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Research Article
Essential Oil Effect on the Physicochemical Characteristics of Different Wood Species

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Abstract

Background and Objective: The hydrophobicity and electron donor-electron acceptor properties of wood are considered an important parameters for microbial adhesion and wood degradation. Thus, the physicochemical properties of various wood species before and after treatment with *Mentha pulegium*, *Rosmarinus officinalis* and *Cananga odorata* essential oils were assessed. **Materials and Methods:** The hydrophobicity, electron acceptor (γ^+) and electron donor (γ^-) properties of untreated and treated wood were determined using contact angle measurement. **Results:** All wood species tested have an electron donor character γ^- higher than electron acceptor character γ^+ and were characterized as having hydrophobic character except dibetou and beech woods which exhibit a hydrophilic character. The degree of hydrophobicity has decreased considerably using *Mentha pulegium* essential oil with values of water contact angles varying between 14.80$\pm$0.06 and 34.20$\pm$0.45°C followed by *Cananga odorata* and *Rosmarinus officinalis* essential oils with values of water contact angles ranged from 27.70$\pm$1.54° to 49.80$\pm$0.45° and from 37.00$\pm$0.35° to 58.90$\pm$0.45°, respectively. **Conclusion:** In this study, the hydrophobicity and the electron donor-electron acceptor properties have changed after treatment of wood species surfaces with the three essential oils.

Key words: Wood, essential oils, hydrophobicity, electron donor/electron acceptor properties, surface treatment, contact angle

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Wood has many assets that make it an ideal material for construction. In addition to its aesthetic qualities, wood is a lightweight and durable material. For thousands of years, this material was and still used extensively. Wood is one of the oldest building materials used by man in Morocco. This last marks out our history and has undergone major technological progress in different areas such as building and furnishing[1]. Cedar wood, pine and beech are the most species of wood used in Morocco. However, as a natural material, wood is sensitive to different agents of degradation such as insects and microorganisms (fungi and bacteria). These latter adhere to the wood surface and form biofilms[1]. The microbial adhesion step is considered as critical point in the bio film formation process. The physicochemical interactions involved in this process are mainly the acid-base, electrostatic and Van der Waals types. These latter depend on the physicochemical characteristic of material and the microbial surface especially hydrophobicity, surface tension and electron donor-electron acceptor properties[2-4].

To improve the lifetime of wood and boost its resistance to deterioration, different techniques were used previously as the use of the heat treatment[5-7], surface treatment with zinc oxide and titanium dioxide nanoparticles[8,9], acidic dyestuff and fixing agent[10], silicon containing compounds[11-13] and polyelectrolytes[14]. These methods have a negative environmental impact and cause damage to the ecosystem. Thereby, development of non-biocidal solutions for wood protection with good environmental profile has become an overriding concern these last years. Several studies have been conducted on the effect of plant extracts and essential oil component on the surface physicochemical characteristics of cedar wood and their anti-adhesive activity[15,16]. Other studies reported the surface energy of various wood species[17] and the microbial adhesion on some wood species[18]. Thus, the purpose of this research was to investigate firstly the physicochemical properties of various wood species and secondly, to examine the effect of three essential oils (*Mentha pulegium*, *Rosmarinus officinalis* and *Cananga odorata*) on the physicochemical properties of wood species used in Morocco.

MATERIALS AND METHODS

The present study was conducted during the year 2016-2017. The essential oils extraction and the physicochemical properties of various wood species before and after treatment have taken four months.

Plant material: In this study, the essential oil of three aromatic and medicinal plants was used: *Mentha pulegium*, *Rosmarinus officinalis* and *Cananga odorata.* The aerial parts of *Mentha pulegium* and *Rosmarinus officinalis* was harvested in the region of Sefrou located in the Middle Atlas (Morocco) and *Cananga odorata* (known as Ylang-Ylang) were harvested from the Comoros islands. The plants are then distillated to obtain essential oil.

Essential oils extraction: The extraction of essential oils was carried out by hydrodistillation technique using Clevenger-type apparatus. During each test, 200 g of plant material was distilled in 1 L of water for 3 h in a ball surmounted by a vigreux column of 65 cm length (29/32) connected to a condenser. The essential oils recovered were dried with anhydrous sodium sulfate (Na_2SO_4, $\geq$99%, Aldrich) and stored at 4°C in the dark.

Wood species preparation: In this work, six wood species (Cedar: *Cedrus atlantica*, Mahogany: *Khaya ivorensis*, Oak: *Quercus robur*, Pine: *Pinus sylvestris*; Beech: *Fagus sylvatica* and Dibetou: *Lovoa trichilioides*) which are employed in different forms of construction and decoration in Morocco were used. The various wood species were cut into pieces with precise dimensions: length = 3 cm, thickness = 0.4 cm and width = 1 cm. The roughness of the surface wood pieces was set at 0.8 µm using a rugosimeter (Model: Mitutoyo Sj 301). At the end, the samples were cleaned six times with distilled water and then autoclaved for 20 min at 121°C.

Wood species treatment: The effect of essential oils on the physicochemical properties of wood species studied was performed as previously described by Sadiki *et al.*[19]. Brief, a volume of 20 µL of pure essential oil was applied by deposit for 15 min at room temperature (25$\pm$2°C) to the surface of the different wood species. Then, after a good drying and absorption of essential oil tested, the samples were analyzed with contact angle measurements.

Contact angle measurements and surface tension components: The Lifshitz-Van der Waals, acid-base and surface free energy of various wood species were calculated from contact angle measurements which were performed by the sessile drop method using a goniometer (GBX Instruments)[17]. Three measurements of contact angles were made on each wood species using three liquids (of which two must be polar) with well-known surface energy components (Table 1)[20]. Once the contact angles were measured, the

Lifshitz-Van der Waals and acid-base surface tension components were obtained by the three equations of the following form[3]:

$$\gamma_L(\cos\theta+1)=2\left(\gamma_S^{LW}\gamma_L^{LW}\right)^{1/2}+2\left(\gamma_S^{+}\gamma_L^{-}\right)^{1/2}+2\left(\gamma_S^{+}\gamma_L^{-}\right)^{1/2} \quad (1)$$

Where:

- θ : The contact angle
- γ^{LW} : The Van der Waals free energy component
- γ^{+} : The electron acceptor component
- γ^{-} : The electron donor component, (S) and (L) stand for solid surface and liquid phases respectively

The Lewis acid-base component is expressed as:

$$\gamma_S^{AB}=2\left(\gamma_S^{-}\gamma_S^{+}\right)^{1/2} \quad (2)$$

The wood species hydrophobicity was evaluated by the approach of Van Oss *et al.*[20] through contact angle measurements. In this approach, the degree of hydrophobicity of a given material is expressed as the free energy of interaction between two entities of this latter when immersed in water (w): ΔGiwi. So, we said that the material is hydrophilic whether the interaction between the two entities is lower than the interaction of each entity with water (ΔGiwi>0); otherwise, the material is considered as hydrophobic Giwi<0. ΔGiwi is calculated as reported in the following formula:

$$\Delta Giwi=-2\gamma_{iw}=-2\left[\begin{array}{l}\left(\left(\gamma_i^{LW}\right)^{1/2}-\left(\gamma_w^{LW}\right)^{1/2}\right)^2+2\left(\left(\gamma_i^{+}\gamma_i^{-}\right)^{1/2}\right.\\+\left(\gamma_w^{+}\gamma_w^{-}\right)^{1/2}-\left(\gamma_i^{+}\gamma_w^{-}\right)^{1/2}-\left.\left(\gamma_w^{+}\gamma_i^{-}\right)^{1/2}\right)\end{array}\right] \quad (3)$$

Table 1: Surface tension properties of pure liquids used to measure contact angles[21]

Liquid	γ^{LW} (mJ m^{-2})	γ^{+} (mJ m^{-2})	γ^{-} (mJ m^{-2})
Water (H_2O)	21.8	25.5	25.5
Formamide (CH_3NO)	39	2.3	39.6
Diiodomethane (CH_2I_2)	50.5	0	0

RESULTS

Physicochemical characterization of the various wood species before treatment: From the results shown in Table 2, almost all wood species studied have a hydrophobic character qualitatively and quantitatively especially: the cedar, mahogany, oak and the pine wood with values of the water contact angles ranged from 64.6±0.6 to 81.50±0.73° and values of surface energy ranged from -44.81 to -64.38 mJ m^{-2}. Cedar wood has a very hydrophobic character followed by mahogany, oak and pine. In contrast, beech and dibetou have a hydrophilic character qualitatively and quantitatively with values of θ_W = 48.60±0.75° and θ_W = 43.00±0.53° respectively. The results also showed the electron donor-electron acceptor properties of the wood species, it can be seen that all wood species have an electron donor character γ^{-} more than electron acceptor character γ^{+} with higher values for the beech and the dibetou (γ^{-} = 38.63±0.34 mJ m^{-2} and γ^{-} = 34.50±0.85 mJ m^{-2}, respectively).

Physicochemical characterization of the various wood species after treatment: The Table 3 summarized the contact angles values and the surface energies, together with their γ^{LW}, γ^{-} and γ^{+} of the various wood species after treatment. As can be seen, the treatment of wood species surfaces with the three essential oils has changed the degree of hydrophobicity qualitatively and quantitatively. Indeed, the treatment with *Mentha pulegium* essential oil has decreased dramatically the hydrophobicity parameter of all wood species studied. Thus, beech and dibetou presented the lowest values of water contact angle θ_W = 16.80±0.25° and θ_W = 14.80±0.06° respectively. The same statements were obtained with *Rosmarinus officinalis* essential oil, except beech and dibetou wood which have kept their initial hydrophobicity character. Likewise for *Cananga odorata* essential oil, the results showed the decrease of the degree of hydrophobicity for cedar, mahogany, oak and the pine wood with values of the water contact angle ranged from 27.7±1.54° to 49.80±0.45° after treatment. Beech and dibetou have kept

Table 2: Contact angles values, surface energies and their components of wood species before treatment

	Contact angles (±SD)[a]			Surface energy (mJ m^{-2})			
Wood species	θ_w	θ_f	θ_p	γ^{LW}	γ^{+}	γ^{-}	ΔGiwi
Cedar	81.5±0.73	54.5±0.57	21.90±0.2	47.10±0.06	0.44±0.05	3.74±0.3	-64.38
Mahogany	74.6±0.28	42.7±0.8	25.60±0.23	45.84±0.07	0.63±0.1	4.48±0.45	-58.84
Oak	73.1±0.49	38.7±0.93	33.50±0.54	42.64±0.23	1.59±0.19	4.21±0.45	-52.43
Pine	64.6±0.6	30.2±1	18.70±1.04	48.05±0.28	1.10±0.13	8.38±0.49	-44.81
Beech	48.6±0.75	47.3±0.98	21.30±1.5	47.26±0.47	0.30±0.03	38.63±0.34	11.19±0.22
Dibetou	43.0±0.53	32.7±0.58	25.05±0.09	46.01±0.02	0.15±0.03	34.54±0.85	6.44±1.48

[a]±Standard deviations of three measures

Table 3: Contact angles values, surface energies components of the various wood species after treatment

	Contact angles (±SD)[a]			Surface energy (mJ m^{-2})			
Wood species	θ_w	θ_f	θ_p	γ^{LW}	γ^+	γ^-	Δgiwi
Treatment: EO of *Rosmarinus officinalis*							
Cedar	51.7±0.48	12.9±1.93	14.8±1.61	49.00±0.35	1.48±0.13	16.29±0.58	-26.49
Mahogany	44.4±0.14	7.5±0.08	20.8±0.26	47.43±0.08	1.48±0.01	23.21±0.13	-13.48
Oak	57.7±0.51	17.0±0.58	6.4±0.3	50.38±0.03	1.44±0.05	11.16±0.43	-38.2
Pine	58.9±0.45	35.35±0.38	12.4±0.54	49.43±0.11	0.21±0.04	15.79±0.71	-31
Beech	37.0±0.35	29.9±0.21	14.8±0.57	49.02±0.13	0.03±0.01	40.76±0.56	15.13±0.85
Dibetou	43.2±0.14	28.2±1	16.5±0.69	48.62±0.18	0.19±0.05	31.75±0.59	0.12±1.07
Treatment: EO of *Cananga odorata*							
Cedar	27.7±1.54	10.9±0.42	9.5±0.43	50.00±0.06	0.35±0.03	42.47±1.52	14.55±2.23
Mahogany	48.7±0.25	32.2±0.82	14.3±0.16	49.13±0.04	0.12±0.04	27.05±0.74	-8.19
Oak	47.1±0.51	10.8±0.54	18.0±0.33	48.22±0.08	1.39±0.03	20.81±0.52	-17.99
Pine	49.8±0.45	32.4±0.31	9.5±0.23	49.99±0.03	0.09±0.02	25.82±0.69	-11.2
Beech	32.8±0.28	34.6±1.08	16.8±0.1	48.54±0.03	0.30±0.02	49.69±0.83	28.39±0.7
Dibetou	36.0±0.56	16.0±0.83	13.4±0.12	49.31±0.02	0.47±0.03	35.01±0.61	3.99±0.96
Treatment: EO of *Mentha pulegium*							
Cedar	18.2±0.45	13.0±0.51	9.7±0.36	49.97±0.05	0.16±0.02	51.54±0.5	28.04±0.84
Mahogany	29.2±0.35	16.5±0.16	11.2±0.8	49.73±0.14	0.25±0.01	42.79±0.32	15.72±0.44
Oak	34.2±0.45	16.1±1.19	14.8±0.18	49.02±0.04	0.44±0.03	37.06±0.34	7.25±0.55
Pine	28.3±0.26	16.5±0.56	14.2±0.28	49.14±0.07	0.27±0.02	43.67±0.48	17.21±0.79
Beech	16.8±0.25	14.3±0.76	19.2±0.28	47.90±0.07	0.25±0.02	52.79±0.16	30.13±0.38
Dibetou	14.8±0.06	27.3±0.64	10.9±0.45	49.78±0.07	0.04±0.02	61.70±0.53	43.09±0.92

EO: Essential oil, [a]±Standard deviations of three measures

their hydrophilic character with value of ΔGiwi = 28.39±0.70 mJ m^{-2} and ΔGiwi = 3.99±0.96 mJ m^{-2}, respectively.

The results also showed the increasing of electron donor character for wood species characterized as hydrophobic after treatment with the different essential oils especially with *Mentha pulegium* essential oil. The electron donor character using *Mentha pulegium* essential oil was on average 9-fold higher to the one of untreated woods. It was also 6-fold higher using *Cananga odorata* essential oil and 3-fold greater using *Rosmarinus officinalis* essential oil.

For dibetou and beech characterized as hydrophilic, the electron donor character was slightly increased using the different essential oils. This latter was on average 1-fold higher than untreated ones.

DISCUSSION

In line with Vogler[22] and the approach of Van Oss *et al.*[20,23], a surface is considered as hydrophobic when the value of the contact angle with water is higher than 65° and the surface free energy is negative and hydrophilic when θ_W is lower than 65° and ΔGiwi is positive. The contact angle with water gives a qualitative evaluation of the hydrophobicity, while the surface free energy assesses the hydrophobicity quantitatively. Almost all wood species studied have a hydrophobic character qualitatively and quantitatively except beech and dibetou that presented a hydrophilic character. After treatment with the different essential oils, it was noted a modification of the surface properties of the various wood species. This modification was attributed to the chemical composition of each essential oil used for the treatment of various wood species. *Mentha pulegium* essential oil has decreased the hydrophobic character of the different wood species followed by *Cananga odorata* and *Rosmarinus officinalis* essential oil. This can be explained by the high percentage of oxygenated monoterpenes and sesquiterpenes presents for *Mentha pulegium* essential oil (94.43%) principally represented by menthone and pulegone compared to *Cananga odorata* (46.62%) and *Rosmarinus officinalis* essential oil (26.87%) (Essential oils composition results are not published yet). In other words, the presence of oxygen atoms in oxygenated monoterpenes and terpene alcohols decrease the hydrophobicity by creating interactions with water molecules. Oxygen atoms are also responsible for the increase of the electron donor character because of their high basicity.

The initial physicochemical properties of the various wood species corroborated with the results obtained by several researchers who determined the surface energetic of natural wood. De Meijer *et al.*[17] presented the surface energy of various wood species notably pine (θ_W = 64±2°) and cedarwood (θ_W = 69±2°). The hydrophobic character of Moroccan cedar wood was reported in different studies with values of surface energy ΔGiwi = -58.81 mJ m^{-2}, ΔGiwi = -81.98 mJ m^{-2} and ΔGiwi = -6.09 mJ m^{-2} [18,19,24].

Mohammed-Ziegler *et al.*[25] have shown that European oak wood has a hydrophobic character qualitatively with value of θ_W = 81±4°>65° whereas, Gerardin *et al.*[26] have shown hydrophilic character of beech (θ_W = 54.5°<65°). These researchers have also indicated that wood species tested have a sizeable γ^- and a weak γ^+ thus confirming current results[16,18,25,26].

The modification obtained after treatment consistent with those found by Sadiki *et al.*[19,27], who reported that the untreated cedar wood sample, which was hydrophobic has become more hydrophilic after treatment with crude and fractionated *Thymus vulgaris* extract (ethyl acetate, hexane-ethyl acetate and methanolic fractions). Similar trends were observed in the case of cedar wood treated with essential oil components which was more hydrophilic than the untreated sample (θ_W = 42.2±0.3°, θ_W = 39.8±0.3°, θ_W = 46.5±0.4° and θ_W = 39.9±0.6° for carvacrol, carvone, β-ionone and 1.8-cineol treatment, respectively)[16,28]. Other studies reported the decrease of the contact angle with water after microwave plasma treatment for tiger wood and after modification of the birch wood surface by acidic dyestuff and fixing agent[10,29].

For electron donor-electron acceptor character, current findings were corroborated with the results obtained by Sadiki *et al.*[19], who reported that the electron donor character of cedar wood was increased after treatment with *Thymus vulgaris* extract obtained by maceration and ultrasound. In other study, Sadiki *et al.*[27], showed also that the electron donor character obtained after treatment of cedar wood surface with ethyl acetate, hexane-ethyl acetate and methanolic fractions of *Thymus vulgaris* extract were higher than untreated sample (γ^- = 61.4 46 and 56.4 mJ m^{-2}, respectively). Mohammed-Ziegler *et al.*[25] reported in their work that the treatment of European oak wood with chlorotrimethylsilane (CTMS) and octadecyltrichlorosilane (OTS) affected the electron donor parameter: γ^- (oak treated with OTS) = 2.3 mJ m^{-2}, γ^- (oak treated with CTMS) = 5.4 mJ m^{-2} and γ^- (oak untreated) = 0.3 mJ m^{-2}. The same results were found by Jiang and Kamdem[30], which demonstrated that the electron donor character of red oak wood was increased after treatment with PVC-copper amine. Moreover, the findings of this work were in contradiction with the results obtained by Gérardin *et al.*[26], which showed a decrease of the electron donor component of pine and beech wood after heat treatment. These contradictions can be explained by the difference of the nature of treatment between the two studies.

CONCLUSION

The study has shown clearly the modification of the physicochemical characteristics of six wood species by treatment with plant essential oil. Indeed, the treatment has decreased the hydrophobic property of all wood species studied. The most important effect was given by *Mentha pulegium* essential oil due to the oxygenated fraction presented with a high percentage followed by that of *Cananga odorata* and *Rosmarinus officinalis* ones. In addition, the results showed that these essential oils have made the surface of all wood species studied more electron donor than acceptor compared to the untreated ones. These results support the use of these essential oils as natural product to prevent bio film development, thereby limiting wood biodeterioration.

SIGNIFICANCE STATEMENTS

This study discovers the physicochemical properties of six wood species before and after treatment with *Mentha pulegium*, *Rosmarinus officinalis* and *Cananga odorata* essential oils that can be beneficial for the prevention of bio film development. This study will help the researcher to uncover the effect of essential oils used on the hydrophobicity and the electron donor-electron acceptor properties of various wood species. Thus, a new theory on wood protection may be arrived at by modifying the physicochemical properties of wood surface thus preventing microbial adhesion.

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