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SURFACE TENSION, CONTACT ANGLE AND WETTABILITY OF WOOD

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Abstract

Surface tension is an internal force due to an unbalance in molecular forces that occurs when two different materials, such as a wood surface and adhesive, are brought into contact with each other, forming an interface or boundary. The force is due to the tendency for all materials to reduce their surface area in response to the unbalance in molecular forces that occurs at their points of contact. Wetting properties between solids and liquids are of major importance for most industrial products and processes, such as adhesives, paint and lacquers, photograph films, printing inks, finishing, and textiles. Contact angle analysis is a widely used method to study the wetting characteristics of solid materials. There are several methods to determine the contact angles of a liquid on a wood surface. However, the most widely applied method is the sessile drop method.

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1. Surface Tension, Wettability and Contact Angle

When a drop of liquid is dropped on a flat solid material surface, it may spread completely on the surface or, more likely, remain as a drop, forming a certain angle (θ - contact angle) with the solid surface. The contact angle (θ) is a function of the liquid and the solid surface. When the forces of attraction between the liquid and the solid surface (adhesion) are equal to or greater than those between the liquid and the solid surface (cohesion), the contact angle is 0. When the adhesion forces between the liquid and the solid surface are smaller than the cohesion forces in the liquid, the contact angle approaches infinity (∞) (Shaw, 1970).

When two different materials (such as a drop of liquid on a solid surface) come into contact with each other to form an interface or boundary, an internal force called "surface tension" is generated due to the imbalance in the molecular forces that arise. This force is caused by the fact that all materials try to reduce their surface area in response to the imbalance in the molecular forces that occur at the contact points (Aydin, 2004a). Figure 1 shows the forces of attraction that cause surface tension.

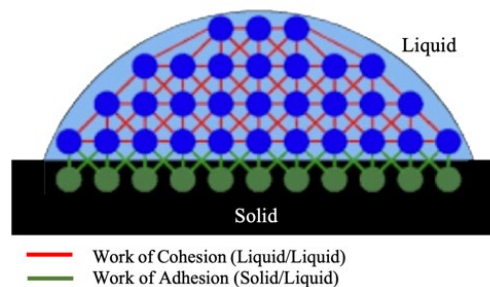


Figure 1: Attraction forces between molecules on the surface and inside a liquid drop (Web-1)

Surface tension is the cohesive force at the liquid's surface that tends to minimize surface area. It is a crucial factor in determining how a liquid interacts with a solid surface. In wood materials, surface tension affects the spreading and penetration of liquids, and the surface tension of both the wood and the interacting liquid significantly affects processes like finishing and adhesion (Smith, 2018).

Several factors can influence wood's surface tension, including moisture content, surface roughness, and chemical composition. A Higher moisture content generally reduces surface tension. Increased roughness can lead to higher surface tension due to capillary effects. Variations in lignin, cellulose, and extractives also affect the surface tension of wood (Wu, 1982).

Materials can be broadly classified based on their affinity with water into hydrophobic (water-repelling) and hydrophilic (water-attracting). Hydrophobicity and hydrophilicity are critical concepts in chemistry, biology, and materials science. They describe the affinity of substances for water, which influences a variety of phenomena, from molecular interactions to materials' macroscopic properties. The interaction of materials with water is expressed by the terms hydrophobic (water repellent) or hydrophilic (water loving). Hydrophilic materials have the ability to absorb water rapidly (Figure 2-a). Surface chemistry allows such materials to wet by forming a film of water or coating agent on their surface. Hydrophilic materials have large surface tension values and have the ability to form hydrogen bonds with water (Aydin, 2004a).

Hydrophobic materials, compared to hydrophilic materials, react inversely to the interaction with water. Such materials absorb little or no water and tend to form bubbles on their surfaces (Figure 2-b). Hydrophobic materials have small surface tension values and lack active groups in the chemical structure of their surfaces to form hydrogen bonds with water (Aydin, 2004a).

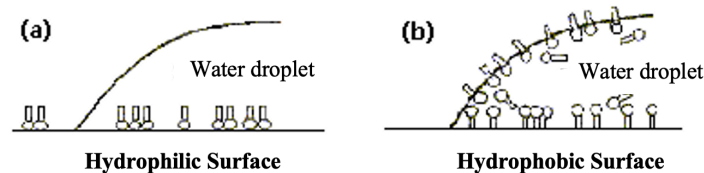


Figure 2: Movement of water molecules on hydrophilic and hydrophobic surfaces (Shaw, 1970)

Wettability is a critical characteristic of wood that affects its performance in various applications, including adhesion, coating, and impregnation processes. Good wettability ensures that adhesives and coatings can spread uniformly and adhere strongly to the wood surface. Effective adhesive bonding requires high wettability to ensure adequate spread and penetration of the adhesive (Marra, 1992). Improved wettability results in stronger and more durable bonds. It is a measure of how easily a liquid can spread across a wood surface, which in turn is influenced by the wood's surface energy, roughness, and chemical composition. Understanding the wettability of wood is essential for optimizing its use in construction, furniture making, and other industries where wood-water interactions are significant (Kamke and Lee, 2007).

Wettability is also important for wood impregnation processes, where liquids such as preservatives, fire retardants, or dyes are introduced into the wood's cellular structure. High wettability facilitates deeper and more uniform penetration of these substances, enhancing the wood's performance in various applications. For example, pressure-treated wood, which involves impregnating wood with preservatives, relies on good wettability to ensure effective protection against decay and pests (Lebow, 2010).

The wettability of wood also affects its susceptibility to biological degradation and weathering. Hydrophobic treatments that reduce wettability can enhance the durability and lifespan of wood products by preventing moisture ingress and subsequent microbial attack (Jones et al., 2016). Recent studies have focused on developing eco-friendly and sustainable treatments that balance wettability and environmental impact (Kim et al., 2022).

The wettability of wood is influenced by its surface characteristics, including chemical composition, surface roughness, and the presence of contaminants (Gray, 1962). Various treatments can enhance the wettability of wood, such as plasma treatment, chemical modification, and sanding (Gardner et al., 1996). Plasma treatment modifies the wood surface at a molecular level, improving wettability by introducing polar functional groups (Griffin, 1962). Chemical treatments can alter the surface energy of wood, making it more hydrophilic or hydrophobic, depending on the intended application (Rowell, 1984).

Different wood species exhibit varying degrees of wettability due to their unique anatomical and chemical characteristics. For instance, Gindl et al. (2013) demonstrated that hardwood species generally have lower contact angles compared to softwoods, attributed to differences in surface energy and porosity. Similarly, Bächle et al. (2019) found that the presence of extractives in certain wood species can significantly reduce wettability, emphasizing the need for species-specific treatment approaches.

Surface roughness is also a critical determinant of wood's wettability. Studies have shown that smoother wood surfaces tend to have higher contact angles, indicating lower wettability (Kúdela and Paprčka, 2016). The interplay between surface roughness and contact angle is complex; rougher surfaces can either increase or decrease wettability depending on the scale and pattern of the roughness (Laskowska and Kozakiewicz, 2021). Techniques such as sanding and mechanical planing are often employed to modify the surface roughness and, consequently, the wettability of wood.

Chemical composition, including the presence of lignin, cellulose, and hemicelluloses, plays a significant role in wood wettability. Treatments that alter the chemical composition, such as thermal modification and chemical grafting, have been extensively studied. Thermal modification, for example, generally decreases wood wettability by increasing hydrophobicity and reducing surface energy (Hakkou et al., 2019). Chemical treatments, such as acetylation and silanization, have also been shown to enhance the hydrophobic properties of wood surfaces (Li et al., 2017).

Contact angle measurements are the most common method used to determine the wettability of a material and to describe the adhesion between a solid and a liquid (Collet, 1972; Kazayawoko et al., 1997; Aydin and Çolakoglu, 2002; Aydin, 2004a; Aydin, 2004b). The contact angle, also called the wetting angle, is the angle between the plane tangent to the liquid surface and the plane tangent to the surface of the solid (Woodward, 2000; Aydin and Çolakoglu, 2002; Jones et al., 2020). It quantifies the wettability of a surface: a low contact angle indicates high wettability, while a high contact angle suggests low wettability (Kalnins and Feist, 1993; Shi and Gardner, 2001).

Several factors impact the contact angle on wood, such as surface energy, roughness, chemical composition, moisture content, and the presence of surface treatments. A higher surface energy typically

reduces the contact angle. Increased roughness can lead to higher contact angles due to air entrapment (Berg, 1993). The chemical composition of the wood, including the presence of extractives and treatment chemicals, significantly affects wettability. For instance, a higher lignin content generally reduces wettability (Gray, 1962). The moisture content of wood influences its surface energy and, consequently, the contact angle. Higher moisture content usually results in lower contact angles due to the swelling of wood fibers (Wu, 1982). Chemical modifications can either increase or decrease the contact angle, depending on the treatment (Sharma and Chattopadhyay, 2021). Surface treatments such as sanding, plasma treatment, or chemical modification, can alter the surface energy of wood, thereby affecting contact angles. For example, plasma treatment increases surface energy and reduces contact angles, enhancing wettability (Hosseinaei et al., 2015).

When a small drop of liquid is dropped on a flat solid surface, there are three possible wetting patterns. These are complete wetting, partial wetting, and non-wetting as can be seen from Figure 3 (Aydin, 2004a).

$\theta = 0$	$\theta < 45^\circ$	$\theta = 90^\circ$	$\theta > 90^\circ$	$\theta = 180^\circ$
Complete Wetting	Partial Wetting - Hydrophilic		Partial Wetting - Hydrophobic	Non-Wetting

Figure 3: Wetting behaviors on wood (Aydin, 2004a).

For perfect wetting, a contact angle $\theta=0^\circ$ is required. In this case, the liquid spreads as a thin film on the solid surface. The case $\theta=180^\circ$ is practically not observed. This is not feasible because it would require the adhesion between the liquid and the solid to be zero, or the surface tension between the liquid and the air to be infinite. There is always some force of attraction between the solid and the liquid. If $\theta < 90^\circ$, it can be said that the liquid wets the solid surface; if $\theta > 90^\circ$, it does not (Bodig, 1962). Both partial wetting with $\theta < 90^\circ$ and complete wetting ($\theta=0^\circ$) provide acceptable interface-free energy for adhesion (Aydin, 2004a). Table 1 also indicates the degree of wetting and interaction strength depending on the contact angle.

Table 1: Degree of wettability and interaction strength depending on the contact angle

Contact Angle	Degree of wettability	Forces of Attraction	
		Solid-liquid	Liquid-liquid
$\theta = 0$	Complete wetting	Strong	Weak
$0 < \theta < 90^\circ$	High wettability	Strong	Strong
$90^\circ \leq \theta < 180^\circ$	Low wettability	Weak	Strong
$\theta = 180^\circ$	Non-wetting	Weak	Strong

It is thought that there are three forces affecting the liquid drop that forms an angle on the surface when dropped on the surface of a solid, and that these forces are in equilibrium. These forces are surface tension between solid and liquid (γ_{SL}), surface tension between solid and gas (γ_{SG}) and surface tension between liquid and gas (γ_{LG}) (Wålinder, 2000; Clint, 2001).

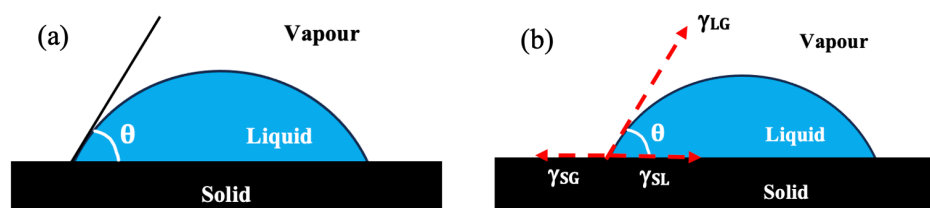


Figure 4: Formation of the contact angle (θ) (a) and the balance of forces acting on the liquid drop on the solid surface (b) (Aydin, 2004a)

The relationship between the surface tensions of a solid and a liquid, the interfacial tensions between the solid and the liquid, and the contact angle (θ) formed by a drop of liquid dropped on a flat surface was formulated by Thomas Young in 1805. This equation, also called Young's Equation, is given below (Kalnins and Feist, 1993):

$$\gamma_{LG} \cos \theta = \gamma_{SG} - \gamma_{SL}$$

Dupré developed an equation for the thermodynamic energy of the interaction between these interface forces, commonly known as the work of adhesion (Mantanis and Young, 1997). According to this equation:

$$W_A = \gamma_S + \gamma_L - \gamma_{SL}$$

Combining the two equations above yields the following equation for adhesion work (Clint, 2001):

$$W_A = \gamma_L (1 + \cos \theta)$$

Thus, the thermodynamic energy of the interaction at an interface can be calculated using the surface tension and contact angle values of the liquid.

2. Contact Angle Measurement Methods to Determine the Wettability of Wood Surfaces

Wetting properties between solids and liquids are critical for many industrial products, including adhesives, surface treatments such as paints and varnishes, printing inks, photographic films, video and audio tapes, and so on. Understanding wood's wettability is critical for optimizing its use in a variety of industrial applications. Controlling and enhancing wettability significantly improves wood adhesion, coating, and impregnation processes, resulting in more durable and reliable wood products.

The wettability of a material is usually determined by contact angle measurements. The accurate measurement of contact angles on wood surfaces is crucial for assessing wettability. There are many techniques available for measuring the contact angles of a liquid on a wood surface. The most commonly used methods are optical and gravimetric techniques. Direct measurement of contact angles from the profile of a liquid drop on the wood surface (Sessile Drop Method) is the most widely used method (Herczeg, 1965; Jordan and Wellons, 1977; Nguyen and Johns, 1979). With the optical technique known as drop shape analysis (Sessile Drop Method), contact angles are measured visually. This method involves placing a liquid droplet on the wood surface and capturing its profile with a high-resolution camera. The contact angle is determined by analyzing the shape of the droplet using software. In the sessile drop method, the contact angle (θ) is defined as the angle between the tangent to the liquid droplet surface and the solid surface at the three-phase contact line. A droplet of liquid is dispensed onto the wood surface, and the equilibrium contact angle is recorded once the droplet stabilizes (Gardner, 1996). The process typically involves the following steps:

1. Surface Preparation: The wood surface is cleaned and conditioned to remove contaminants.
2. Droplet Placement: A microliter syringe or a similar device is used to place a droplet of distilled water or another test liquid on the surface.
3. Image Capture: A high-speed camera captures the side view of the droplet.
4. Angle Measurement: Image analysis software calculates the contact angle from the droplet profile.

The sessile drop method is favored for its simplicity and directness. Although this technique has the advantages of requiring small quantities of test liquid and a small sample of wood, the contact angle of a liquid on a wood surface is difficult to obtain. The reason for this difficulty is the problem of accurately plotting the tangent on the drop profile at the point of contact with the wood surface (Casilla et. al., 1981). Wood surfaces are rough and heterogeneous. A drop of liquid (such as glue) on such a surface like wood does not have perfect axial symmetry. If a single contact angle is measured with such a technique, this measurement may not be representative of all drops on the wood surface, as the contact angles will vary from point to point. Furthermore, an accurate contact angle reading using this technique depends on the experience and skill of the operator taking the measurement (Kazayawoko et. al., 1997). This method can often give different results depending on the operator performing the measurement (Aydin and Çolakoglu, 2002). In addition, contact angle measurements with this method are quite time consuming. Because the angle changes in a short time due to the interaction between the liquid and the solid, direct determination

of the contact angle on solid surfaces is very difficult (Kalnins et. al., 1988). Since porous materials are highly absorbent, it is difficult to obtain contact angles with an optical technique (Aydin and Çolakoglu, 2002).

Recent advancements in goniometry and image analysis have improved the precision of contact angle measurements. Shi et al. (2018) highlighted the importance of using sessile drop methods combined with high-resolution imaging to capture the dynamic changes in contact angle over time. Furthermore, the use of environmental scanning electron microscopy (ESEM) has enabled researchers to study the wettability of wood under various humidity conditions, providing deeper insights into the moisture-related behavior of wood surfaces (Zhao et al., 2020).

In order to overcome the difficulties encountered in the direct measurement of contact angles by sessile drop analysis and to obtain more precise measurements, devices equipped with video cameras and computers have been developed. In today's applications, automated devices equipped with video cameras and computer support, schematically shown in Figure 5, are mostly used to observe the behavior of a liquid drop on a solid surface after contact with the surface, to directly measure the contact angles with precision, and to calculate the surface tension.

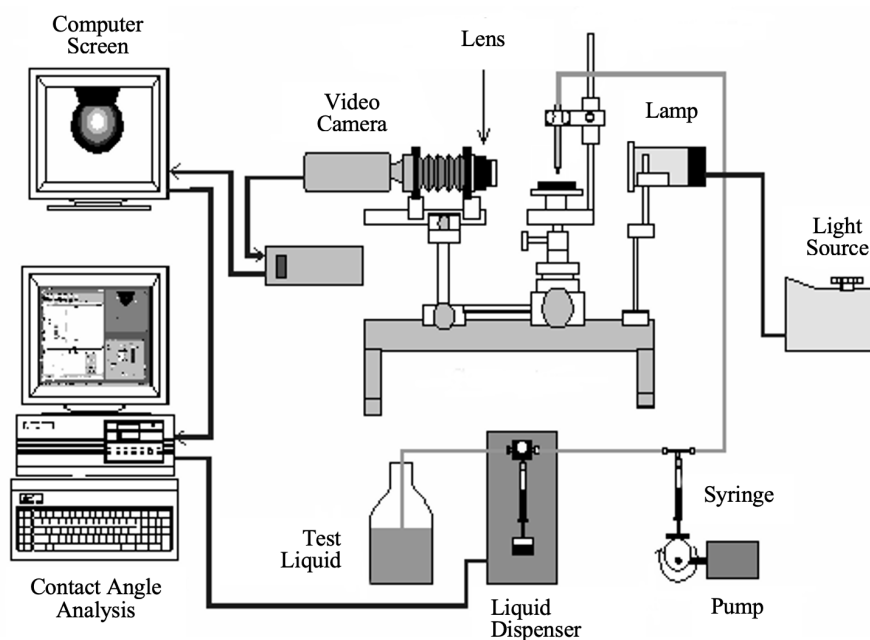


Figure 5: A computer-aided automatic contact angle measurement and analysis (Aydin, 2004a)

The Wilhelmy Plate method offers an alternative approach, particularly suitable for dynamic wettability studies. This method is widely used due to its simplicity, accuracy, and ability to provide insights into the wetting behaviors of various materials. The Wilhelmy Plate Method operates on the principle that a thin, vertical plate made of a known material is partially immersed in a liquid. The liquid wets the plate and climbs up its surface, creating a meniscus whose shape and height are influenced by the contact angle (Adamson and Gast, 1997). It measures the force exerted on a vertically immersed plate as it interacts with a liquid, providing contact angle data based on the force balance. The force exerted by the liquid on the plate is measured as a function of depth, allowing the calculation of contact angles. This technique is particularly useful for assessing the contact angle of wood fibers and small samples (van Oss et. al., 1988).

As can be seen from the schematic diagram of the Wilhelmy Plate Method (Figure 6), when a vertically suspended plate touches a liquid surface or interface, then a force F , which correlates with the surface tension or interfacial tension σ and with the contact angle θ according to the following equation, acts on this plate (Web-2):

$$\gamma = \frac{F}{L \times \cos \theta}$$

The wetted length L of the plate is equal to its perimeter. To measure the force F , the plate is attached to the force sensor of a tensiometer.

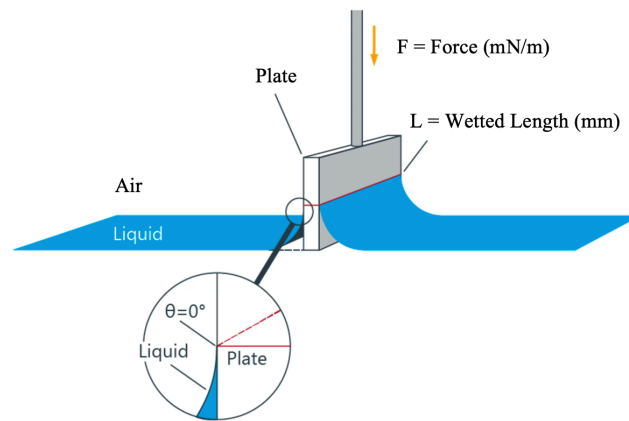


Figure 6: Schematic diagram of the Wilhelmy Plate Method (Web-2)

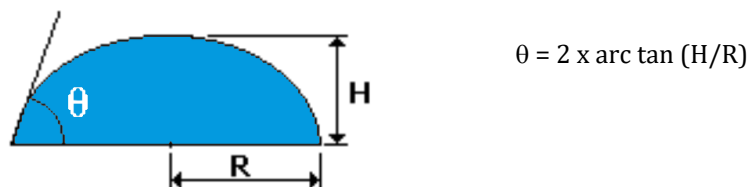
Dynamic contact angle measurements involve determining the contact angle during the advancing and receding motions of the liquid on the solid surface. This is critical for understanding hysteresis, which is the difference between advancing and receding contact angles and is indicative of surface heterogeneity and roughness (Kwok and Neumann, 1999). With the Wilhelmy method, a dynamic contact angle is normally measured by slowly immersing and then withdrawing the solid. The advancing angle is determined during the wetting process and the receding angle during the de-wetting process (Web-2). The advancing contact angle is measured as the plate is immersed in the liquid. The meniscus climbs up the plate, and the contact angle increases until it stabilizes at a maximum value. This angle reflects the liquid's tendency to wet the surface. The receding contact angle is measured as the plate is withdrawn from the liquid. The meniscus descends, and the contact angle decreases until it stabilizes at a minimum value. This angle indicates the liquid's tendency to de-wet the surface (Kwok and Neumann, 1999).

The Wilhelmy Plate method is straightforward and requires minimal sample preparation. It provides precise measurements of contact angles and is applicable to a wide range of liquids and solid materials. On the other hand, it is highly sensitive to surface roughness and heterogeneity, which can affect accuracy. Temperature and humidity can also influence the measurements and need to be controlled.

Casilla et al. (1981) developed an alternative approach to determining the wettability of wood surfaces. This approach is a modified version of the Wilhelmy technique. This modified version determines a wettability index (the area under the force-dip curve) by immersing a conical wood sample in a solution (Kazayawoko et al., 1997). In contrast to drop shape analysis methods, the Wilhelmy technique does not directly obtain a quantitative contact angle value. Although this technique is an alternative tool to characterize the wettability and surface properties of wood, its inability to provide a quantitative contact angle value is a major obstacle since most thermodynamic wetting and adhesion theories require contact angle information (Kazayawoko, 1996; Kazayawoko et al., 1997). However, in specific applications such as gluing and varnishing, dynamic contact angle has been reported to be more informative (Boehme and Hora, 1996).

Rotenberg et al. (1983) developed an alternative method called ADSA (Axisymmetric Drop Shape Analysis) to calculate the contact angle based on measurements of the drop shape.

It is also possible to approximate the contact angles on a material surface using the dimensions of the liquid drop on the surface using the following equation (Liptáková and Kúdela, 1994):



Where θ is the contact angle, H is the height of the drop and R is the radius of the drop base.

Using the Dynamic Contact Angle (DCA) method, it is possible to measure the contact angle at equilibrium. The time required for a liquid drop to reach equilibrium on the surface where it is dropped depends on the liquid used. For example, for a water drop, the equilibrium state is reached in a few seconds, while for oil drops, the time to reach equilibrium is about 3 minutes. The contact angle before reaching equilibrium is called the advancing contact angle. After reaching the equilibrium state, the contact angle

starts to retreat towards the initial state, and the contact angle in this state is called the retreating contact angle (Liptáková and Kúdela, 1994). The lack of thermodynamic equilibrium in non-ideal systems leads to the formation of a contact angle due to the formation in the field of motion of the fluid. This phenomenon is called contact angle hysteresis (Wulf et al., 1997).

There are some factors that cause the formation of contact angle hysteresis. Surface roughness and the heterogeneous structure of the surface are the most important of these factors (Kazayawoko, 1996; Extrand, 1998; Dominigue; 2000). As the liquid drop spreads, it can be contaminated by some contaminants on the surface, and this changes the surface tension of the liquid. The contamination of the liquid drop is also considered among the factors that cause the formation of contact angle hysteresis (Extrand, 1998). Another factor shown to cause hysteresis is the reorganization of molecules and functional groups on the surface of the solid after contact with the liquid (Wålinder; 2000). The occurrence of the hysteresis phenomenon poses a difficulty in the practical measurement of contact angles (Kazayawoko, 1996).

3. Conclusions

The surface tension, contact angle, and wettability of wood are critical parameters influencing its performance in various applications. By understanding and manipulating these properties, we can enhance wood processing techniques, improve product quality, and expand the range of wood's practical uses. Future research should focus on developing more efficient surface treatments and better understanding the interactions between wood surfaces and different liquids.

Understanding wood's wettability is critical for optimizing its use in a variety of industrial applications. Factors such as surface energy, surface roughness, and chemical treatments significantly influence wettability. Accurate measurement of wettability through techniques like contact angle goniometry and the Wilhelmy plate method provides valuable insights into wood-liquid interactions. Controlling and enhancing wettability significantly improves the performance of wood in adhesion, coating, and impregnation processes, resulting in more durable and reliable wood products.

Contact angle measurement is a valuable tool for investigating the wettability and surface properties of wood. By understanding the factors influencing contact angles and employing appropriate measurement techniques, researchers and industry professionals can optimize wood treatment processes, enhancing the performance and longevity of wood products.

Contact angle measurement methods, including the sessile drop, Wilhelmy plate, and dynamic measurements, are fundamental in assessing the wettability of wood surfaces. Each method has its own unique advantages and limitations, making them suitable for different aspects of wettability studies. Understanding these methods enables researchers to select appropriate techniques for specific applications, ultimately improving wood surface treatments and applications.

Disclosure Statement

No potential conflict of interest was reported by the author.

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