of the mixture. The flask opening was closed immediately after the cessation of nitrogen gas. The mixture was heated at 70 °C in an oil bath under reflux for 24 h with continuous stirring. At the end of the treatment time, the mixture was decanted. The mixture was kept motionless to allow the formation of two separate phases. The solvent, which was on the top, was decanted up to the boundary between the phases, and the main mixture was washed with xylene three times to remove free phthalimide particles. Finally, it was poured into a petri dish and placed in an oven at 50 °C for 24 h.

Because the modified talc particles were agglomerated after the chemical modification, the talc was ground using a Fritsch planetary ball mill (Pulverisette 5 classic line, Idar-Oberstein, Germany) to make them smaller and to obtain a uniform particle size. The powder and balls were placed into separate bowls at a powder/ball weight ratio of 1 to 10. The balls were made of tempered steel, and the big ball/small ball ratio was 1 to 8. The final composite filler was prepared after grinding at 200 rpm for 30 min.

Table 2. Suspensions Used for Handsheet Preparation

Treatment	Identification Code	Suspension Type	
		Filler Loading Level (%)	Components
B	B	-----	Blank (DIP pulp without any additives)
T	T3	3	DIP pulp + talc only (based on the dry weight of the fiber)
	T5	5	
	T7	7	
	T15	15	
	T20	20	
TS	TS3	3	DIP pulp + talc + 2.5% saponified rosin in the presence of 5% alum (based on the dry weight of the fiber)
	TS5	5	
	TS7	7	
	TS15	15	
	TS20	20	
TP	TP3	3	DIP pulp + talc + 0.2% cPAM (based on the dry weight of the fiber)
	TP5	5	
	TP7	7	
	TP15	15	
	TP20	20	
MT	MT3	3	DIP pulp + modified talc (based on the dry weight of the fiber)
	MT5	5	
	MT7	7	
	MT15	15	
	MT20	20	

Spectroscopy and Microscopy Analysis

Wide-angle X-ray diffraction (XRD) patterns were performed for the powder specimens on a Bruker D8 Advance (Karlsruhe, Germany) using Ni-filtered CuKα radiation (35 KV and 30 mA). Fourier-transform infrared (FTIR) spectra were recorded using a Perkin-Elmer Spectrum RXI spectrometer (Rodgau, Germany) with KBr pellets. The chemical compositions of the commercial and composite talc were determined by a Bruker energy dispersive X-ray fluorescence spectrometer (XRF) (AXS GmbH-S4 EXPLORER). Field emission scanning electron microscope (FE-SEM) images were recorded using a Hitachi S4160 (Tokyo, Japan) to investigate the particle surface. TAPPI test method T534 om-03 (2003) was used to determine the filler brightness.

Handsheets

Paper sample preparation

TAPPI standard method T205 sp-02 (2002) was used to make handsheets. For this purpose, 21 different suspensions were prepared (Table 2). The circular sheets with a grammage of 60 gsm and surface area of 200 cm^2 were made and then, pressed for 4 min using a hydraulic press. Finally, they were air-dried for 24 h and conditioned in a climate chamber in standard conditions (50% relative humidity, 23 °C).

Paper Properties Analysis

Filler retention

Filler retention was measured by burning the paper sheets in a furnace at 525 ± 25 °C for 60 min and Eq. 1, according to TAPPI standard T211 om-02 (2002).

$$Filler\ retention = \frac{Ash\ weight\ after\ burning\ in\ the\ furnace\ (g)}{Filler\ weight\ used\ for\ each\ sheet\ (g)} \times 100 \qquad (1)$$

Physical Properties

After conditioning, the filled and unfilled hand sheets were weighed, and the grammage was calculated by Eq. 2, according to TAPPI standard T410 om-02 (2002):

$$Grammage\ (g/m^2) = \frac{Sheet\ weight\ (g)}{Sheet\ area\ (m^2)} \qquad (2)$$

Sheet thickness was measured using a PTI N1101 manual micrometer (Birkenau, Germany) according to TAPPI standard T411 om-05 (2002). Sheet density was calculated using Eq. 3.

$$Density\ (g/cm^3) = \frac{Sheet\ grammage\ (\frac{g}{m^2})}{Sheet\ thickness\ (mm)} \qquad (3)$$

Air resistance depends on paper permeability and is measured by different methods. The Gurley method, which measures the amount of time required for a certain volume of air to pass through a test specimen, was used to determine sheet air resistance on a Rycobel Group Gurley standard densometer (model 4110N, Deerlijk, Belgium) according to TAPPI standard method T460 om-02 (2002).

Optical Properties

Paper brightness is based on the amount of light reflection in the blue part of the visible light spectrum (250 nm) compared with magnesium oxide. Sheet brightness was measured using a PTA TS 40600 brightness-colorimeter according to TAPPI standard T452 om-02 (2002).

Opacity is generally calculated as a contrast ratio, *i.e.*, the ratio of light reflection from the surface of a single sheet with a non-reflective black surface backing to the reflection from the surface of a stack of the paper. TAPPI standard T425 om-01 (2001) was used to determine sheet opacity on a PTA TS 40600 brightness-colorimeter.

Mechanical Properties

Bursting strength is expressed as the resistance of a paper sheet against deformation by a rubber diaphragm with a 31.5 mm diameter until the paper bursts. The sheet burst strength was measured using a PTI F18533.e010 burst tester according to TAPPI standard method T403 om−02 (2002).

Tearing resistance is the amount of energy required to tear paper at a specified distance after the initial tear. TAPPI standard method T414 om−04 (2004) was used to determine the sheet tearing resistance on a PTI Elmendorf Tear Tester F53984.

Barrier Properties

Water absorptiveness (Cobb value) is expressed as the amount of water absorbed by 1 m^2 of paper that is placed under 1 cm of water for a specified time. The sheet Cobb value was measured by Cobb tester a4665 according to TAPPI standard method T441 om−04 (2004).

FE-SEM Images

FE-SEM images were taken to study the particle distribution in the prepared handsheets and effects on the fiber network.

Statistical Analysis

Experiments were performed in triplicates (n=3). Results were tested for three-stage nested design and Duncan's post hoc test. The statistical analysis was performed using SAS software (SAS Institute Inc., Ver. 9.0, Cary, NC, USA). Significant difference was defined at $p \leq 0.05$.

RESULTS AND DISCUSSION

Efficiency of Modification Process

The XRD patterns in Fig. 1 show the presence of basal diffractions of talc (T) (001), (002), (003), and (020) in the structure of the composite talc (Ferrage *et al.* 2003b), but their positions were not affected. Therefore, the talc kept its crystalline structure.

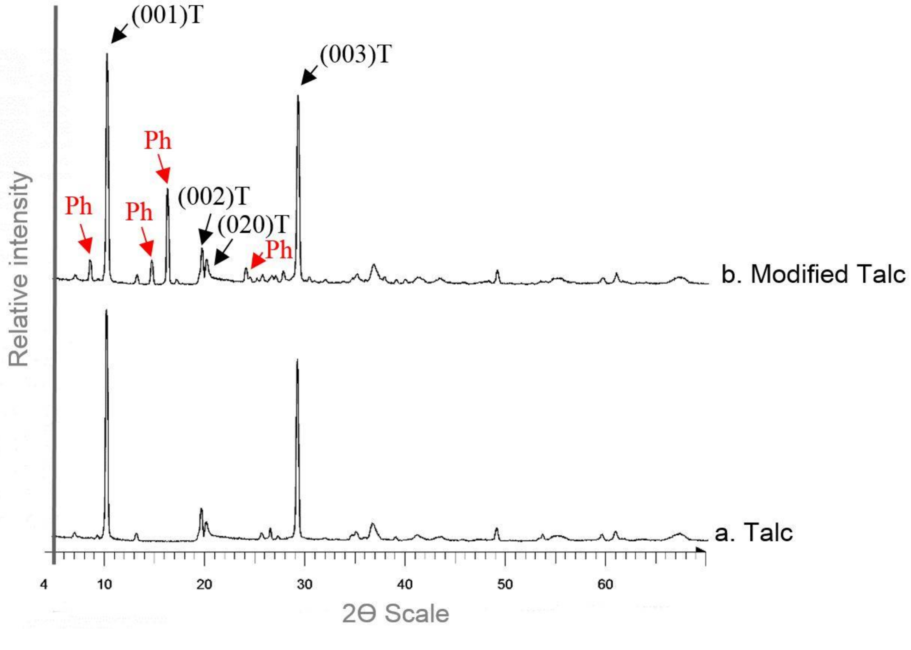

Fig. 1. XRD patterns of (a) commercial talc and (b) composite talc

The presence of phthalimide (Ph) diffractions in the XRD pattern is related to the covering of the particle surface by a thin layer of phthalimide.

Four typical peaks were specified in the FTIR spectrum of the commercial talc, with the 1019 cm^{-1} peak corresponding to the Si-O tetrahedral layer and the 3679, 675, and 458 cm^{-1} peaks representing the MgO/MgOH octahedral layer (Fig. 2a) (Schroeder 2002). As observed in the spectrum of the phthalimide (Fig. 2c), the 1600 cm^{-1} peak shows the stretching vibration of C=C bonds in the aromatic ring, and the 3305 and 1713 cm^{-1} peaks are related to the stretching vibrations of secondary N-H amide bonds and conjugated C=O bonds, respectively.

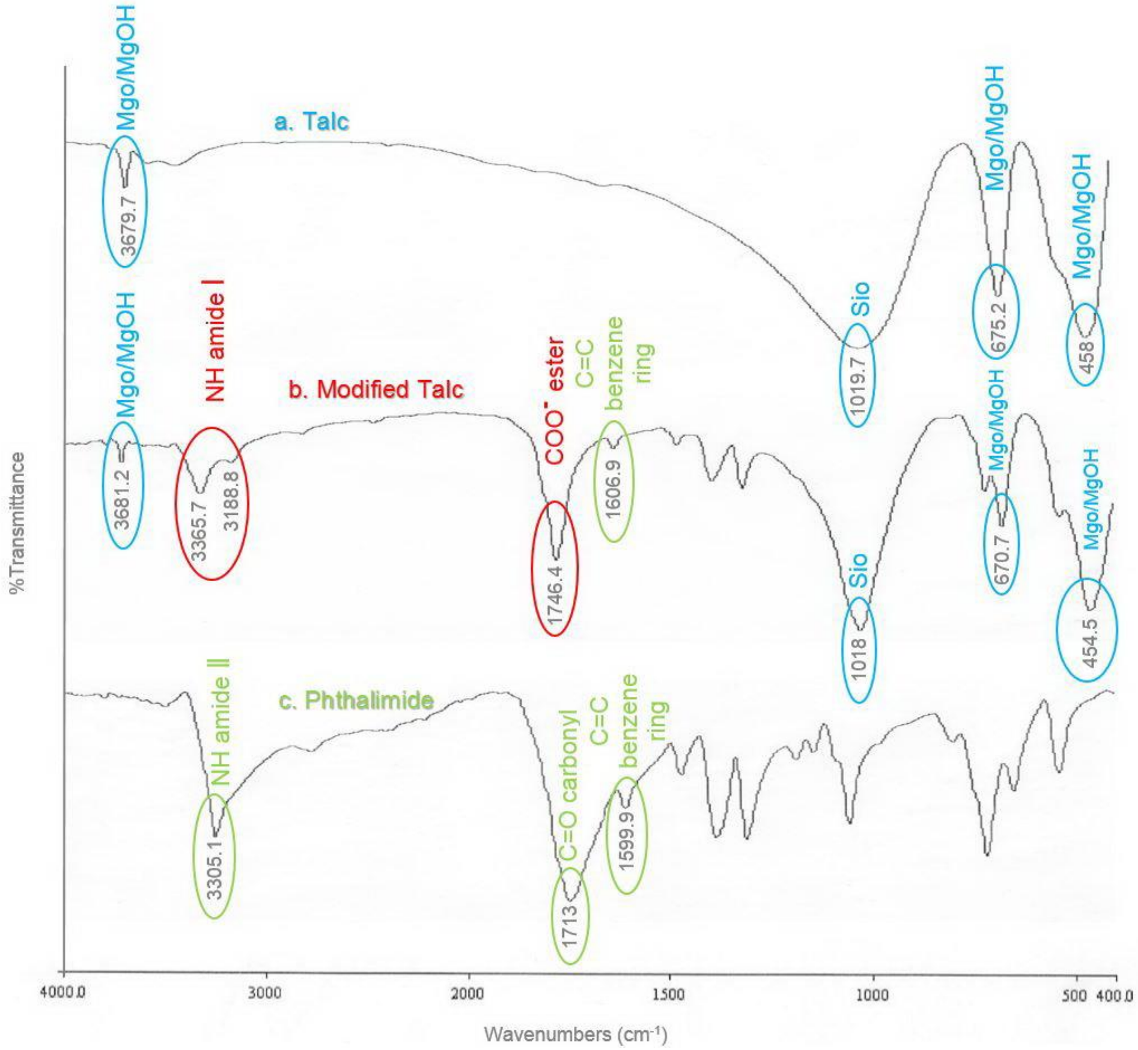

Fig. 2. FTIR spectra of (a) commercial talc, (b) composite talc, and (c) phthalimide

The peak at 1746 cm^{-1} shows that ester bonding (COO-) occurred, and the 3188 and 3365 cm^{-1} peaks reveal the formation of primary N-H amide bonds (Fig. 2b) (Sherman-Hsu 1997; Pavia *et al*. 2009). This result demonstrates that phthalimide formed ester bonds with the surface hydroxyl groups of the talc particles according to the proposed chemical reaction shown in Fig. 3.

Phthalimide molecules contain N-H groups that can develop hydrogen bonds; therefore, the commercial talc changed from being hydrophobic to relatively hydrophilic due to the chemical modification, which is an important phenomenon in the papermaking system. Commercial mineral fillers with poor bonding ability strikingly reduce the filled paper strength (Shen *et al.* 2010), while composite talc establishes hydrogen bonds with fibers to develop fiber-to-fiber bonding. This fact reveals the importance of chemically modified fillers in composites.

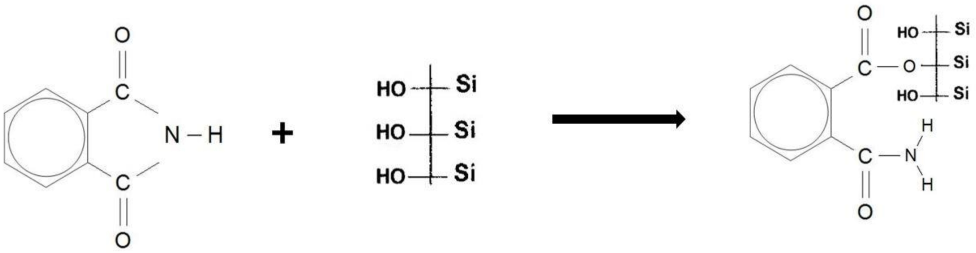

Fig. 3. The chemical reaction between the talc surface and phthalimide

In the SEM image of commercial talc, the particles appeared as flat tabular crystals (Fig. 4a). Some changes occurred in the surface morphology of the talc particles due to the chemical modification. Most of the modified talc particles looked like huddled clustered masses (Fig. 4b). Due to grinding, the large particle flocs were crushed, and a more uniform distribution of particle size was obtained (Fig. 4c,d).

Table 3 shows the chemical compositions of the commercial and composite talcs, as determined by the XRF analyzer. The results reflect the fact that the impurities of the composite talc decreased for some compounds: TiO_2, P_2O_5, CaO, Fe_2O_3, and Al_2O_3. In addition, the metal compounds Pt and TeO_2 were completely removed from the modified talc structure. This may be due to the extraction effect of xylene used as the reaction media. The loss on ignition (L.O.I.) significantly increased in the modified talc (approx. 26%), which corresponds to the presence of the organic compound of phthalimide. Due to reducing the impurities and metal compounds (especially Fe_2O_3, which has a significant effect on the yellowness of the powder), and good coating of talc particles by phthalimide molecules, the brightness of the composite talc significantly increased from 80% to 84% ISO, which is of great importance to the paper industry.

Table 3. Chemical Composition of Commercial and Composite Talc

Talc Samples	Commercial Talc	Modified Talc
Pt	0.082	-
TeO_2	0.259	-
TiO_2	0.031	0.022
P_2O_5	0.044	0.031
CaO	0.317	0.224
Fe_2O_3	0.700	0.515
Al_2O_3	1.015	0.749
MgO	31.667	22.559
SiO_2	58.155	42.515
L.O.I. (1050 °C, 1 h)	7.73	33.40
L.O.I., Loss on ignition		

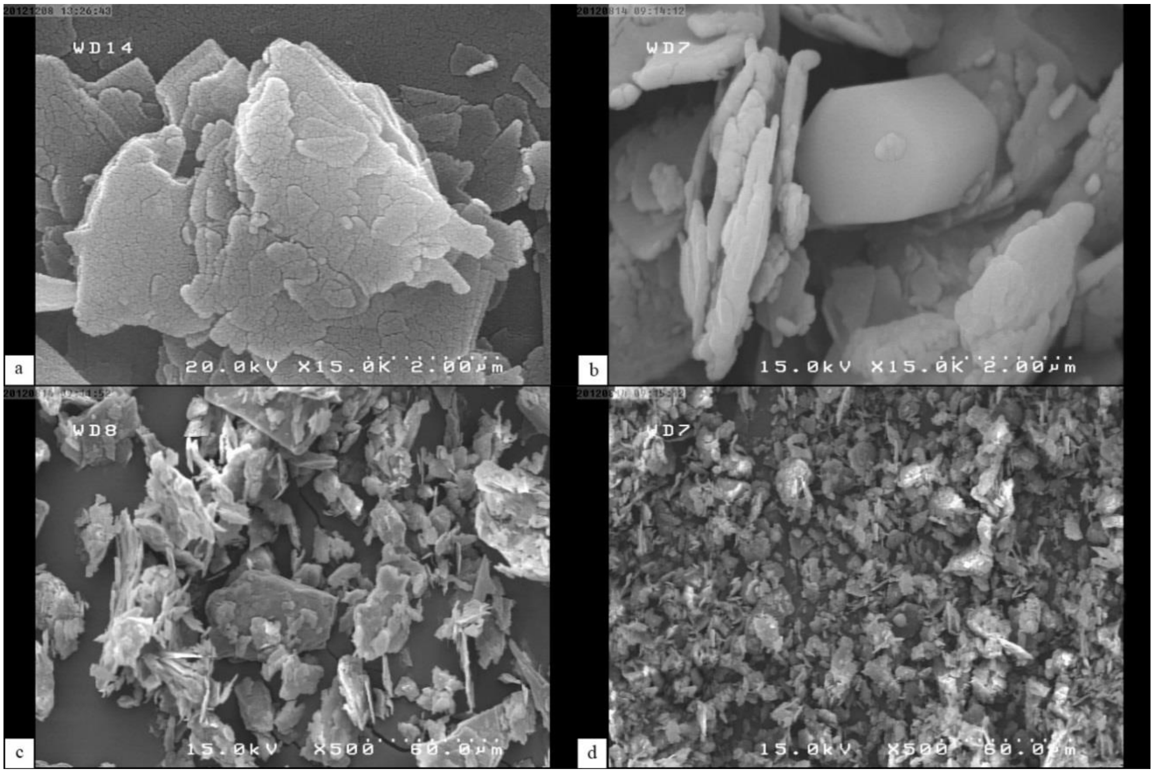

Fig. 4. SEM images of (a) commercial talc, (b, c) unground, and (d) ground composite talc

Filler Efficiency in Papermaking

The results of the paper tests are summarized in Table 4. Filler retention has great effects on the final product price, papermaking system cleanliness, and pollution load of the wastewater system (Vengimalla *et al.* 1999). As shown in Table 4, filler retention in the TP treatment was greater than in the other treatments, with the MT treatment having the lowest retention. This represents the high ability of cPAM adsorbing onto the fiber, fines, and fillers and forming polymeric bridges between the particles (Lindström and Glad-Nordmark 1978; Tanaka *et al.* 1999; Zhang *et al.* 2003; Hubbe *et al.* 2009). Composite talc contains about 26% phthalimide (Table 3), and it is an organic material in which only minerals remain during paper burning. Thus, at the same filler loading level, less minerals remained in the MT treatment than in the other treatments.

Apparent density and air resistance are two other essential parameters in papermaking. Different fillers may have different effects on these parameters. Studying the effects of filler addition on the air resistance and apparent density can lead to a better understanding of the filler effects on the paper properties.

The density of the filled papers was more than in the blank sheets, and it was enhanced with increasing filler loading level. This may be because the filler had a greater density than the fiber. In fact, the density of most fillers is in the range of 2.5 to 3 g/cm^3 (talc: 2.8 g/cm^3), which is much greater than the density of cellulosic fibers (approximately 1.5 g/cm^3) (Gill and Hageymeyer 1992; Velho 2002). The MT treatment resulted in a greater density than that of the other treatments. Because the composite talc contained N-H groups due to modification by phthalimide molecules, it could develop hydrogen bonds with the fiber surface and thus form sheets of less bulk (Fig. 5e).

Table 4. Properties of the Different Handmade Sheets

Treatment	Filler Retention (%)	Density (g/cm^3)	Air Resistance (S)	Brightness (%)	Opacity (%)	Burst Index (KPa.m^2/g)	Tear Index (mN.m^2/g)	Cobb Value (g)
B	-----	0.4688	1	81.15	89.20	1.542	6.995	105
T3	63.3	0.4725	1.067	81.11	89.41	1.480	6.979	90.67
T5	60.0	0.4754	1.156	81.07	89.54	1.450	6.952	82
T7	56.5	0.4783	1.244	81.04	89.62	1.401	6.924	75
T15	52.7	0.4844	1.444	80.93	89.83	1.298	6.880	69.67
T20	48.2	0.4898	1.6	80.83	90.04	1.212	6.846	63.33
TS3	83.2	0.4701	1.122	81.05	89.44	1.446	7	20.67
TS5	75.0	0.4743	1.256	80.97	89.65	1.343	6.945	19.33
TS7	69.7	0.4792	1.367	80.90	89.86	1.229	6.895	18.67
TS15	64.5	0.4870	1.578	80.78	90.07	1.092	6.831	17.33
TS20	59.2	0.4921	1.778	80.70	90.28	1.003	6.787	16.67
TP3	90.2	0.4731	1.267	81	89.62	1.527	7.022	87.67
TP5	82.6	0.4765	1.467	80.87	89.86	1.434	6.977	78.67
TP7	78.0	0.4801	1.667	80.75	90.07	1.350	6.937	71.33
TP15	72.4	0.4909	1.978	80.54	90.31	1.193	6.866	60.67
TP20	68.5	0.4962	2.178	80.45	90.52	1.104	6.81	52.33
MT3	58.4	0.4742	1.089	81.25	89.25	1.540	7.033	78
MT5	53.5	0.4784	1.244	81.4	89.44	1.531	7.018	72.67
MT7	47.1	0.4834	1.467	81.65	89.60	1.492	7	67.67
MT15	42.8	0.4952	1.778	82.51	90.04	1.431	6.950	54.33
MT20	38.4	0.5032	2.1	83.01	90.25	1.380	6.918	45

Paper's air resistance is reduced by decreasing the filler particle size and increasing the particle aspect ratio. Particle shape is also an important parameter in relation to air permeability of filled papers. Plate-like fillers can be oriented in parallel with the paper surface by the pressure applied during pressing and calendaring and thus improve paper gloss and smoothness. For a certain particle size, they can significantly reduce the sheet air permeability (Velho 2002). The reason for this phenomenon is unclear; the plate-like particles may form masses and close the fiber network structure, blocking the air passageways (Perng and Wang 2012).

The air resistance of the blank sheet was relatively low. This may be due to the open and porous structure of the sheets made from the deinked pulp, which is related to the poor bonding ability of used fibers. By adding filler, the air resistance of the filled papers increased significantly, and it was greatest for the TP treatment. This results from having a higher ash content and thus more blocked air passageways (Fig. 5d). The paper filled with the modified talc had greater air resistance than the two other treatments (*i.e.*, T and TS treatments) while it also had less ash. This may be due to the bonding ability of the composite filler with the fiber surface to form less porous sheets (Fig. 5e). An increase in filler loading level resulted in enhanced air resistance for all treatments. This increase was more significant in the MT treatment, as there was no significant difference between the MT and TP treatments at the 20% loading level.

In addition to reducing the production costs, improving the optical properties of paper is usually the most important role of fillers. When development of new filler production technologies is studied, the effect of filler addition on the optical properties of papers should always be considered (Shen *et al.* 2010). Generally, fillers have a significantly higher brightness and refractive index than cellulosic fibers. The performance of mineral fillers for the improvement of paper opacity depends on the filler's ability to scatter light, which is affected by the size, shape, and refractive index of the particles, and their uniform distribution in the paper (Velho 2002).

The initial brightness of the commercial talc used in this research was 80% ISO, which was less than that of blank paper; therefore, the filled papers had a lower brightness than that of the blank paper. The lowest paper brightness was found in the TP filled papers, which had a higher ash content than the other treatments. With increasing filler loading, these papers had the greatest decrease in brightness. The brightness of the composite talc increased from 80% to 84% ISO due to chemical modification; therefore, the greatest brightness was obtained from the papers filled with the composite filler and contrary to other treatments, an increase in filler loading led to enhanced brightness for this treatment. In contrast to brightness, the opacity of the filled papers was greater than that of the blank paper, and it was enhanced for all treatments with increasing filler loading level. Therefore, the TP treatment had the highest opacity due to having the highest ash content.

There is an effect of fillers, especially at relatively high loading levels, that should be considered. They can strikingly reduce the paper strength due to two important reasons. First, mineral fillers have poor bonding ability compared with the fibers, and when placed at fiber-to-fiber contact points, they can certainly interfere with fiber-to-fiber bonding. Second, substitution of fibers with mineral fillers reduces the number or amount of fibers in the paper network, which can inescapably play a negative role in the development of paper strength (Fairchild 1996; Kurrle 1996; Silenius 2000; Subramanian *et al.* 2005, 2006, 2007, 2008; Yan *et al.* 2005; Zhao *et al.* 2005, 2008; Yoon and Deng 2006a,b, 2007; Yoon 2007; Fahmy and Mobarak 2008, 2009; Song *et al.* 2009a,b; Deng *et al.* 2011; Laleg *et al.* 2011).

Generally, the burst index of blank paper was greater than that of the other treatments. By adding the filler to the papermaking suspension, the burst index decreased due to disrupting the fiber-to-fiber bonding. Deinked pulp contains sticky contaminants, and talc particles have a greater ability to trap them in the presence of cationic charge supplying materials (*i.e.*, alum and cPAM) (Gao *et al.* 2011). Therefore, some of the fiber-to-fiber contact points were inhibited, which caused a severe reduction in burst index of these treatments, especially at higher loading levels. In the presence of talc, which is a hydrated magnesium silicate, the magnesium ion can cause premature agglomeration of the rosin size.

This agglomeration may cause the carboxyl groups on rosin to become blocked, and no longer able to participate in interactions with the alum as a mordant material. Thus, the rosin size remained in an untreated form with the paper sheet, even after the paper was dried (Fig. 5c). This effect can explain the lower burst index of the TS treatment as compared with the other treatments.

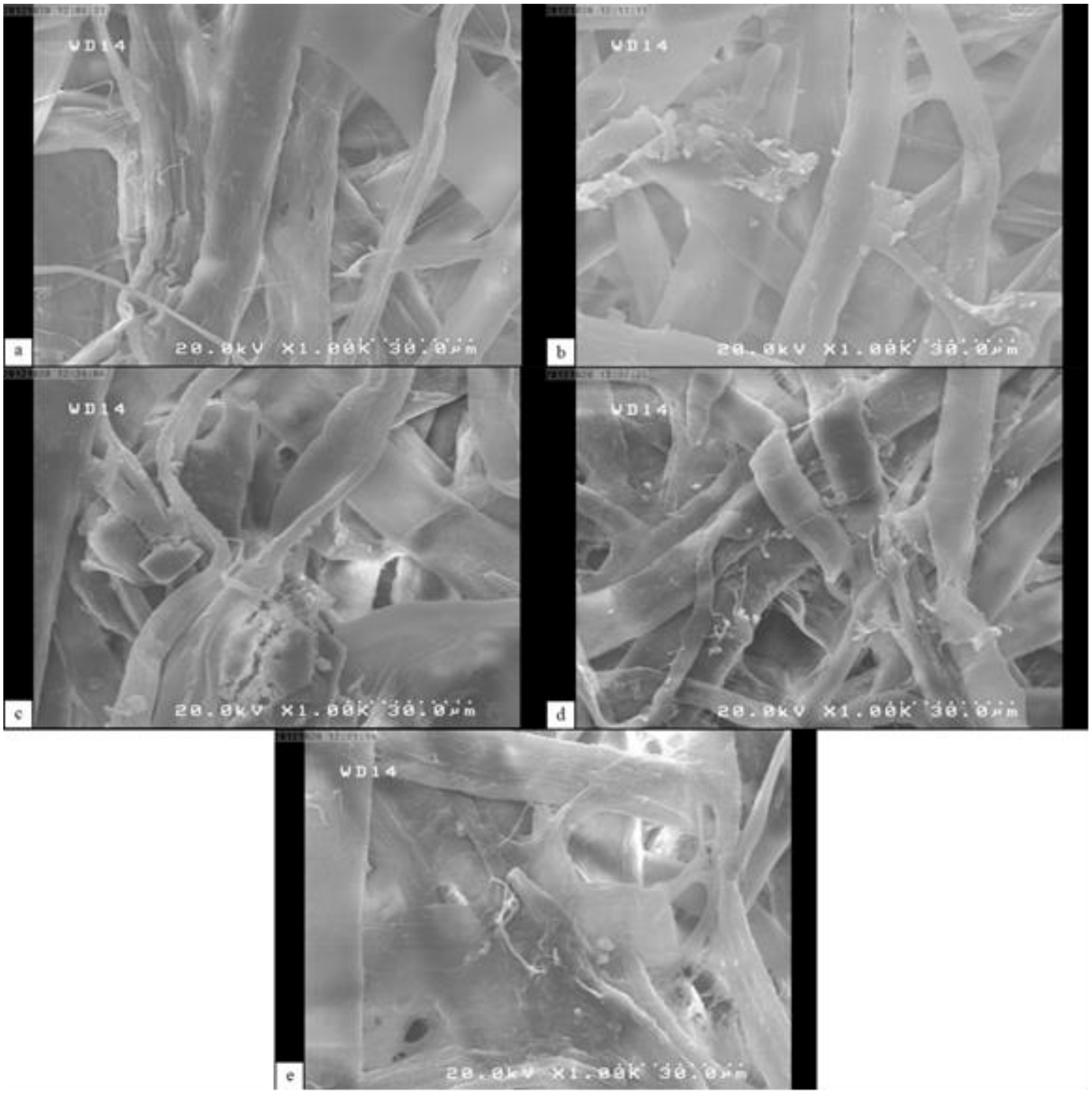

Fig. 5. FE-SEM images taken from the surface of (a) blank paper, (b) T-filled paper, (c) TS-filled paper, (d) TP-filled paper, and (e) MT-filled paper at 3% loading level

The burst index of the MT-filled papers was greater than that of the other treatments for each loading level; therefore, there was no significant difference between the burst indices of this treatment and the blank paper at the 7% filler loading level. This manifests the development of fiber-to-filler bonding in the fiber network (Fig. 5e).

Tear strength of paper depends on the fiber strength, which is related to fiber length and flexibility (Casey 1981). Decreasing the tear index is a result of increase in paper stiffness due to filler addition, which reduces the paper flexibility (Casey 1981), and the reduction of the fiber-to-filler ratio in the paper, which decreases the fiber-to-fiber bonding (Shen *et al.* 2010). Tear index was the lowest in the TS treatment, which can be related to poor fiber-to-fiber bonding due to placing the premature agglomerated rosin particles in the fiber-to-fiber contact points. Unlike the TS treatment, there was no significant difference in the tear indices of the blank paper and the MT filled papers at the 7% loading level, which implies the high bonding ability of the composite filler.

As shown in Table 4, the water absorptiveness of the filled papers was strikingly less than that of the blank paper due to blocking of the fiber-to-fiber contact points, which are free hydroxyl groups on the fiber surface that can absorb water. Rosin particles contain abietic acid, which is a polycyclic compound (Strazdins 1989; Lee *et al.* 2006). In general, it is hydrophobic unless it contains a carboxylic acid (Hubbe 2006). When there is a mordant material such as alum in the media, these molecules are oriented in such a way that the hydrophobic end is outward and then, sticks to the alum and deposits on the fiber. The fiber surface becomes hydrophobic and tends to absorb less water; therefore, the water absorptiveness of the TS treatment was much lower than in the other treatments.

Phthalimide has a hydrophilic head (-NH) and a hydrophobic end, and composite talc develops hydrogen bonds with the fiber surface at the hydrophilic head. During paper drying, the filler particles are oriented in such a way that the hydrophobic end and hydrophilic head are outward and toward the fibers, respectively, and consequently, the free energy of the paper surface decreases. Additionally, water resistance depends on the reduction in free energy of a paper surface (Krueger and Hodgson 1995; Irvine *et al.* 1999), thus the water absorptiveness of the MT-filled papers was less than that of the other treatments (*i.e.*, T and TP treatments), in spite of its lower ash content.

CONCLUSIONS

1. Due to the chemical modification and the formation of primary N-H amide groups on the particle surface, the composite filler can develop fiber-to-fiber bonding by forming hydrogen bonds with fibers.
2. The impurities of the composite talc significantly decreased, which resulted in an improvement in filler brightness.
3. By adding the composite filler to the sheets, the mechanical properties decreased much less than for that of the other treatments.
4. The optimal filler loading level was 7% and there was no significant difference in the burst and tear indices of the filled and blank papers.
5. The paper opacity increased for all treatments, but brightness improvement occurred only in the MT-filled papers, which also had a relatively high water repellence.

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