Preparation and Antifungal Activities of Microcapsules of Neem Extract Used in *Populus tomentosa* Deteriorated by Three Mold Fungi

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Microcapsules of neem extract (MNE) were observed using an optical microscope (OM) and scanning electron microscope (SEM). The antifungal activity of the extract was evaluated by an agar diffusion assay. The MNE were induced into the wood material by a full-cell process. The diameters of the microcapsules were measured by OM, and the distribution of microcapsules in wood was observed by SEM. Wood blocks of *Populus tomentosa* were treated with the MNE, neem extract (NE), and an acid mixture of melamine formaldehyde resin (MF) and sodium dodecyl sulfate (AMS); their antifungal properties against *Penicillium citrinum*, *Trichoderma virens,* and *Aspergillus niger* were visually assessed. The microcapsules prepared by MF, 1% sodium dodecyl sulfate (SDS), and 10% NH_4Cl showed regular shape and good dispersion. The agar diffusion assay showed that the neem extract had significant inhibition against all tested fungi, and the optimum concentration of NE was 10%. The diameters of the microcapsules were normally distributed in a range of 0.4 μm to 4 μm, and the microcapsules were unevenly distributed in the vessels and surface of *Populus tomentosa*. Wood specimens treated with MNE observed complete inhibition to all studied fungi, and the mark grades of specimens treated with MNE against three fungi all reached 5 (no growth of fungi).

Keywords: Antifungal activities; Microcapsules; Neem; Extract; Populus tomentosa

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INTRODUCTION

Poplar is widely distributed in the central plains of China. *Populus tomentosa* is one of the most widespread species in this family and has the potential to be a source of fuel ethanol (Jin *et al*. 1988; Wang *et al*. 2012). However, *P. tomentosa* has poor decay resistance, and that limits its service life (Ge *et al*. 2017).

Molds use wood carbohydrates to sustain mycelia or spore growth and form flocculent or speckled stains on wood surfaces, causing discoloration, and they can grow more easily on wood with a high content of moisture (Zabel and Morrell 1992; Nasser *et al*. 2017). Some typical molds such as *Penicillium citrinum, Trichoderma virens,* and *Aspergillus niger* can cause economic loss (Kositchaiyong *et al*. 2014b). Mildew usually occurs on wood surfaces and does not destroy the wood structure. However, molds can affect the appearance of wood. Cellulose and lignin are degraded when wood is exposed to mold for a long time, which increases the permeability of liquid and promotes wood staining (Zu and Huang 1987; Duan 2005). Many conventional preservatives such as chromated copper arsenate (CCA) are harmful to people and the environment (Gao *et al*.

2005). Therefore, the development of an eco-friendly and sustainable anti-mildew agent for wood is urgent.

Natural extracts contain compounds that protect wood materials against mold and destructive fungi (Damjan *et al*. 2006; Bento *et al*. 2014). The secondary metabolites of plants have antimicrobial potential and insect resistance (Xu *et al*. 2011). Neem (*Azadirachta indica* A. Juss) is one of the most respected trees in India. Different parts of neem extract, including flowers, leaves, seeds, and bark, have shown great antibacterial property and biological activity (Gupta *et al*. 2017). Neem produces a variety of chemicals that protect against wood-decay fungi; azadirchtin-A, nimbin, and salannin are the major triterpenoids that have bactericidal capacity (Dhyani *et al*. 2004; Ali *et al*. 2017). However, botanical extracts are sensitive to rain, hyperthermia, and UV radiation so that the biocides are easily leached from wood. Therefore, it is necessary to develop new formulation techniques that reduce biocide leaching yet are still eco-friendly.

Microencapsulation is the technology of coating solid and liquid into the form of tiny particles using film-forming materials. Microcapsules have many advantages over conventional plant extracts, and encapsulation can reduce the release of agents and protect agents against leaching and UV-induced degradation (Jämsä *et al*. 2013). In addition, wood biocides with poor water-solubility are easier to introduce into wood by encapsulation (Liu *et al*. 2002). Microencapsulation has been widely used in medical science (Kumar *et al*. 2011), food technology (Silva *et al*. 2014), and agriculture (Jiang *et al*. 2008). However, microencapsulation used in wood preservation has been less reported. In the present study, a method was developed for the encapsulation of neem seed extract. The microcapsules were characterized in terms of appearance, particle size distribution, and antifungal effects against *Aspergillus niger*, *Trichoderma virens*, and *Penicillium citrinum*. To the authors' knowledge, this is the first report of a plant-based microencapsulation applied to wood preservation.

EXPERIMENTAL

Materials

Neem seeds were purchased from Kunming, Yunnan province in China in July, 2017. Ethanol, dimethyl sulfoxide (DMSO), polyoxyethylene (20) sorbitan monooleate (Tween 80), sodium dodecyl sulfate (SDS), NH_4Cl, and acetic acid were bought from Fusite Technology Co., Haerbin, China. The chemicals were of analytical grade. Melamine formaldehyde (MF) resin and urea formaldehyde resin (UF) were bought from Hongming Chemical Co., Jinan, China. Sapwood samples of *P. tomentosa* were obtained from a woodworking factory in Haerbin, China. The wood samples (20 mm × 20 mm × 5 mm) were dried at 105 °C to a constant weight and then autoclaved at 121 °C for 20 min. Three common mold fungi—*P. citrinum*, *T. virens*, and *A. niger*—were provided by the College of Engineering and Technology at Northeast Forestry University, Harbin, China. All abbreviations for materials used in this paper are shown in Table 1.

Preparation of Neem Extract

Neem seeds were washed and ground to 20 mesh after being air-dried at room temperature. Approximately 20 g of neem powder was soaked in 280 mL of ethanol (60%). The container was shaken in a 50 °C water bath for 90 min. The solvent was evaporated by a rotary evaporator (Yarong Instrument Co., Shanghai, China).

Preparation of Microcapsules of Neem Extract

The surfactants (polyoxyethylene (20) sorbitan monooleate and SDS) were diluted to 0.5%, 1%, and 2% in distilled water, and 20 g of neem extract was added. The emulsion was prepared using a stirrer (Xinrui Instrument Co., Jiangsu, China) at 1500 rpm at room temperature. The encapsulation of neem extract was carried out in a 1-L beaker *via in-situ* polymerization. First, 60 g of pre-polymers (MF and UF) were added into the emulsion, respectively. And the pH of the emulsion was adjusted to 5.5 using 10% acidifying agents (NH_4Cl and acetic acid) for 3 h at 50 °C, respectively.

Table 1. Abbreviations for Materials Used

Full Name	Abbreviation
Microcapsules of Neem Extract	MNE
Neem Extract	NE
Acid Mixture of Melamine Formaldehyde Resin and Sodium	AMS
Potato Dextrose Agar	PDA
Melamine Formaldehyde Resin	MF
Urea Formaldehyde Resin	UF
Sodium Dodecyl Sulfate	SDS
Optical Microscope	OM
Scanning Electron Microscope	SEM
Dimethyl Sulfoxide	DMSO

Evaluation of Microcapsules

The distribution and size of the microcapsules were observed using an optical microscope (OM, BX53, Laishi Electronic Technology Co. LTD, Shanghai, China). Sizes of microcapsules were measured by 0.01-mm micrometer of OM, and the amount of microcapsules was more than 500. The morphology of microcapsules and their distribution in wood material were examined by SEM (Quanta 200, FEI Co., Hillsboro, OR, USA). The condition of SEM was 2nd electron detection mode, under 5 kV of accelerating voltage, gold sputter coating.

Agar Diffusion Assay

Approximately 15 mL of potato dextrose agar (PDA) was poured into each Petri dish, and the fungal spore suspension was spread evenly on the agar with sterilized cotton swabs. Five holes were punched in each dish with a 5-mm hole punch. Different concentrations of extract solution were added into the holes with a pipette. Neem extract was prepared at different concentrations of 15%, 12.5%, 10%, 7.5%, 5%, and 2.5% by diluting the respective amount of extract in 40% ethanol and 0.5% DMSO. A solution of 40% ethanol and 0.5% DMSO was used for the control group. The inhibitory zone diameters were measured with vernier caliper *via* intersection method.

Wood Treated by Tested Reagents

Wood blocks were autoclaved at 121 °C for 20 min to be sterilized. Samples were treated under 0.1 MPa, full-cell pressure vacuum for 60 min (Xu *et al*. 2013). Different solutions (MNE, NE, and AMS) were induced into wood samples for 10 h, and then samples were air-dried for 24 h (Salem *et al*. 2016).

Biodeterioration of Wood by Mold Fungi

Mold fungi 15 day-old PDA cultures were prepared. The wood specimens were inoculated with a 5-mm disc of each fungus in a petri dish after treatment by MNE, NE, and AMS. Each dish contained 15 mL of PDA and was incubated for five days at 26 ± 1 °C (Kositchaiyong *et al.* 2014a). Three replicates were used for each solution, and untreated wood specimens were used for control samples. Antifungal properties were evaluated by fungal growth retardation after 14-days observation, using the visually determined marks recommended by Humar and Pohleven (2005), as shown in Table 2.

Table 2. Fungal Growth Retardation Marks

Mark	Growth of Mycelium
0	mycelium growth more intense than control
1	normal growth, insignificant retardation (area of colony ≥ 90% of area of controls)
2	visible signs of retardation (colony < 90% and > 60% of controls)
3	pronounced retardation (colony < 60% and >25% of controls)
4	very marked retardation (colony < 25% of controls)
5	no growth

RESULTS AND DISCUSSION

Effects of Wall Materials on Preparation of Microcapsules

Microcapsules with different wall materials have different permeability and compactness; appropriate wall materials can improve the appearance and coating effect of microcapsules (Wang *et al.* 2006). The microcapsules prepared by MF and UF are shown in Fig. 1. The surfactant and acidifying agent were 1% polyoxyethylene (20) sorbitan monooleate and 10% NH_4Cl, respectively.

The microcapsules prepared with MF had a regular morphology and were densely distributed (Fig. 1a). The size of microcapsules prepared by UF was not uniform (Fig. 1b). UF contains carbonamide group, which is easy to hydrolyze, and the molecule contains hydroxymethyl groups, carboxyl groups, amino groups, ether bonds, and other hydrophilic groups so that UF has poor water resistance (Zhang *et al*. 2009). Melamine in MF can react with hydroxymethyl groups and amino groups, which reduces the number of hydrophilic groups and increases the hydrophobicity of MF, so MF has good water resistance (Lee *et al*. 2002; Shi and Cai 2006). Besides, MF has both heat resistance and chemical resistance. Therefore, it would be a better choice as the wall material.

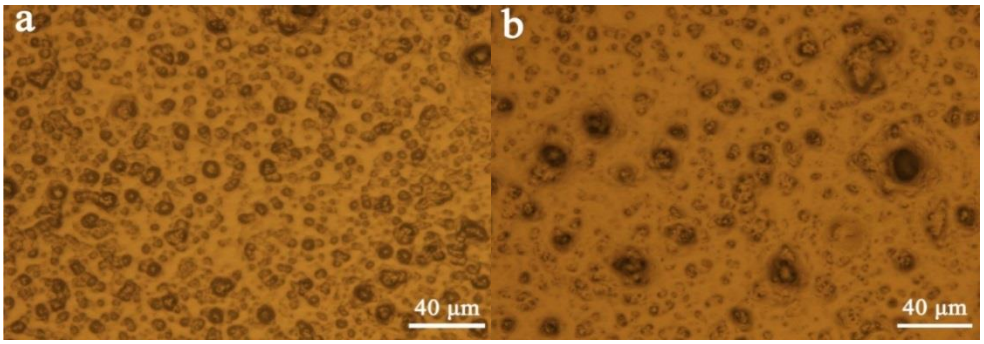

Fig. 1. Microcapsules prepared with (a) MF and (b) UF

Effects of Surfactants on Preparation of Microcapsules

The formation of surfactant/prepolymer complexes can change the adsorption layer around the oil phase, which improves the stability of emulsion and promotes the formation of an outer membrane (Petrovic *et al*. 2010). However, the type and dosage of surfactants can have a significant influence on the particle size and wall thickness of microcapsules. Therefore, an appropriate surfactant is important for the formation of microcapsules (Chao 1993). Using MF and 10% NH_4Cl as wall material and acidifying agent, respectively, the microcapsules prepared by polyoxyethylene (20) sorbitan monooleate and SDS are shown in Table 3 and Fig. 2. Compared with the microcapsules prepared by polyoxyethylene (20) sorbitan monooleate, microcapsules prepared with SDS had better dispersion as a whole. As the concentration of surfactants was increased, the microcapsules became denser. The morphology of microcapsules was irregular, and the dispersion was not uniform using 0.5% concentration of both surfactants (Fig. 2a, d). When the concentration reached 2%, the microcapsules all became aggregated seriously (Fig. 2c, f). In addition, the dispersion of microcapsules with 1% SDS was uniform compared with microcapsules with 1% polyoxyethylene (20) sorbitan monooleate (Fig. 2b, e). The micelles were less when using surfactants at low concentration, so the microcapsules were less and irregular. Besides, the microcapsules were overabundant and easily aggregated when using excessive concentration of surfactants. The microcapsules with 1% SDS had a better appearance and narrower particle size distribution, while microcapsules with polyoxyethylene (20) sorbitan monooleate tended to agglomerate and break. Because SDS is more hydrophilic than polyoxyethylene (20) sorbitan monooleate, SDS can promote the formation of smooth outer wall on microcapsules.

Table 3. Effects of Surfactants on the Distribution of Microcapsules

Dosage of Surfactants (%)	Types of Surfactants	
	Tween 80 *	SDS
0.5	Poor encapsulation	Poor encapsulation
1	Agglomeration	Good Dispersibility and Appearance
2	Agglomeration	Agglomeration

* polyoxyethylene (20) sorbitan monooleate

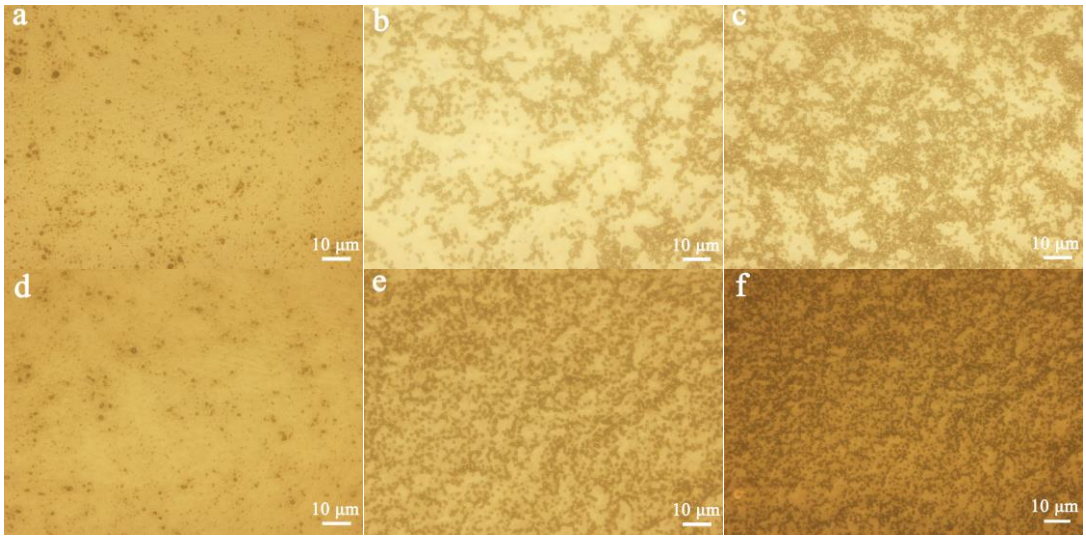

Fig. 2. Microcapsules prepared with (a) 0.5% polyoxyethylene (20) sorbitan monooleate (Tween 80), (b) 1% Tween 80, (c) 2%Tween 80, (d) 0.5% SDS, (e) 1% SDS, and (f) 2% SDS

Effects of Acidifying Agents on Preparation of Microcapsules

The acidifier promotes the chain-end polymerization of MF, forming the outer network structure of microcapsules. Microcapsules prepared with 10% acetic acid and 10% NH_4Cl are shown in Fig. 3. The microcapsules were broken down and agglomerated using 10% acetic acid, while the microcapsules using 10% NH_4Cl possessed a smooth appearance and were finely dispersed. Acetic acid can dramatically improve the crosslinking reaction speed, which makes the pre-polymers quickly reduce the reactivity and limit the maximum of conversion efficiency (Ye *et al*. 2006). Thus, the crosslink density network has low molecular weight, and microcapsules are easily broken using acetic acid. As for NH_4Cl, the pH is more easily adjusted to a lower value, and NH_4Cl can consume free formaldehyde and act as a curing agent. Therefore, 10% NH_4Cl was chosen as the acidifying agent.

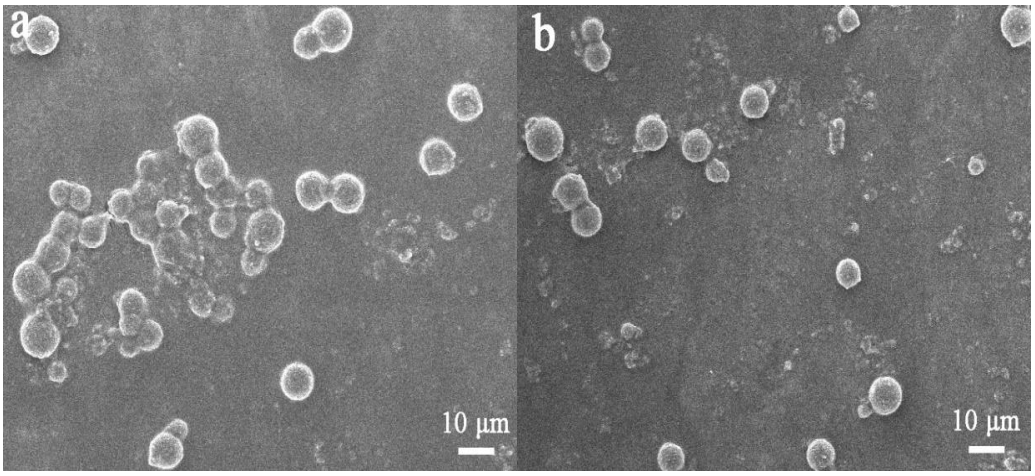

Fig. 3. Microcapsules prepared by (a) 10% acetic acid and (b) 10% NH_4Cl

Size of Microcapsules and Distribution in Wood

Microcapsules were prepared by MF, 1% SDS, and 10% NH_4Cl according to the preliminary optimization experiments. The particle size was normally distributed in a range of 0.4 μm to 4 μm (Fig. 4), and the mean size was 1.97 μm.

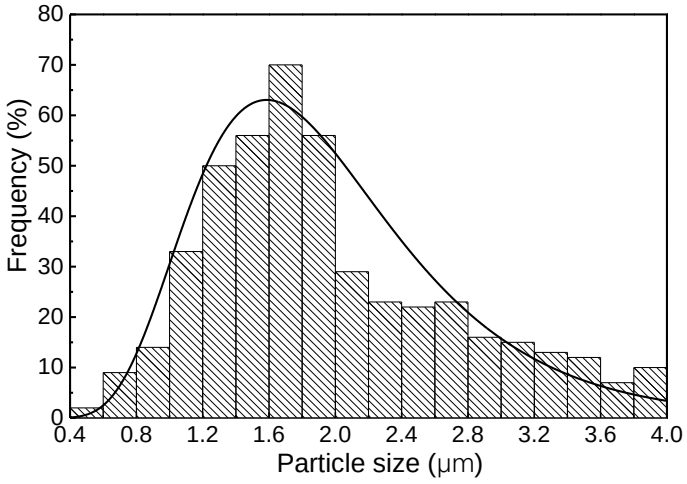

Fig. 4. Size distribution of microcapsules of neem extract

The main function of vessels in wood is translocating water and inorganic salt. The vessels diameters of hard wood range from 16 to 500 μm (Li 2002). Microcapsules can be induced into the wood blocks. Thus, the size of the microcapsules prepared in this work was feasible.

Microcapsules were injected into wood material by "full-cell process", and tangential and transverse sections were observed by SEM. Microcapsules were attached stably to the wood vessels. The wood vessels were filled with microcapsules, which were round in shape and smooth in appearance (Fig. 5a, b). Additionally, microcapsules were observed on the surface of wood specimens (Fig. 5c, d). However, because the size and distribution of microcapsules were not homogenous, it is expected that hypha can grow on some areas of wood after a long period of time (Flemming and Wingender 2010).

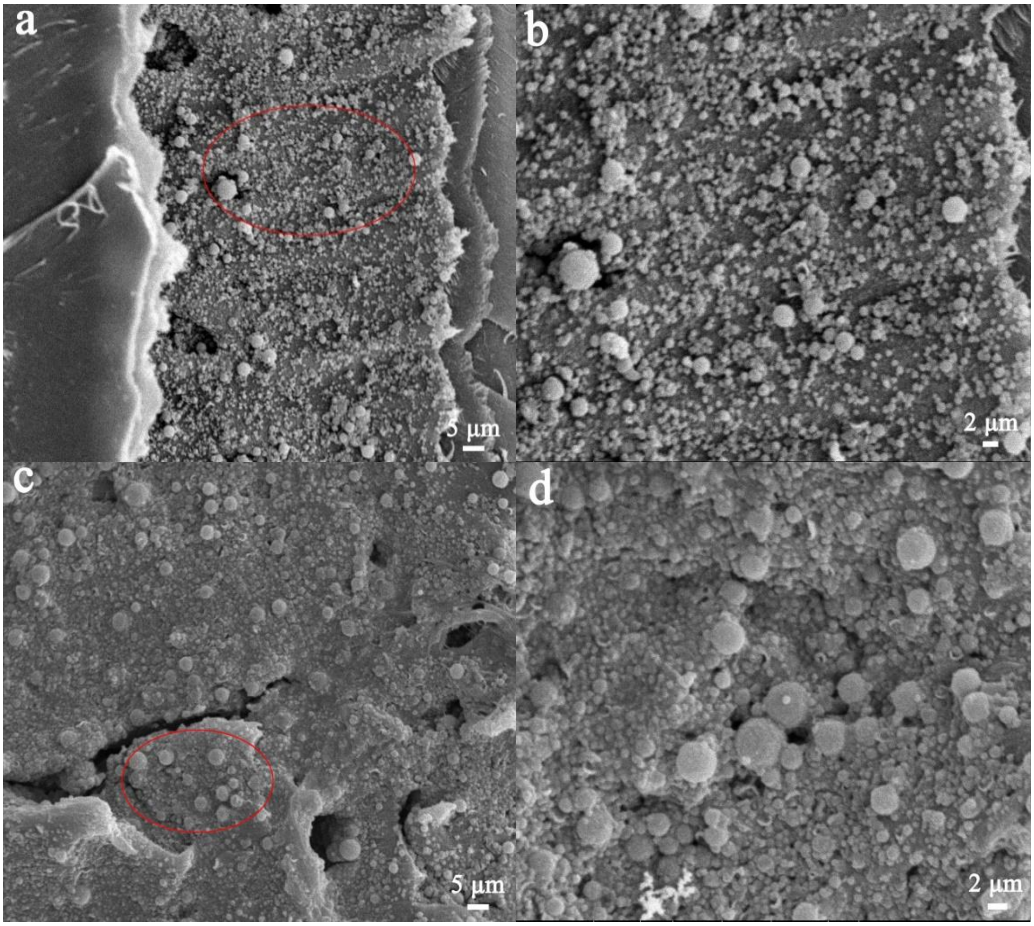

Fig. 5. Tangential (a, b) and transverse (c, d) sections of *Populus tomentosa*

Table 4. Means of Inhibition Zones (mm) of Neem Extract *vs.* Concentrations against Growth of *Penicillium citrinum*, *Trichoderma virens*, and *Aspergillus niger*

Strains	Concentration (%)						
	Control	2.5	5	7.5	10	12.5	15
P. citrinum	N.D.	10.51	12.34	14.22	15.79	16.10	16.80
T. virens	N.D.	11.42	13.86	13.90	15.12	14.97	14.05
A. niger	N.D.	12.22	14.78	15.55	16.36	16.47	16.83
N.D.= Not detected							

Agar Diffusion Assay

The inhibition zone (IZ) depends on the diffusion of bacteriostatic composition and the growth rate of tested strains, and IZ value can be used to evaluate the sensitivity of tested fungi to drugs (Zhao *et al.* 2015). Results from the agar diffusion assay are shown in Table 4. On the whole, the growth of *A. niger*, *T. virens*, and *P. citrinum* was inhibited by neem extract at all concentrations. The antifungal effect was better on the three fungi at high concentrations. Among the three mold fungi monitored, *A. niger* was the most sensitive to neem extract, while *P. citrinum* had the lowest sensitivity. The largest inhibition zone on an agar plate of *A. niger*, *T. virens*, and *P. citrinum* was 16.83 mm, 15.12 mm, 16.8 mm, respectively. Notably, the 10% concentration produced good inhibition against all three fungi, but the antifungal effect decreased using a higher concentration against *T. virens.* Thus, the 10% concentration was selected for follow-up experiments.

AntiFungal Properties of Wood Treated with Microcapsules of Neem Extract

Antifungal effects of neem extract applied to *P. tomentosa* against three mold fungi were evaluated and compared with control samples. The antifungal effect is presented in Fig. 6. Application of microcapsules of neem extract (MNE) to the *P. tomentosa* showed complete inhibition against the growth of the three studied mold fungi.

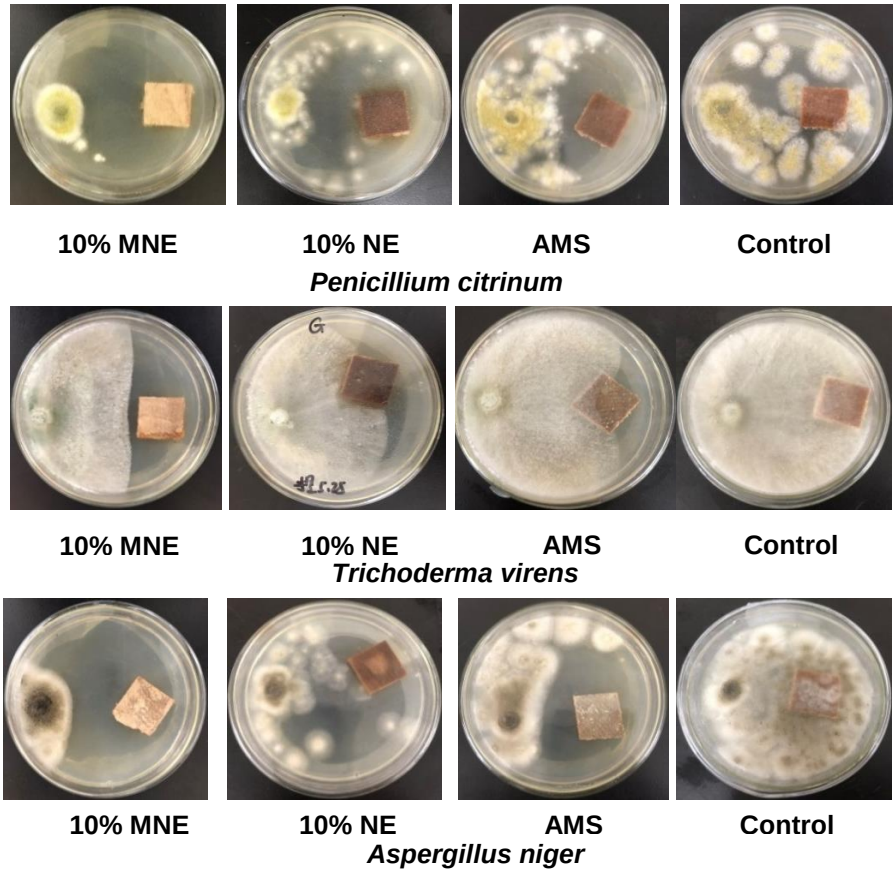

Fig. 6. Antifungal effect of *Populus tomentosa* treated with 10%MNE, 10%NE, and AMS

The application of neem extract (NE) for *P. tomentosa* observed good inhibition against *A. niger* and *P. citrinum* and little inhibition against *T. virens. P. tomentosa* applied with acid mixture of melamine formaldehyde resin and lauryl sodium sulfate (AMS) had good inhibition against *P. citrinum*, little inhibition against *A. niger*, and no inhibition against *T. virens.*

Effect of MNE, NE, and AMS on the Growth of Three Molds over Surface of *P. tomentosa*

Table 5 summarizes the antifungal activity after 14 days' inoculation of fungi (Humar and Pohleven 2005). Specimens applied with MNE showed complete retardation of all three tested fungi, where the marks reached 5. The microcapsules have a slow-release effect, and the core material can be kept longer in *P. tomentosa*. Additionally, the retardation of wood specimens applied with NE and AMS ranged from 2 to 4. In general, the inhibition effect of NE was better than AMS. The synergism of microcapsules with NE and AMS was stronger than the single one.

Table 5. Antifungal Activity was Estimated by Fungal Growth Retardation Using Following Visually Determined Marks

Treatments	Fungal Growth Retardation Marks		
	Penicillium citrinum	*Trichoderma virens*	*Aspergillus niger*
10%MNE	5	5	5
10%NE	4	3	4
AMS	4	2	2
Control	1	1	1

CONCLUSIONS

1. The best conditions for the preparation of neem extract microcapsules was the combination of melamine formaldehyde resin (MF), 1% lauryl sodium sulfate (SDS), and 10% NH_4Cl. The microcapsules possessed regular shape, good dispersion, and uniform particle size.
2. The neem extract inhibited the growth of all tested fungi. The most sensitive fungus was *A. niger*, and *P. citrinum* had the lowest sensitivity. The largest inhibition zones on an agar plate of *A. niger*, *T. virens*, and *P. citrinum* were 16.83 mm, 15.12 mm, and 16.8 mm, respectively. The optimum antifungal concentration of neem seed extract against the three mold fungi was 10%.
3. The diameters of microcapsules were normally distributed in a range of 0.4 μm to 4 μm, and the particles were unevenly distributed in vessels and surface of *P. tomentosa*.
4. Wood specimens treated with MNE completely inhibited all studied fungi, and NE and AMS exhibited weaker effects.
5. The *P. tomentosa* treated with MNE can reach mark 5 against all three molds, while specimens treated with NE and AMS only reached mark 2 to mark 4. Encapsulation of neem extract can enhance the effect of antifungal activities and the service life of

natural extract, and these findings demonstrate the potential use of microcapsules of nature extract in wood mildew resistance.

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