

Examination of selected Synthesis parameters for composite adhesive-type Urea-Formaldehyde/activated carbon adhesives

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Abstract— This paper addresses synthesis of activated carbon particles by pyrolysis of wood fibers and their dispersion in urea formaldehyde with an aim to optimize a stable activated carbon particles/urea formaldehyde adhesive. All the adhesive hybrids were characterized with X-ray diffractometry (XRD) and Fourier transform infrared spectroscopy (FTIR), while the dispersion of activated carbon particles was studied with Field Emission scanning electron microscopy (FESEM). Thermo gravimetric analysis (TGA) shows that activated carbon particles have little high effect in the thermal stability of the UF adhesive. It is observed in the TGA graph that the thermal stability of the UF based activated carbon particles is higher than UF only. The scanning micrographs provided confirmation of the smoother surfaces in the UF adhesive made with activated carbon particles. This was attributed to the better encapsulation of activated carbon particles by the polymer matrix.

Index Terms— Wood Fibers, Urea-Formaldehyde, Activated Carbon Particles

I. INTRODUCTION

Urea - formaldehyde adhesives have been widely used by the wood composites industry for over 100 years, as well their concert in the manufacture of composite wood panels, because they have a low cost and high reactivity. Their weakness is lower water resistance and high formaldehyde emissions from wood panel, resulting in lower bond strength amino - methylene it.

To address this problem, this has made such a struggle, to change the method of synthesis using various types of adhesives and additives or hardener, and others [1-3]. The main goal for modern adhesives industry is to produce effective Urea Formaldehyde adhesive with very low formaldehyde emissions. In addition to the drastic reduction in the emission of formaldehyde with a significant increase in resistance Urea Formaldehyde stability bound composite wood can extend applications and markets for these products. To reduce formaldehyde emissions preferably one effective approach is to reduce the molar ratio F/U for adhesives synthesized [4-6]. But this shows that crosslinking is reduced and thus, lower performance of the adhesive, with respect to water resistance and mechanical properties [4]. Several studies have focused on modifying the synthesis parameters Urea Formaldehyde adhesive than lower molar ratio F/U. By controlling parameters such as pH of reaction [7-9], the introduction of a second addition of urea [10] and the use of additives [11, 12].

Additives have an effect on the properties of the renewal adhesive Urea Formaldehyde [13]. The addition of a small quantity of melamine has been used so far in the case of the more challenging applications. The use of other additives, such as formaldehyde catcher also had been tested [14]. Addition trimethoxymethylmelamine and dimethoxymethylmelamine as crosslinking agents available both for resistance [15]. The carbon fiber in Urea Formaldehyde has been reported to improve the mechanical properties of the medium density fibre board [16] and, therefore, need to be studied in depth.

This work aims to analyze the effect of using activated carbon particles in the Urea Formaldehyde adhesive in order to optimize the stable of activated carbon particles/urea formaldehyde adhesives. Therefore, this study investigates the effect of activated carbon particles on thermal behavior of UF adhesive, using TGA. Also, a thorough characterization of the new hybrid was performed using FTIR, FESEM and XRD. The novelty of this work is the use of activated carbon particles in UF adhesive for the first time.

II. EXPERIMENTAL

A. Materials

Urea-formaldehyde (UF) adhesive liquid used for this study were collected from Dynea Malaysia Sdn. Bhd. The viscosity UF resin at 30 °C is 170 centipoises, 8:27 pH, density of 1.286 kg/m³ and gel time at 100 °C is 36s.

B. Activated carbon particles

Activated carbon particles were synthesized by pyrolysis of wood fibers using a furnace in inert conditions at 450 °C for 2 h. Carbon particles were pulverized using a Retsch ZM 200 Chemical Grinder at 18000 rpm for 30 seconds. The carbon content of activated carbon obtained 74.09 % using the system element analysis (CHNS analyzer).

C. UF adhesive/ activated carbon particles hybrids synthesis

The final ratio of urea-formaldehyde (UF) adhesive collected from Dynea Malaysia Sdn. Bhd has an F: U = 1.07. Activated carbon particles added in adhesives Urea Formaldehyde in stage 1, 2.5, 3.5 and 5 % (w / w). The UF / activated carbon particles have a mechanical mixture i.e., stirred for 30 minutes before use. The pure UF resin

samples were named CF -0 while the mixture is named CF - 1, -2.5 CF, CF - 3.5 and CF - 5 each.

D. Fourier Transform Infrared spectroscopy

Hybrid adhesive was studied by FTIR as a partially cured UF/activated carbon particles adhesive in solid form. The partially cured adhesive was prepared by drying the liquid adhesive in an oven at 105 ° C for 2 h. The FTIR transmittance spectra were obtained with a Perkin Elmer Spectrum 1000 - spectrometer in the spectral region 400-4000 cm^{-1} , with a resolution of 2 cm^{-1} and 50 scans. For adhesive in solid samples, KBR pellets with a weight of 1 % from the resulting powder material.

E. Field Emission scanning electron microscopy (FESEM)

Morphological structure prepared samples were investigated in JEOL JSM - 7500F. The sample consists of carbon -coated to provide good conductivity electron beam. Operating conditions have been accelerating voltage of 20 kV; investigate the current 45 nA, and calculate time 60 s.

F. X-ray diffraction (XRD)

X -ray Diffraction (XRD) measurements of soled Urea Formaldehyde adhesive containing activated carbon particles and without activated carbon particles were studied. The X-ray diffraction (XRD) was carried out in an XRD analyzer. The samples were scanned in between 3-80 ° 2 θ at 1deg / min. The spacing between the layers (D002) carbon particles was calculated according to the Bragg equation: $\lambda = 2d \sin\theta$.

G. Thermogravimetric analysis (TGA)

Thermo Gravimetric Analysis (TGA) was used to measure the amount and rate of change in material weight, are important as a function of temperature or time in a controlled atmosphere. This technique can characterize materials that exhibit weight loss or gain due to decomposition, oxidation, or dehydration. The Standard Practice for Thermo gravimetry follows the ASTM 1582 method.

Thermal stability were investigated by non- isothermal thermo gravimetric analysis (TGA) using a TA Instruments. Samples (6 ± 0.2 mg) stored in alumina crucibles. An empty alumina crucible was used as

reference. The samples were heated 30-600 °C in a stream of 50ml/min nitrogen with a heating rate of 10 °C/min.

III. RESULTS AND DISCUSSION

A. Characterization of resins, interactions with activated carbon particles

Urea Formaldehyde adhesive chemical structure can be expressed as a poly (methylene hydroxymethylureas methylene ether which caused by condensation reaction with an aqueous solution of urea formaldehyde. Activated carbon particles have a surface carbon and some hydrogen and nitrogen that can react with the hydroxyl end groups of macromolecules mostly through condensation reactions [17]. In the first step of a side reaction between urea and formaldehyde 1, 3 - bishydroxymethyl (dimethylolurea) urea is produced, which has two hydroxyl groups and can interact with the carbon and make C-OH attraction mode. To confirm the formation of UF/activated carbon particles mixed adhesive, all samples were studied by FTIR spectroscopy and the spectrum is recorded is shown in Fig. 1. diverse and broad peaks in the spectrum CF- 0 resin is caused by entanglement adhesive polymer structure .

In the spectrum of adhesives Urea Formaldehyde, broad peak around 3350-3450 cm^{-1} can be attributed to hydrogen bond O- H and N-H. C- OH attraction mode can be found at 3440 cm^{-1} and 1508 cm^{-1} , respectively. Peak occurs at 1161 cm^{-1} is characteristic C-O is stretching in lactonic, alcohol groups and carboxylate moieties [18]. The peak from 1600 to 1650 cm^{-1} multiples and several overlapping peaks appear in the spectrum of pure Urea Formaldehyde adhesive. This peak is assigned to CO stretching amide I and II, as well as -N-H scissors of the amide I. In the area of 1500-1600 cm^{-1} peak overlap is caused by -N-H bending vibration of the amide II. Multiple peaks at 1460-1470 cm^{-1} can be associated with C-H bending vibration of $\text{CH}_2\text{-N}$ group, while in 1320-1450 cm^{-1} small peak can be attributed to the strain C - N vibrations of amide I and II , he was also appointed for C - H stretching and -O-H bending vibration of alcohol [19] . Strong and broad peak at 1250 cm^{-1} assigned to the C-N stretching vibration of amide II [20]. Peak at 1161 cm^{-1} is caused by two strains of asymmetry N- $\text{CH}_2\text{-N}$ and asymmetric stretching -O-C-C-ether network [21]. The FTIR spectrum and adhesives Urea Formaldehyde UF / CF are shown in Table 1.

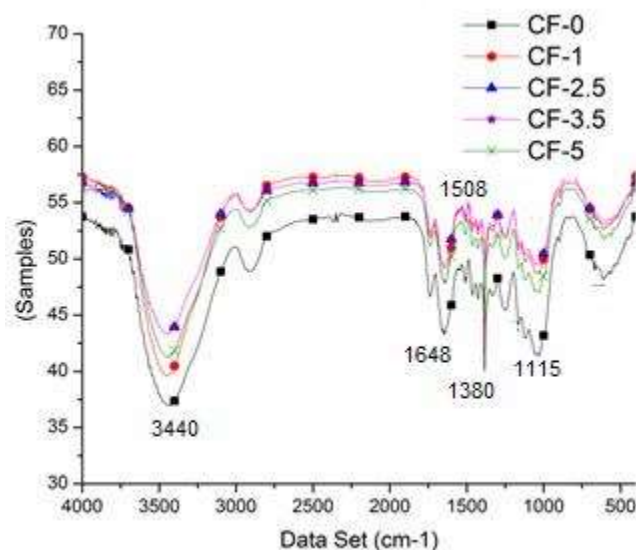


Fig. 1. FTIR transmittance spectra of CF-0, CF-1, CF-2.5, CF-3.5 and CF-5.

Table 1: FTIR peaks observed for UF resin and UF/Activated carbon resin.

Chemical Assignment	UF and UF/Activated carbon resins				
	Wavenumbers/cm ⁻¹				
	CF-0	CF-1	CF-2.5	CF-3.5	CF-5
(NH) 2° amine and OH	3440.34	3441.99	3440.98	3440.97	3441.98
(C=O) in -CONH ₂ (amide I)	1648.01	1639.08	1647.98	1647.99	1640.75
(NH) in NH-CO in 2° amine (amide II)	1508.56	1509	1509.06	1509.08	1509.10
CH- deformation; asymmetric in -CH ₃ -and -CH ₂ -	1460.30	1460.10	1460.11	1460.92	1461.92
(CH) in CH ₂ /CH ₂ OH/N-CH ₂ -N)	1380.42	1380.40	1380.45	1380.43	1380.34
(N-CH ₂ -N), √(C-O-C) of ether linkage	1115.66	1161.92	1161.14	1162.69	1162.69
(H-N-H-CH ₂ -CH ₃) 1° amine	1032.19	1031.66	1032.67	1033.67	1034.17

A spectrum UF/activated carbon particles adhesive shows a strong absorption band between 3446-3448 cm⁻¹ region and 3421 cm⁻¹ for pure samples. It is characteristic of hydrogen bonded NH-NH₂ formed by reaction methylenization during cross [20].

Strong absorption bands observed in the spectrum, near 1648, 1639, 1647, 1647 and 1640 cm⁻¹ for CF-0, CF-1, CF-2.5, CF-3.5 and CF-5, assigned to strain C=O (amide -I) in the group -CONH₂ . Very strong absorption band around 1508.56, 1509, and 1509.08 to 1509.10 for CF-0,

CF-1, CF-2.5, CF-3.5 and C-5, it may be caused by -NH (amide II) is given. Stretching vibration between 1350-1400 cm⁻¹ for sample adhesive, represented by CH bending mode in CH₂/CH₂OH/N-CH₂-N. Absorption band intermediate between 1115-1162 cm⁻¹ may arise due to stretching vibration group -N-CH₂-N of ether linkages.

B. X-ray diffraction (XRD)

The X-ray diffraction (XRD) profiles of all studied adhesive hybrids in powder form are presented in Fig. 2.

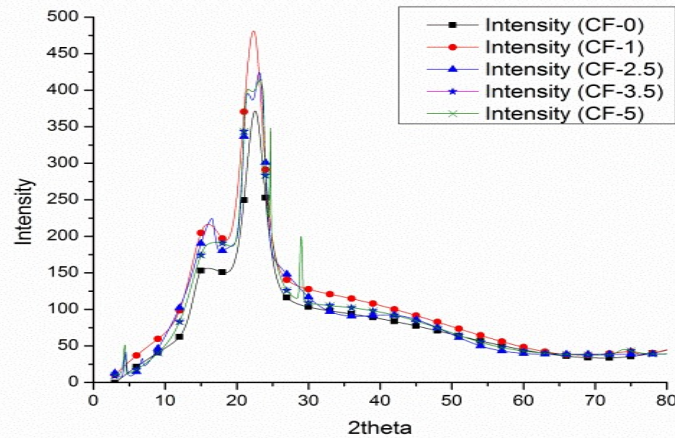


Fig. 2. XRD pattern of UF containing different ratios of activated carbon particles.

All samples XRD patterns confirm that all adhesive hybrids are mainly amorphous with a small degree of order, while the presence of activated carbon particles does not change the appearance of the pattern. Only in CF-1 and CF-3.5 pattern, a peak at 22° seems to be a little wider than in UF-0, which can be caused by the presence of activated carbon particles and a higher degree of amorphisation. In fact, the narrow sharp diffraction peak shows the crystal structure, while the peak width is amorphous structure. XRD pattern of adhesives Urea Formaldehyde containing different ratios of activated carbon particles shows that

changes in the network structure that occurs in the amorphous region of Urea Formaldehyde adhesive.

C. Field Emission Scanning Electron Microscopy (FESEM) Analysis

Adhesive surface morphology was studied by FESEM. All the pictures were taken with an $8000\times$ magnification. In the case of adhesives, some microphotographs of samples CF-0, CF-1, CF-2.5 and CF-5 are presented in Fig. 3.

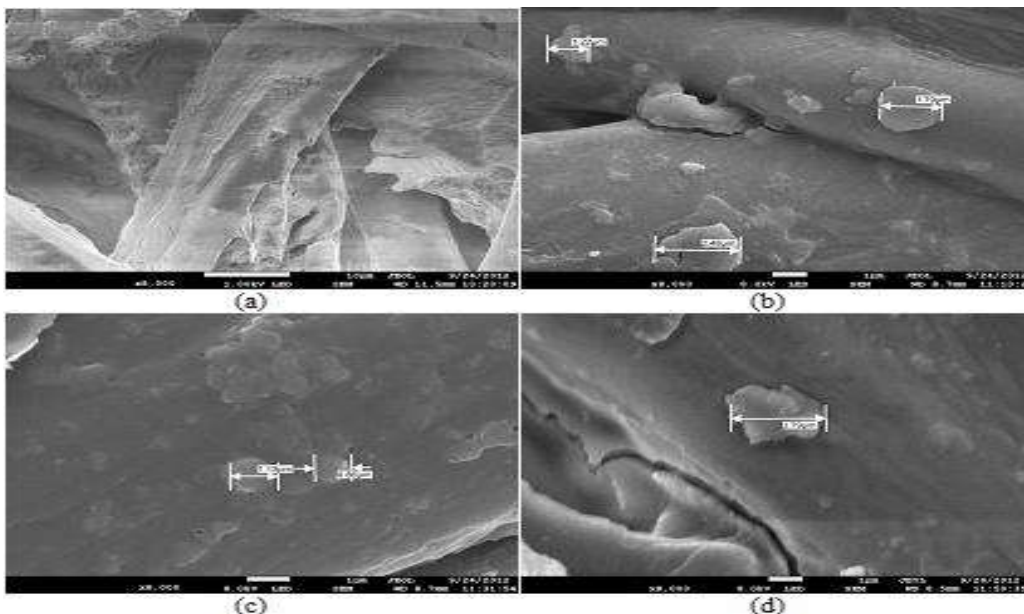


Fig. 3. FESEM microphotographs of (a) CF-0, (b) CF-1, (c) CF-2.5 and (d) CF-5 samples.

These images show that the activated carbon particle concentration increases, a larger amount of light areas appear on the sample. These micrographs clearly show that in comparison to morphological differences compared with the morphology of polymer composites CF-0, CF-1, CF-2.5 and CF-5 (Figure 3a - 3d). It is seen that when Urea Formaldehyde adhesive matrix reinforced with different load activated carbon particles, some morphological

changes occur depending on the bond between the loading of different activated carbon particles and adhesives Urea Formaldehyde. In lower case load of activated carbon particles (1 or 2.5 %) bonding between matrix and reinforcement is higher and when the content of the activated carbon particles are higher loading of lower bond between matrix and reinforcement.

D. Effect of activated carbon particles on the thermal stability of the resins

A carbon material when added to a polymer matrix increases the thermal stability of the polymer [16]. It was

also expected in this study and to evaluate the TGA has been used. In Fig. 4 TGA curves for all samples are presented.

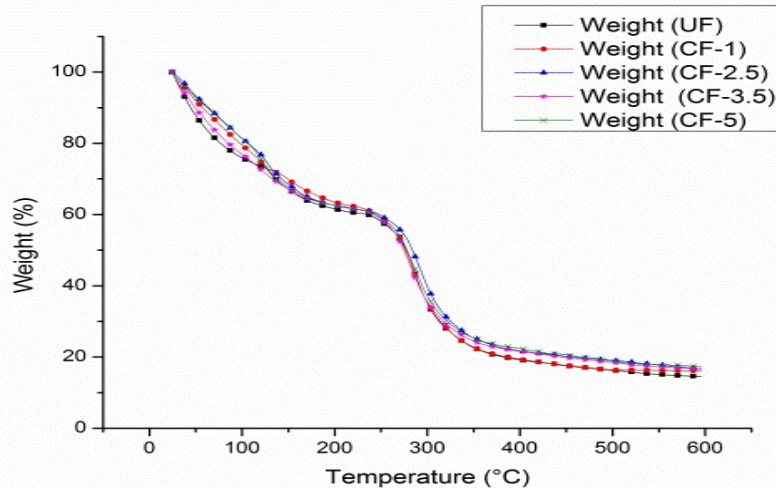


Fig.4. TG curves of all samples: (1) CF-0, (2) CF-1, (3) CF-2.5, (4) CF-3.5, and (5) CF-5.

It is revealed that the addition of activated carbon particles for Urea Formaldehyde adhesives have little effect on thermal stability. From the TGA curves it is clear that the study can be divided into two regions [22]. The first step corresponds to 3-4.5 % loss in mass and it applies to all the samples between 50 and 100 °C. This step corresponds to the evaporation of water from the sample. At a temperature of 100-200 °C slow formaldehyde emissions cause massive losses small in each sample. The main degradation steps initiated above 200 °C, when starting the chain scissions and encourage the formation of radicals formed cyclic structure in the polymer chain. This process leads to solving many of the polymers. Cured adhesive degradation began with the release of formaldehyde from the group dimethylene ether [21] and occurs when the maximum degradation rate of methylene ether stable relationship collapses [22]. Above 200 °C, which compares the mass loss curves for all samples adhesive, it is seen that the hybrids affect the thermal stability of adhesive. The adhesive with a lower content of activated carbon particles have a value intermediate between those CF-1 and CF-2.5, which is slightly higher than UF -0. However, at high temperatures, Urea Formaldehyde adhesive with activated carbon particles have better performance in thermal stability than without particles of activated carbon.

IV. CONCLUSIONS

The purpose of activated carbon particles is added to improve the properties of activated carbon particles/urea formaldehyde adhesives. The degree, which is done, is very dependent on the deployment of additional material into the adhesive. In this work, it is confirmed from FTIR spectroscopy that activated carbon particles can make hydrogen bonds with UF adhesive. However, this is not effective to have a fine dispersion of particles of activated

carbon as individual particles in the polymer matrix and some aggregates are formed. Furthermore, the activated carbon particles as additive in adhesives in UF likely to affect many properties of the hybrid adhesive. TGA graph of this was found that the thermal stability of UF based activated carbon particles is higher than the pure UF. Scanning micrographs provided evidence of the smooth surface at UF with activated carbon particles.

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