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DOI

[10.17306/J.AFW.2025.2.12](https://doi.org/10.17306/J.AFW.2025.2.12)

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Publication date

2025

Document Version

Final published version

Published in

Acta Scientiarum Polonorum Silvarum Colendarum Ratio et Industria Lignaria

Citation (APA)

Kozowyk, P. R. B. (2025). Testing the hydrophobicity of wood tar adhesives and coatings using contact angle measurements. *Acta Scientiarum Polonorum Silvarum Colendarum Ratio et Industria Lignaria*, 24(2), 173-182. <https://doi.org/10.17306/J.AFW.2025.2.12>

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TESTING THE HYDROPHOBICITY OF WOOD TAR ADHESIVES AND COATINGS USING CONTACT ANGLE MEASUREMENTS

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ABSTRACT

Pine wood tar has long been used as a protective coating for wooden structures in the Nordic countries and has recently been identified as an adhesive in stone buildings. However, the conservation of structures historically reliant on wood tar is increasingly threatened by declining expert knowledge, reduced access to high-quality forest resources, and warmer, wetter climates that accelerate decay of both tar and wood. Positive progress is being made in pine tar research and conservation, including efforts to preserve the remaining expertise among craftspeople, yet the cost of regularly reapplying traditionally made pine tar can be prohibitive in many cases. This study presents a pilot investigation comparing the hydrophobicity of wood tar coatings using water sessile drop contact angle measurements. Traditional Finnish pine was compared to a spruce tar byproduct from industrial biochar production, both with and without powdered charcoal filler. Results show that spruce tar exhibits superior hydrophobicity, and the addition of charcoal significantly improves the hydrophobicity of both tars. These findings highlight the importance of preserving traditional material knowledge for the conservation of cultural heritage and for the development of sustainable, biobased materials. Further research on diverse tar types and substrate materials is needed to optimise both traditional and modern wood tar products.

Keywords: spruce, pine, biochar, pitch, water resistance

INTRODUCTION

Before the advent of petrochemicals, wood tars were used for millennia as adhesives, sealants, and protective coatings. Renewed interest in sustainable, biobased materials has spurred research into historical wood tar technologies (e.g. Karlsson and Lin, 2024; Kozowyk et al., 2017; Lang et al., 2018; Langejans et al., 2022; Li and Suzuki, 2010; Lindblad et al., 2021; Singh and Singh, 2011; Tintner et al., 2021; Wang and Ben, 2020). Wood tar is a viscous black or dark brown liquid obtained by pyrolysing or destructively distilling wood components – cellulose, hemicelluloses and lignin – often incorporating resins, extractives, and their degradation products. The term *wood tar pitch*

generally refers to solid, meltable residues produced during tar distillation at room temperature (Collin and Höke, 2005). However, “pitch” is sometimes also applied to tapped crude or heated conifer resins (Langenheim, 2003), which can be confusing; therefore, the term is mostly avoided here.

Tars are chemically complex mixtures containing hundreds of organic molecules, including acids, alcohols, aldehydes, ketones, phenols, benzenes, and sugars (Sui et al., 2022). Their precise composition depends on both the raw material and the production method. The earliest known tars were derived from birch bark by Neanderthals hundreds of thousands of

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years ago (Mazza et al., 2006; Niekus et al., 2019), with birch bark tar in use well into the Middle Ages (Stacey et al., 2020). From the Iron Age onwards, resin-rich pine wood (*Pinus sylvestris*) became the predominant raw material for tar production across Europe (Hennius, 2018; Hjulström et al., 2006; Samojlik et al., 2013; Tintner et al., 2021). This technology survives most prominently in Scandinavia, where it continues to preserve historic buildings (Bakken, 2016), though it remains marginal and is increasingly threatened by a changing climate, economic constraints, and loss of expert knowledge (Lindblad et al., 2021). To refine traditional material recipes and identify sustainable alternatives, this pilot study presents the first comparison of the hydrophobicity of traditional pine wood tar and an industrial spruce tar byproduct from biochar production, using water contact angle measurements.

Archaeological and historical evidence demonstrates that pine wood tar (and solid wood tar pitch produced by further distilling pine wood tar) was widely used across Europe. Examples include a Norwegian boat from 1250 BC (White and Stern, 2017), an Etruscan ship from 600 BC, and King Henry VIII's flagship, the *Mary Rose* (Robinson et al., 1987). In Stavanger Cathedral, Norway, soapstone blocks were repaired with pine wood tar adhesives (Fig. 1a-b), radiocarbon dated to 1027–1217 AD (Ebert, 2024). Pine tar was also applied on Norwegian stave churches (Fig. 1c), 28 of which survive (Anker, 2016), and on wooden shingles in Sweden and Finland (Lindblad et al., 2021).

The remarkable longevity of these structures underscores the exceptional preservation qualities of properly made, applied, and maintained pine wood tar. For example, Borgund Stave Church in Norway, built over 800 years ago, still retains shingles that have likely never been replaced (Mehlum, 2016). Similarly, many Swedish churches preserve their original wooden shingles, with the oldest dating back to the 14th century (Lindblad et al., 2021). This enduring condition is likely due to the tar rather than the inherent quality of the wood. Analyses indicate that many stave churches and other Norwegian timber buildings of the period were constructed from fast-grown wood, comparable in quality to timber grown today (Thun et al., 2016).

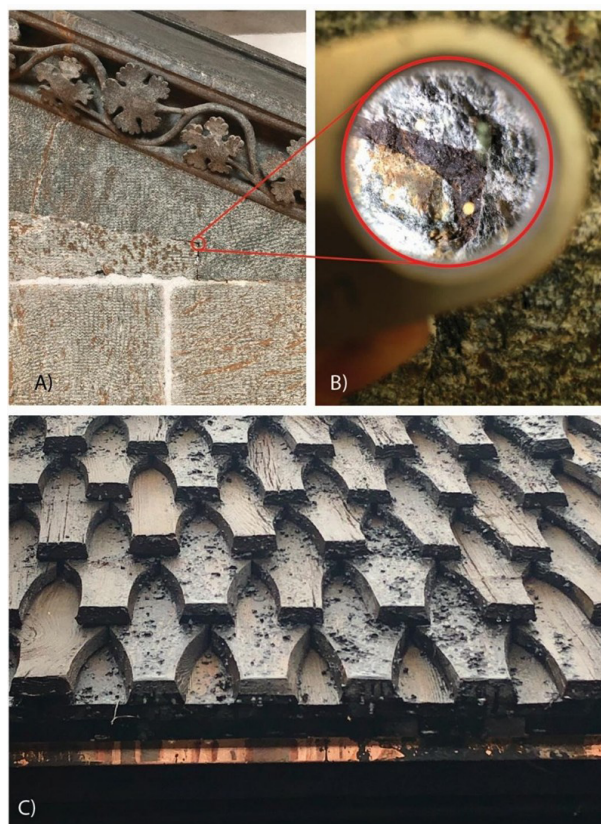


Fig. 1. Surviving examples of pine wood tar used as an adhesive and sealant: Glue applied to stone indents in Stavanger Cathedral, Norway (C); Close-up of the tar adhesive bond line shown in 1A (B); Wood tar coating on the stave church at the Maihaugen Open Air Museum in Lillehammer, which received new shingles and was retarred through the Stave Church Preservation Programme (C) (2005–2009)

Ryc. 1. Zachowane przykłady wykorzystania dziegciu sosnowego jako kleju i środka uszczelniającego: Klej nałożony w zagłębieniach kamiennych w katedrze w Stavanger w Norwegii (A); Zbliżenie spoiny klejowej z dziegciu pokazanej na ryc. 1A (B); Powłoka z dziegciu drzewnego na kościele słupowym w Muzeum na Wolnym Powietrzu Maihaugen w Lillehammer, który otrzymał nowe gonty i został ponownie pokryty dziegciem w ramach Programu Ochrony Kościołów Słupowych (C) (2005–2009).

The longevity of historic tar adhesives and coatings can be attributed to two primary factors. The first is regular maintenance and reapplication, although this was likely less relevant when tar was used as a stone adhesive, such as in Stavanger Cathedral. Historical

sources indicate that tar was reapplied at least every three years (Egenberg et al., 2003). In contrast, modern tar degrades more quickly, requiring recoating every one or two years (Lindblad et al., 2021), which substantially increases maintenance costs. The second factor is the quality of the tar and the specific admixture recipes, which historically contributed to the material's durability and long-term preservation.

Historic tars were likely applied as full-bodied mixtures, produced by seething or boiling tar into a more solid “stir-tar” or wood tar pitch (Egenberg et al., 2003) and often enriched with pigments or fillers such as red earth (clay containing iron oxide), sand, or crushed charcoal (Källbom, 2015). This approach allowed a thick, protective coating to build up over time with each new application, enhancing long-term durability. In contrast, modern tars are typically applied more thinly, without pigments or fillers, and rarely survive or are reapplied often enough to form a substantial protective layer (Lindblad et al., 2021).

Historically, tar production was a time- and labour-intensive process, requiring approximately eight to ten man-days to produce a single barrel (125 litres) of tar (Källbom, 2015). A single church roof could require thousands of litres; for example, between 1781 and 1783, a total of 17,000 litres of tar was used on the Røros church roof, at a rate of 10 litres per square meter (Källbom, 2015). Today, the Church of Sweden requires roughly 6,000 to 12,000 litres of tar annually (Lindblad et al., 2021). Traditionally, mature, dense, resin-rich pine stumps and lower trunks were harvested, demanding careful planning and management of forest resources (Lindblad et al., 2021). Local production of traditionally made pine tar is now insufficient to meet current demand, and high costs necessitate importing tar, where quality and material sources cannot be guaranteed (Lindblad et al., 2021). These challenges are compounded by the declining number of skilled craftspeople with detailed knowledge of tar preparation and application, as well as by changing climate conditions.

Exposure to direct sunlight is one of the primary causes of tar degradation on churches in Norway, with rain and snow playing a secondary role (Egenberg et al., 2003). Combined with tar deterioration, warmer and wetter climates also accelerate the decay of the underlying wood (Lindblad et al., 2021). More variable

weather, including hotter and wetter extremes, places many historic tarred buildings at increased risk. Meteorological records in Sweden indicate a general rise in temperatures across all seasons over the past 60 years, along with increased precipitation over the last 30 years (Kjellström et al., 2022). Rediscovering and improving historic tar recipes thus offers two potential benefits. First, it can help preserve historic buildings and cultural heritage that have endured for centuries. Second, the technology provides a unique opportunity to mitigate the effects of climate change on both new and existing structures.

Wood tar is an often underutilised byproduct of biochar production (Sui et al., 2022), which itself is used to enhance body, viscosity, strength, and UV resistance in biobased materials (Papadopoulou et al., 2024), much like charcoal was historically combined with tar (Källbom, 2015). It is important to clarify terminology: although both biochar and charcoal are produced by the pyrolysis of biomass, charcoal typically refers to material derived from wood intended as fuel, whereas biochar is used primarily for carbon sequestration and is not burned (Hagemann et al., 2018). Because this distinction did not exist historically – and most pyrolysed wood was intended for fuel – the term “charcoal” is used here for historic or traditional materials, whereas “biochar” refers to material produced in modern industrial processes without the intention of being burned.

Biochar has received significant attention in recent years due to its favourable properties, including high carbon content, cation-exchange capacity, large specific surface area, and structural stability (Wang and Wang, 2019). These characteristics make it a promising material for mitigating environmental degradation. Current applications include soil remediation and amelioration, carbon sequestration, organic solid waste composting, and water decontamination (Senadheera et al., 2025; Wang and Wang, 2019); see also (Tan and Yu, 2024). The global biochar market is expanding rapidly, with production in Europe increasing from 5,000 tons per year in 2021 to 50,000 tons per year in 2023 (Senadheera et al., 2025). This growth ensures the availability of significant quantities of waste tar produced locally throughout Europe. Utilising wood pyrolysis by-products for the preservation of wooden buildings presents an untapped opportunity

to sequester carbon while reducing the construction industry's environmental impact (Evans et al., 2022).

The purpose of this pilot study on the hydrophobicity of wood tar coatings is therefore twofold: first, it seeks to determine how conifer wood tars produced during biochar manufacture compare with traditionally made pine tar – an important step toward lowering costs and enabling broader application where strict cultural heritage conservation standards are not required. Second, it investigates the effect of charcoal or biochar additives on the hydrophobicity of wood tar. This has direct relevance for the preservation of cultural heritage buildings, as it supports the reproduction of tars that perform more effectively while more closely reflecting historic materials. Taken together, the results provide insights that can help us learn from traditional practices while developing sustainable strategies to protect cultural heritage and prepare for future environmental challenges.

MATERIALS AND METHODS

To compare wood tars from different sources and evaluate the effect of charcoal on the hydrophobicity of wood tar coatings, contact angle measurements were carried out using water and the sessile drop method. This technique is one of the most accessible and widely applied methods for assessing surface wettability and water resistance of solid materials and coatings (Hagens et al., 2007; Lijesh et al., 2023; Wenzel, 1936; Zhao and Jiang, 2018). Wetting occurs when a liquid comes into contact with a solid and spreads across its surface. Accordingly, higher wettability corresponds to smaller angles at the liquid-solid-gas interface, whereas lower wettability corresponds to larger contact angles. In general, surfaces with contact angles below 90° are considered hydrophilic, while those with angles above 90° are considered hydrophobic (Fig. 2).

Two Finnish wood tars were examined: a traditional kiln-produced pine (*Pinus sylvestris*) tar purchased in 2022, and a spruce (*Picea abies*) tar obtained as a byproduct of industrial biochar production by Carbofex (Nokia, Finland). The unprocessed tars were applied to glass microscope slides and heated to approximately 200°C for one hour to drive off volatiles and create a uniform solid pitch coating. In a parallel experiment, each wood tar was mixed with 30 wt.%

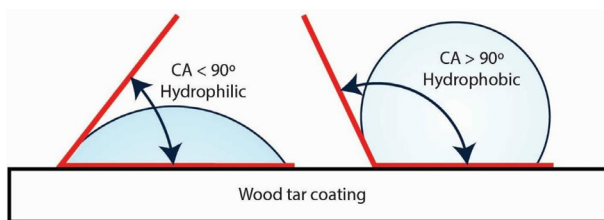


Fig. 2. Illustration of water droplets on a solid surface. Contact angles below 90° indicate hydrophilic behaviour, while contact angles above 90° indicate hydrophobicity, corresponding to greater resistance to wetting

Ryc. 2. Ilustracja kropli wody na powierzchni stałej. Kąty zwilżania poniżej 90° wskazują na właściwości hydrofilowe, natomiast powyżej 90° oznaczają hydrofobowość, czyli większą odporność na zwilżanie

beech (*Fagus sylvatica*) charcoal (Kremer Pigmente, Aichstetten, Germany), and the same preparation procedure was followed. All slides were then conditioned at approximately $20\text{--}25^\circ\text{C}$ and 60% relative humidity for 48 hours prior to testing.

Measurements were performed using a KSV CAM 200 optical contact angle meter equipped with an automatic dispenser. Drops of $5\ \mu\text{L}$ of distilled water were placed on the surface of the wood tar coatings, and photographs were captured approximately 2 seconds after contact (Fig. 3a–b). Contact angles were calculated using the Young–Laplace curve fitting method in Attension Theta software (NanoScience

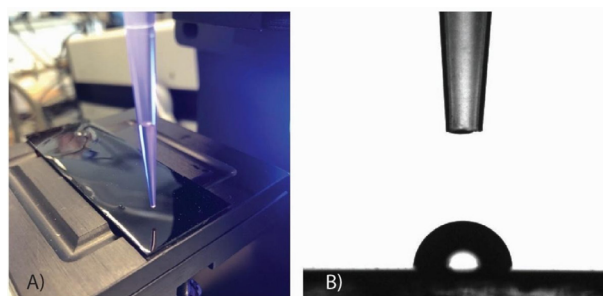


Fig. 3. Wood tar coating applied to a glass microscope slide (A); $5\ \mu\text{L}$ water droplet as observed through the device camera (B)

Ryc. 3. Powłoka z dziegciu drzewnego naniesiona na szkiełko mikroskopowe (A); Kropla wody o objętości $5\ \mu\text{L}$ widoczna przez kamerę urządzenia (B)

Instruments, Phoenix, AZ). According to standard procedures, two measurements (left and right side) are typically taken on three individual drops for each material (ASTM, 2013). To account for surface irregularities caused by the inhomogeneity of these natural materials, two measurements were performed on a minimum of five individual drops per material.

RESULTS

Contact angle

Traditional kiln-produced pine tar exhibited the lowest mean contact angle (77.4° , $n = 5$, std dev = 2.2). Industrially produced spruce tar showed the second-lowest mean contact angle (85.6° , $n = 6$, std dev = 3.1). The addition of 30 wt.% charcoal uniformly increased the contact angle by nearly 50% for both materials, resulting in 114.5° for pine tar ($n = 6$, std dev = 5.3) and 124.8° for spruce tar ($n = 6$, std dev = 4.0; Fig. 4). Samples containing charcoal exhibited greater surface irregularities, which likely contributed to the higher variation observed in these measurements. A one-way ANOVA with a post-hoc Tukey HSD test, conducted using OriginPro (v.10.1.0.178, OriginLab Corporation), confirmed that,

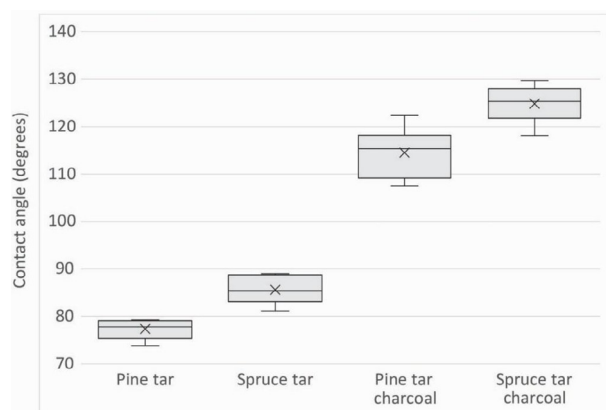


Fig. 4. Mean contact angles for each material. Spruce tar exhibits a higher contact angle than pine tar, and the addition of charcoal significantly increases the contact angle of both materials

Ryc. 4. Średnie kąty zwilżania dla każdego materiału. Dziegieć ze świerku wykazuje wyższy kąt zwilżania niż dziegieć sosnowy, a dodatek węgla drzewnego znacząco zwiększa kąt zwilżania w obu przypadkach

at the 0.05 significance level, the mean contact angles of each material differed significantly. Individual measurement results are presented in Table 1.

Table 1. Averages of two contact angles (left and right) for each drop on all materials

Tabela 1. Średnie wartości dwóch kątów zwilżania (lewego i prawego) dla każdej kropli na wszystkich badanych materiałach

Drop number	Pine tar	Spruce tar	Pine tar-charcoal	Spruce tar-charcoal
1	76.9	86.5	116.8	118.1
2	73.8	83.8	115.1	125.6
3	77.8	88.7	122.3	125.0
4	79.0	81.2	109.7	129.7
5	79.3	84.5	115.6	127.4
6		89.0	107.5	123.0
Mean	77.4	85.6	114.5	124.8
Std Dev	2.2	3.1	5.3	4.0

DISCUSSION

The results provide two key insights: first, regarding the wood type, modern industrial spruce tar outperformed traditional pine tar. This is promising for future applications and the development of novel tar-based materials, but it raises the question of why pine was historically preferred. Second, the findings support the notion that the addition of pigments or fillers – as likely used in historic tars (Källbom, 2015) – served a functional purpose.

Wood type

Outside Scandinavia, tars are produced from other conifers, such as *Cedrus libani*, *Cedrus atlantica*, and various juniper (*Juniperus*) species in Morocco and Turkey (Julin, 2008; Kurt et al., 2008), typically for veterinary or medicinal use. Unsurprisingly, *Pinus sylvestris* was not used in Morocco, as it lies outside its natural range. However, it is unclear why *Picea abies* was not used in Scandinavia, where it is abundant (Caudullo et al., 2017; Samojlik et al., 2013). Possible

explanations include limited archaeological evidence and sparse written sources from small-scale production sites, or material properties favouring pine tar that have not yet been systematically tested.

If the historic preference for pine tar was not performance-related, it may relate to resin production and tar yield. Resin canal size and abundance are the primary anatomical determinants of resin output (López-Álvarez et al., 2023), and pines generally possess larger and more abundant canals than other conifers (Connors, 2015; Wu and Hu, 1997). Pine is widely tapped for resin globally (Cunningham, 2012), and higher resin production likely yields more tar, since resin acids constitute a substantial portion of the product (Egenberg et al., 2003; Tintner et al., 2021). Historical methods specifically targeted resin-rich pine wood, supporting the hypothesis that pine was selected for maximum tar yield (Lindblad et al., 2021). In labour-intensive tar production, trees with the highest tar yield were probably preferred, which may have excluded spruce.

Today, biochar and tar can be produced from diverse organic feedstocks, including wood pulp, agricultural residues, and sewage waste (Lehmann and Joseph, 2009). Automated, mobile, and industrial systems (Maroušek and Trakal, 2022) reduce the labour and skill intensity, removing historical limitations. While pine remains essential for the maintenance of protected heritage buildings, spruce tar shows potential for new materials, leveraging the long-standing success of pine tar. Pine tar produced in the same manner as the spruce tar tested here may also offer an economically and environmentally sustainable option for conservation. Nevertheless, numerous factors influence the performance of wood tar coatings, including substrate material, weather exposure, thermal resistance, ageing and production temperature (e.g. Egenberg et al., 2003), which were not addressed in this study. Further research is needed to clarify why pine was historically favoured and whether spruce or other tars could provide superior long-term performance.

Additives

In regions historically producing large quantities of tar, charcoal was a valuable byproduct with multiple uses, ranging from gunpowder and artists' pigments to fuel (Gray et al., 1982; Tomasini et al., 2012). Charcoal

powder was also used as an additive in historic tar (Källbom, 2015), though it is rarely included in modern wood tar products intended for conservation (Lindblad et al., 2021). The results of this study demonstrate that charcoal improves the hydrophobicity of wood tar and may also serve additional functions. Among the favourable properties of biochar when incorporated into other materials is its role as an efficient UV-protective additive in biocomposites (Papadopoulou et al., 2024). In addition to enhancing hydrophobicity and serving as an important source of modern wood tar, biochar may also improve the UV resistance of adhesives and coatings. This is particularly significant given the accelerating effect of sun exposure on the degradation of traditional tar coatings (Egenberg et al., 2003). While this study used powdered beech wood charcoal, further experiments with different biochar materials – especially those derived from wood tar production – represent an important next step. Such investigations offer a promising avenue for improving the performance and durability quality of wood tar-based materials.

CONCLUSION

This first study of the water contact angle of pine and spruce wood tar has yielded promising results for both enhancing traditional materials used in heritage conservation and developing new materials inspired by historical knowledge. The nearly 1,000-year preservation of wood tar adhesives and tar-coated wooden structures under the harsh conditions of western Norway, along with the enduring historical knowledge associated with them, provides an invaluable source of insight into biobased materials and construction techniques. A simple modification of traditional pine tar with charcoal – guided by historic practices – significantly improves its hydrophobicity, underscoring the value of expert material knowledge from the past and demonstrating its potential for modern applications (e.g. Ebert, 2024; Ebert and Bjelland, 2023). Moreover, the performance of industrially produced spruce tar highlights additional opportunities for improving wood tar materials beyond heritage conservation contexts (c.f. Chen et al., 2022; Firouzbehi et al., 2021; Lis et al., 2016). It remains unclear why spruce tar was not historically used alongside pine. Further studies investigating other material properties, degradation

processes, and compound mixtures will help answer such questions regarding past material choices and may inspire innovative wood tar-based materials for future applications.

ACKNOWLEDGEMENTS

This research is part of the project “Sticking Stones: Rediscovering medieval wood tar adhesives for stone conservation” (Project 344868), funded by the Research Council of Norway. We thank Bettina Ebert, Silas Ploner, Uta Pottgiesser, and members of the Nordic Tar Network and the EU-PoTaRCh COST Action (CA22155) for their inspiring discussions. Special thanks to Hans Poulis and Roy Awater from the Faculty of Aerospace Engineering and the Aerospace Structures and Materials Laboratory at TU Delft for the generous use of lab space and equipment, and to Kim Lehiö from Carbofex for providing the spruce tar sample.

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BADANIE HYDROFOBOWOŚCI KLEJÓW I POWŁOK ZE SMOŁY DRZEWNEJ PRZY UŻYCIU POMIARÓW KĄTA ZWILŻANIA

ABSTRAKT

Smoła sosnowa ma długą historię stosowania jako powłoka zabezpieczająca drewniane konstrukcje w krajach nordyckich. Niedawno odkryto jej zastosowanie jako spoiwa w budynkach kamiennych. Utrzymanie i konserwacja wielu obiektów, w których tradycyjnie i historycznie stosowano smołę drzewną, jest dziś zagrożone. Produkcja przystępnej cenowo, czystej i jednnorodnej smoły drzewnej o odpowiedniej jakości napotyka poważne trudności z powodu zaniku wiedzy eksperckiej oraz ograniczonego dostępu do wysokiej jakości zasobów leśnych. Problem pogłębiają cieplejsze i wilgotniejsze warunki klimatyczne, które przyspieszają degradację zarówno samej smoły, jak i drewnianego podłoża. Równocześnie podejmowane są pozytywne działania w zakresie badań i konserwacji smoły sosnowej, m.in. w celu zachowania wiedzy rzemieślniczej, jednak koszty regularnego stosowania tradycyjnie wytwarzanej smoły bywają w wielu przypadkach zbyt wysokie. W niniejszym artykule przedstawiono wyniki badania pilotażowego porównującego hydrofobowość powłok smołowych przy użyciu pomiaru kąta zwilżania metodą kropli osiadłej. Tradycyjna smoła sosnowa produkowana w Finlandii została porównana ze smołą świerkową, będącą produktem ubocznym przemysłowej produkcji biowęgla. Oba rodzaje smoły badano również po dodaniu wypełniacza w postaci sproszkowanego węgla drzewnego. Wyniki wskazują, że smoła świerkowa charakteryzuje się wyższą hydrofobowością, a dodatek węgla drzewnego istotnie zwiększa hydrofobowość obu rodzajów smoły. Badanie to podkreśla znaczenie zachowania tradycyjnej i historycznej wiedzy materiałowej zarówno dla ochrony dziedzictwa kulturowego, jak i dla rozwoju nowych, zrównoważonych materiałów pochodzenia biologicznego. Konieczne są jednak dalsze badania, obejmujące szerszą gamę smołowych produktów i rodzajów podłoża, aby zoptymalizować zarówno tradycyjne, jak i nowoczesne materiały na bazie smoły drzewnej.

Słowa kluczowe: świerk, sosna, biowęgiel, smoła, odporność na wodę