

Synthesis of Microcrystalline Cellulose—Polyvinyl Alcohol Stabilized Polyvinyl Acetate Emulsion

Ravindra V. Gadhave , S. K. Vineeth 

Department of Polymer and Surface Engineering, Institute of Chemical Technology, Mumbai, India
Email: ravi.gadhav3@gmail.com, vineetheskay@gmail.com

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Abstract

Polyvinyl alcohol (PVA) colloid stabilized Polyvinyl acetate (PVAc) based wood adhesive has poor performance in highly humid conditions. Currently, the addition of natural fillers in the wood adhesive is one of the most effective ways to enhance the performance of PVAc wood adhesive in highly moist conditions. Microcrystalline cellulose (MCC) are strong renewable, bio-based material and has great potential in a reinforcement of the polymeric matrix. Hence, the present work investigates the applicability of microcrystalline cellulose incorporated 3% and 5% in situ emulsion polymerization PVAc wood adhesives. Effect on physical, thermal and mechanical properties was studied by viscosity, pH, contact angle measurement, differential scanning calorimetry (DSC) and pencil hardness test of films. Emulsions with different proportions of MCC were prepared and the shear strength of the applied adhesive on wood was measured. The viscosity of the adhesives was increased by increasing the concentration of MCC. The mechanical properties like tensile strength of adhesives with MCC were measured by universal tensile machine (UTM). Thermal stability was studied by differential scanning calorimetry (DSC). The tensile shear strength demonstrates that MCC can improve bonding strength as compared to PVAc Homo based adhesive in the wet condition which was validated through a contact angle study. The hardness of PVAc films were also changed positively by the addition of MCC. Here, we studied the effect of the addition of different concentrations of MCC materials in situ polymerization of PVAc on their performance properties.

Keywords

Wood Adhesive, Polyvinyl Acetate, Microcrystalline Cellulose, Bio-Based, Emulsion

1. Introduction

Adhesives play an essential role in the wood joinery industry. Wood adhesives can be classified according to their origin in natural and synthetic [1] [2]. Natural wood adhesives can be of vegetable origin or proteins of animals, while synthetic ones have hydrocarbon raw material [3] [4] [5] [6] [7]. The replacement of hydrocarbons with natural polymers has gained much importance in this aspect.

Traditionally, polyvinyl alcohol (PVA) is employed as a protective colloid in polyvinyl acetate (PVAc) wood adhesive [8]. PVAc is a yellow-white adhesive that can be applied directly to the wood substrate and cured at room temperature or through high radio frequency. After curing this adhesive exhibit high mechanical properties; however, its use is not suggested in environments with high temperatures and high relative humidity [9] [10]. PVAc-based adhesives offer numerous advantages, such as economic, good adhesion to a wood substrate, easy processing with a simple mixing method and excellent stability [11] [12]. PVAc adhesive is used for any and all wood gluing processes. Major areas include bonding of finger-jointing, laminating and assembling [13] [14] [15]. It is also widely used in the formulation of waterborne adhesive for porous materials including paper, board, textile, ceramic, foils and the like [16] [17].

It has poor water and heat resistance. Although modifications in the chemistry of wood adhesives are a path of optimization of performance properties, the addition of filler presents a possible alternative route of modification [18] [19] [20]. Another alternative to alleviate the shortcoming of PVAc adhesive is the use of nanoparticles such as carbon nanotube [21] on physico-mechanical properties of PVAc, clay [22] to improve the bonding strength of PVAc, graphene [23] to improve PVAc strength and toughness, cellulose [24] to see the effects of filler on thermal stability [22] to improve the performance of PVAc at elevated temperature and high humid condition. These particles were shown to have a beneficial effect on the properties of PVAc wood adhesives [25].

Bio-fillers are among the most studied materials in the scientific world due to their renewability, non-toxicity and biodegradability and ease of availability. Starch provides an option to replace PVA for stabilizing PVAc. Like the structure of starch, cellulose is a member of the polysaccharide family; it is also expected to function in the same manner. Cellulose has inherent binding properties but is too weak to bond with the substrates. Hence cellulose is a potential material in adhesive industry with the added advantage of renewability and biodegradability [26]. Cellulose is composed of a linear polysaccharide chain consisting of repeated β -(1 \rightarrow 4)-D-glucopyranose units [27]. The crystallinity of cellulose is lower due to the presence of amorphous regions. The hydroxyl group on these materials allows potential hydrogen bonding and surface modification [28]. There has been great interest in bio-based binders and various cellulosic materials for application in the wood adhesive field [29]-[35].

Recent studies have focused on the reinforcement of adhesives with micro crystalline cellulose (MCC) to improve their mechanical properties. PVAc adhe-

sive was reinforced with MCC at different concentrations (3%, and 5%) and then shear strength tests were conducted using the universal testing machine (UTM). The MCC improved the bonding strength of PVAc adhesive in dry and wet conditions. The improvement was measured in terms of wood failure percentage in dry conditions and shear strength in wet conditions at room temperatures. In this study, PVAc adhesive was with reinforced MCC with an aim to assess the comparative performance of PVA stabilized PVAc and MCC-PVA stabilized PVAc adhesive. The performance of reinforced PVAc adhesives was analyzed by testing the shear strength of wood joints bonded with these adhesives and water resistance was analyzed by using a water contact angle study.

2. Experimental

2.1. Materials

Partially hydrolyzed polyvinyl alcohol (PH PVA) (CAS No. 9002-89-5, % degree of hydrolysis 87% - 89%, Molecular weight 99,000 - 105,000 g/mol) was obtained from Kuraray Cooperative Limited, India. Vinyl acetate (VAc, CAS No. 108-05-4), sodium lauryl sulfate (SLS, CAS No. 68585-34-2), sodium bicarbonate (SBC, CAS No. 144-55-8), potassium persulfate (PPS, CAS No. 7727-21-1), 2-ethyl hexanol (2 EH, CAS No. 104-76-7) and Isothiazolinones preservative (CAS No. 1003-07-2), Microcrystalline cellulose (MCC, pH 6.4, Bulk density (untapped 0.3 g/cm³, CAS No. 9004-34-6) were acquired from Sigma Aldrich.

2.2. Preparation Method

Polymer emulsions were synthesized in a flat-bottom glass reactor equipped with condenser, stirrer, temperature indicator, metric pump and glass funnel for the addition of additives. Semi-continuous emulsion polymerization process was adopted with continuous supplying thermal energy using a water heating bath. **Table 1** shows the polymer emulsion recipes.

2.3. Colloid Solution Preparation

PVA was first put into a glass reactor. The water was added to them, and the mixture was kept for continuous stirring at 175 rpm. The temperature was slowly raised up to 94°C - 96°C and the final temperature was kept at 94°C - 96°C. The cooking was carried out for 2.5 hr. Then, the solution was cooled down to 25°C and added isothiazolinones preservative. It is used in PVAc Homo.

2.4. PVA-MCC Solution Preparation

PVA and MCC were first put into a glass reactor. The water was added to them, and the mixture was kept for continuous stirring at 175 rpm. The temperature was slowly raised up to 94°C - 96°C and the final temperature was kept at 94°C - 96°C. The cooking was carried out for 2.5 hr. Then, the solution was cooled down to 25°C and added isothiazolinones preservative.

Table 1. Composition of the emulsions.

Components	PVAC Homo	MCC-01	MCC-02
	Weight % (%)	Weight % (%)	Weight % (%)
Vinyl acetate monomer (VAc)	46.6	43.6	41.6
MCC	0	3	5
DIW	45.11	45.11	45.11
PH PVA	2.63	2.63	2.63
SBC	0.11	0.11	0.11
SLS	0.01	0.01	0.01
2-Ethyl hexanol	0.01	0.01	0.01
DIW	1.53	1.53	1.53
PSS	0.06	0.06	0.06
DIW	1.2	1.2	1.2
PPS	0.04	0.04	0.04
DIW	2.3	2.3	2.3
Sodium benzoate	0.1	0.1	0.1
DIW	0.15	0.15	0.15
Isothiazolinones Preservative	0.15	0.15	0.15
Total	100	100	100

2.5. PVA Stabilized Emulsion (PVAc Homo)

PH PVA solution was used as a protective colloid for the stabilization of the emulsion. Sodium lauryl sulphate was used as an anionic surfactant. Sodium bicarbonate used as buffer. The initiator solution was prepared by dissolving potassium persulfate in deionised water. 2 ethyl hexanol was used to control foam generation in batch. Polymerization was carried out by controlled addition of vinyl acetate (VAM) monomer. After the complete addition of monomer, potassium persulfate solution in water was added and the temperature was maintained at 90°C for 1 hr. Then, the emulsion was cooled down to 25°C and added Isothiazolinones preservative.

2.6. PVA and MCC Stabilized Emulsion (MCC-01 and MCC-02)

PH PVA + MCC solution was used as a protective colloid for stabilization of the emulsion. Sodium lauryl sulphate was used as an anionic surfactant. Sodium bicarbonate used as buffer. Initiator solution was prepared by dissolving potassium persulfate in Deionised water. 2 ethyl hexanol defoamer was used to control foam generation in batch. Polymerization was carried out by controlled addition of monomer VAM. After the complete addition of monomer, potassium persulfate solution in water was added and temperature was maintained at 90°C

for 1 hr. Then, the emulsion was cooled down to 25°C and added isothiazolones preservative.

3. Characterization and Testing

3.1. Solid Content

The solids content of emulsion was checked by heating sample (around 1 g) at 120°C for 120 minutes in a hot air oven. Measurements were done in triplicate to minimize error. To measure the solid content the following equation was used,

$$\text{Solid content (\%)} = (W_3 - W_2) / (W_2 - W_1) \times 100\%$$

W_1 = Weight of empty weighing pan.

W_2 = Weight of Pan with sample before heating.

W_3 = Weight of pan with residue after drying till constant weight.

3.2. Viscosity and pH

A Brookfield Viscometer RVT was used for calculating the viscosities of the samples. The values were taken at 20 rpm and at a temperature of 30°C. The pH of the samples was analysed using a digital pH meter, CL 54 + Toshcon Industries, India. To reduce the error, viscosity and pH measurements were taken 5 times.

3.3. Pencil Hardness Test

The pencil hardness test was done using a QHQ-A portable pencil hardness tester, having lead pencils of varying hardness numbers. Using an applicator, 200-micron films were cast over a glass plate. After the films are dried at 28°C for 1 day, the test was conducted following ASTM D 3363 standard.

3.4. Water Contact Angle Measurement

The water contact angle was measured using a Rame-Hart Goniometer, Germany at 28°C and 60% relative humidity. To measure the contact angle, the samples are cast over glass substrates with an applicator of 100 micrometers. The test was conducted 5 times to reduce the error. Contact angle measurement gives an idea of the behaviour of prepared samples with respect to water. Moreover, variations in the hydrophobicity in formulations can be analysed using contact angle measurements.

3.5. Tensile Shear Strength-Wet and Dry

Shear strength was evaluated at different intervals in joints prepared by gluing beech specimens in a normal climate. Bonded assemblies were tested after keeping them at 23°C and 60% relative humidity after 4 hr, 6 hr, 24 hr and 7 days. In this case, strength was calculated in MPa. Wet strength tests were performed according to condition D3-3 of EN 205 after 7 days in a normal climate; specimens were soaked in water for an additional 24 hr and then tested water resistance.

4. Results and Discussions

4.1. Solid Content

The solids content of the emulsion was checked by heating the sample (around 1 g) at 120°C for 120 min in the oven. It was calculated that the solid for both the samples are approximately the same (**Table 2**).

4.2. Viscosity and pH

The viscosity and pH of the prepared samples are shown in **Table 2**. The presence of MCC in PVAc Homo (MCC-01 and MCC-02) sample enhances the pH and makes the system less acidic in nature as compared to PVAc Homo. Compared to the PVAc Homo, increase in viscosity was observed in with increase in the concentration of MCC in PVAc Homo. This is due to the grafting of VAc monomer on hydroxyl group of cellulose and resulted in an increased in viscosity as compared to PVAc Homo.

4.3. Pencil Hardness of Film

The pencil hardness of emulsion polymer film evaluates the mechanical strength property. With the addition of MCC in PVAc Homo (MCC-01 and MCC-02), hardness of the film was increased as compared to PVAc Homo as shown in **Table 2**. Due to grafting of VAc on MCC resulted in a dense structure. Slip from sharp pencil due to needle like structure increased, hence the MCC-01 and MCC-02 samples showed higher values of pencil hardness.

4.4. Water Contact Angle

In general water contact angle correlates with the water resistance properties of the samples. Moreover, to correlate with the water resistance properties of MCC-01

Table 2. Various test results.

Sr. No	Properties	PVAc Homo	MCC-01	MCC-02
1	% Solid	47.40 ± 0.1	47.50 ± 0.1	47.50 ± 0.1
2	Viscosity (Poise)	400 ± 10	620 ± 10	660 ± 10
3	pH	4.90 ± 0.4	5.05 ± 0.4	5.05 ± 0.4
4	Film Hardness	HB	1H	2H
5	Contact angle	45.7 ± 1.39	65.0 ± 1.39	68.0 ± 1.39
	Tensile share strength (Dry) (MPa)			
6	6 hr	2.27 ± 0.4	1.90 ± 0.4	3.80 ± 0.4
	24 hr	5.70 ± 0.4	5.80 ± 0.4	6.70 ± 0.4
	7 Days	6.20 ± 0.4	6.50 ± 0.4	6.80 ± 0.4
7	Wet strength (7 days + 1 day)	1.27 ± 0.3	2.58 ± 0.4	2.73 ± 0.4

and MCC-02 with the PVAc Homo, water contact angle measurement was studied. Distilled water is made in contact with the emulsion samples casted over glass slides and contact angle values were noted. The contact angle of the sample with water is shown in **Figure 1** and the values are labelled in **Table 2**. It is observed that the contact angle value is higher for MCC-01 and MCC-02 as compared to PVAc Homo. Slight reduction in strength of bonded wood specimen in wet condition in PVAc Homo provides evidence for the same observation (Reason for water resistance).

4.5. Tensile Shear Strength Testing-Wet and Dry

The bonding strength of adhesives was tested on the substrates in dry condition and another set of samples in wet conditions as per EN 204 for water resistance. From **Table 2**, it was observed that by the addition of MCC in PVAc Homo CC, water resistance was increased. After 24 hr immersion in water, PVAc Homo has lower strength as compared to MCC-01 and MCC-02 as shown in **Figure 2**. This is due to MCC fibers which act as a barrier for water or moisture through adhesive which was validated in contact angle testing. In addition to MCC, dry strength increased in MCC-01 and MCC-02 as compared to PVAc Homo this was due to cellulosic hydroxyl having a high affinity towards wood cellulose due to secondary force which resulted in a strong bond on wood substrate. Dry strength was shown in **Figure 3**.

5. Conclusion

Cellulose reinforced PVAc adhesives were prepared by mixing MC at different concentrations of 1%, 3%, and 5% during in situ polymerization of PVAc adhesive. The addition of cellulose to PVAc adhesive improved the elastic stiffness of the joints as well as the bond quality. The addition of cellulose in colloid solution improved the mechanical properties. The bonding ability of cellulose reinforced PVAc adhesives is evident from the increased proportion of wood failure. Water resistance was enhanced in PVAc adhesives with cellulose as compared to PVAc Homopolymer based adhesive which was confirmed by a contact angle study. The cellulose added to in situ emulsion polymerization of PVAc improved the water

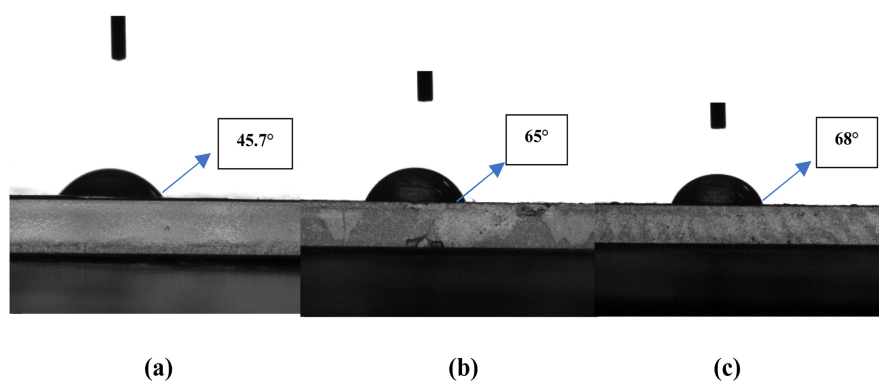


Figure 1. Water contact angle images for (a) PVAc Homo (b) MCC-01 (c) MCC-02.

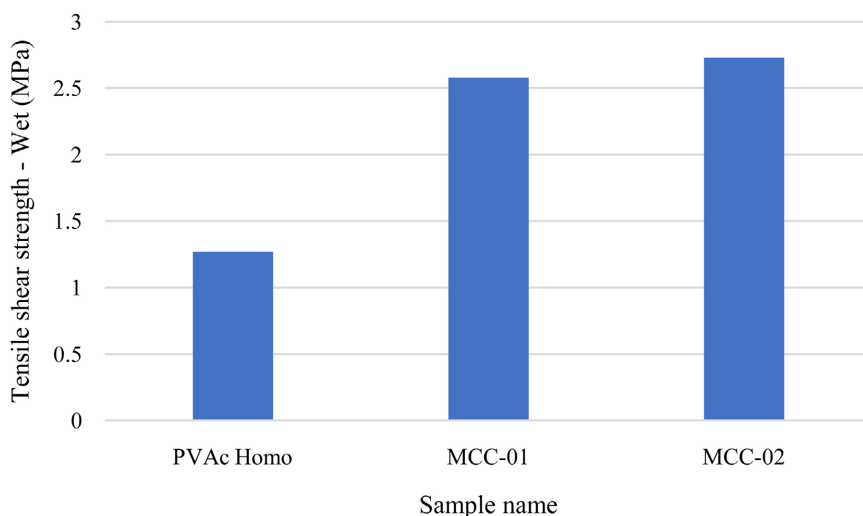


Figure 2. Tensile shear strength in wet conditions.

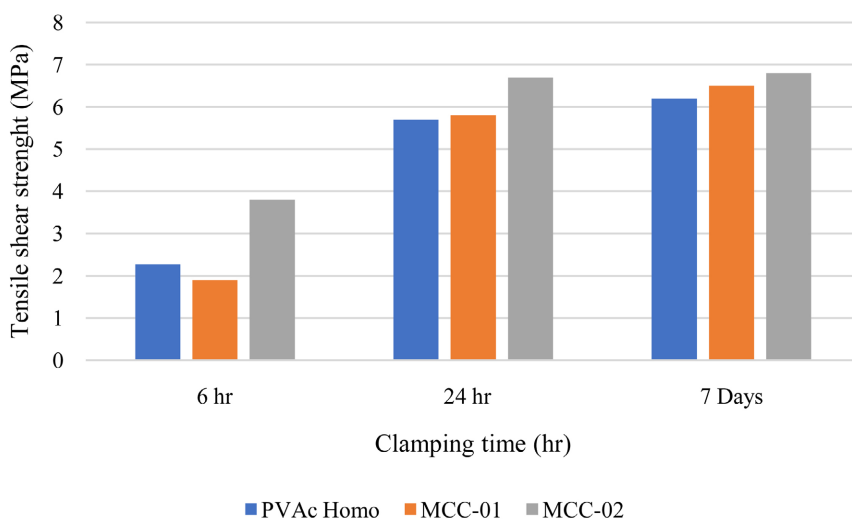


Figure 3. Tensile shear strength in dry condition.

resistance and mechanical properties of adhesives as compared to virgin PVAc Homo. Cellulose Stabilized PVAc based adhesives are mainly used in wood bonding applications due to the binding property with room temperature curing, low toxicity, chemical resistance, and sustainability.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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