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Full Length Article

Cold remote plasma modification of wood: Optimization process using experimental design

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ABSTRACT

A Cold Remote ($N_2 + O_2$) Plasma (CRNOP) process was used in order to create new surface properties on various wooden samples. Two applications were studied and optimized using experimental design. The first one involved samples (fir, pine, beech and oak) treated by the CRNOP in order to increase their impregnability evaluated by dynamic wetting and water absorption measurements. In the best conditions, water absorption was increased by 1.8; 1.9; 2.0 and 5.1 for fir, pine, oak and beech, respectively. The second application involved a plasma polymerization of 1,1,3,3, tetramethyldisiloxane induced by the CRNOP in order to create a superhydrophobic coating on beech sample previously treated by the CRNOP. In optimal conditions, a contact angle equal to 160° could be reached. FTIR spectroscopy give evidence for Si-O-Si, Si-O-C and $Si(CH_3)_n$ bands in a polysiloxane structure.

1. Introduction

Renewability, strength, visual appearance and good thermal insulation properties make wood a material of choice for many applications including building and furniture manufacture. So, it was extensively used during several centuries. However, for last decades it has been gradually replaced by materials such as concrete, steel and synthetic polymer. But recent environmental constraints lead industry to substitute fossil-based materials with green and renewable materials, such as wood, the demand of which is strongly increasing.

As wood is constituted by cellulose, hemicellulose, lignin and extractive substances, its properties are the combination of the chemical properties of these components. Cellulose and hemicellulose are hydrophilic while lignin and extractive substances are hydrophobic. Given that the sum of hydrophilic components is approximately 70% of its dry weight, the wood has a strong affinity with the water and the humidity which is also increased by its natural porous structure. When it remains wet during long periods the lumber swells and deforms what leads to a quick degradation of wooden construction. Moreover, moisture conditions create a very favorable environment for the growth of biological organisms responsible for the degradation of wood. So, the more the wood is dry, the less it risks to be attacked by most types of boring insect or wood-inhabiting fungi which all require moisture and oxygen. The key for the prevention and the control of the growth of these organisms in wood is to choose naturally resistant and durable species which are often expensive. If less durable species are chosen, an alternative is either to keep the wood dry or to use preservative treatment. Sapwood of all species is principally non-durable, while the heartwood of most species is only moderately or slightly durable and requires special protection especially when used in outdoor applications with exposure to weather and wetting conditions. The traditional methods used to prevent the degradation of wood involve wet chemistry which causes environmental concerns because of the use of chemical often dangerous and requires extra cost for the treatment of the unreacted compounds and waste [1,2]. Recently, some less polluting products were developed [3], making impregnation by soaking always used for woods showing a good impregnability. Another alternative to prevent wood degradation is heat treatment but it is to the detriment of mechanical properties [4,5]. To avoid problem of wood degradations and to enhance its durability as well as anti-adhesion, anti-snow, anti-fouling and self-cleaning properties, an interesting way consists in its hydrophobization. For the past few years, works about creation of water repellent wood surface have been reported, including acetylation [6] deposition of metal oxide nanoparticles [7,8], fluoro-containing silicon coatings [9], sol-gel process [10,11] and surface impregnation with various waxes, oils, polyelectrolytes

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and other compounds [12–15]. Super hydrophobic surface can also be prepared combining multi-scale roughness with low surface energy [16,17].

Detrimental effects resulting from wet chemical reactions can be eliminated by using cold plasma techniques which are dry processes and are considered as environmentally friendly green methods [18–20]. Plasma processes can be used either to increase wood wettability and impregnability or to create water repellent coating on wood surface. In the first case, the plasma treatment induces an increase of the polarity of the wood surface by oxidation reaction leading to the formation of polar functional groups which improve wettability, hydrophilicity and so impregnability of wood. Atmospheric corona discharges [21] are often used in this aim as well as radio-frequency discharge plasma [22], dielectric barrier discharge (DBD) [23] or microwave plasma jet [24]. In the second case, the plasma is used to assist a chemical vapor deposition or to initiate polymerization reactions. Polymers formed by plasma polymerization are very different from those created by conventional polymerization even if the same monomer is used for both process. This uniqueness of plasma polymers results from the reaction mechanism of the polymer-forming process which involves fragmentation of the monomer, formation of active sites and recombination of the active fragments [25]. Silicon-based monomers have been widely used in low pressure discharge plasma process for deposition on various substrates as they lead to plasma polymers highly cross-linked, resistant to water permeation and showing a good adhesion with most substrates [26]. The chemical composition and surface properties of the polymer film strongly depend on the operating parameters (plasma gas composition, kind of discharge, pressure, gaseous flow, duration...) and substrate.

While plasma process are often used in order to increase wettability and impregnability of wood, very few works deal about plasma assisted deposition of hydrophobic coating on wood surface and they generally involved DBD [27–29] or capacitively-coupled plasma [30].

The aim of the present work was to use a cold remote ($N_2 + O_2$) plasma (denoted by CRNOP) process in order to modify wood properties. One of the advantages of this kind of reactive gaseous flow, far from the discharge, is its possible large volume extension [31,32]. Two applications were studied:

- The first involved a single CRNOP treatment in order to increase the wettability and/or impregnability of several species of wood. The wettability was evaluated by dynamic wetting studies. Impregnability of wood is generally evaluated from its retention (mass of absorbed preservative by sample unit) and penetration (percentage of depth absorption along the height of the sample) capacities. In this work, water absorption measurements were used as a criterion of the wood samples impregnability.
- The second involved a plasma polymerization of 1,1,3,3-tetramethyldisiloxane (TMDSO) monomer induced by the CRNOP to create a super-hydrophobic wooden surface. As the polymerization occurred in a zone far enough from the discharge such that charged particles are no longer present, the sputtering effect is very low and the deposition rate can reach 100 times that obtained with discharge plasma. In previous works [33–37], such remote plasma used with TMDSO allowed to obtain plasma polymerized TMDSO (ppTMDSO) film showing a polysiloxane-like structure. It was used in a wide variety of applications. Its fire-retardant properties allowed protecting polymer such as polyamide-6 [33,34]. Its barrier effect was efficient in order to hinder Zn^{2+} diffusion from rubber cups to pharmaceutical liquid [35]. It was also used to protect stainless steel substrates (as plate or foam) for high temperature and/or catalytic applications [36] or to protect carbon steel surface against corrosion in NaCl 0.5 M solutions [37].

ppTMDSO coatings deposited on wood were characterized from water contact angles and analyzed by Fourier Transform Infra-Red (FTIR) spectroscopy.

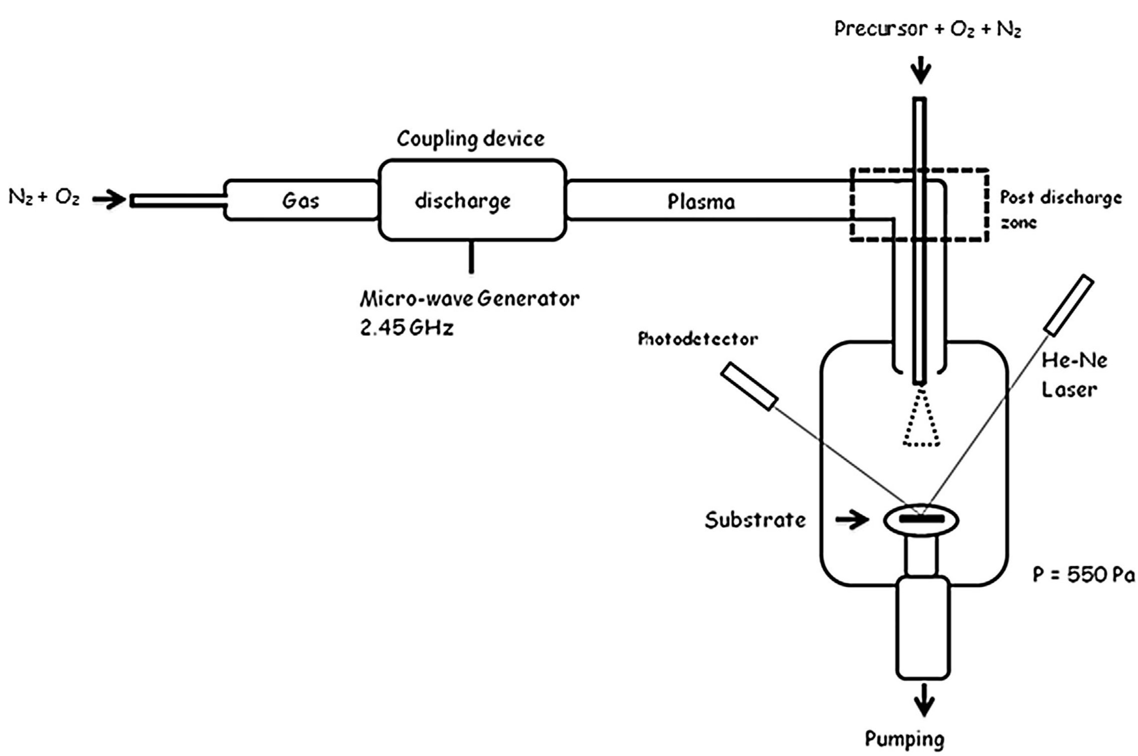

Fig. 1. Experimental set up.