# THE EFFECT OF NATURAL WEATHERING ON STABILITY OF COATING SYSTEM WITH SELF-HEALING MICROCAPSULES

Zuzana Vidholdová – Gabriela Slabejová – Jozef Kúdela – Ján Svocák

# ABSTRACT

The performance of self-healing and standard coating systems on oak wood exposed to natural weathering for two years is investigated in the paper. The changes in coating thickness, colour coordinates  $L^*$ ,  $a^*$ ,  $b^*$ , total colour difference  $\Delta E^*_{ab}$ , impact resistance, and adhesion by Pull-off and Cross-cut tests are evaluated in the study. The results show that while both coating system types displayed slight colour alterations, the self-healing coating system containing microcapsules exhibited more colour stability, maintaining a colour difference ( $\Delta E_{ab}^*$ ) of 2.6. The primary factors contributing to colour change were variations in lightness (L<sup>\*</sup>) and, to a<sup>\*</sup> lesser degree, the b<sup>\*</sup> coordinate leading to subtle darkening and yellowing of the coatings. However, the self-healing coating system exhibited lower impact resistance indicating increased brittleness and susceptibility to cracking corresponding to the standard coating system. Regarding adhesion, the self-healing coating system showed no changes after weathering, as measured by the Pull-off and Cross-cut tests. In contrast, the standard coating system experienced a 32% reduction in adhesion in the Pull-off test. These findings suggest that although self-healing coatings may offer advantages in colour retention, their application on exterior wood surfaces, particularly those exposed to mechanical stress, requires careful consideration.

**Keywords:** adhesion properties; colour stability; impact resistance; self-healing coatings, weathering.

### **INTRODUCTION**

Mechanical damage and crack formation belong to the most harmful defects in wood surface coatings, as they compromise the integrity of the coating film, which leads to a significant reduction in its protective capabilities against environmental factors, particularly water, UV radiation, chemical agents, contamination, and biological organisms such as fungi and algae (Bulian and Graystone, 2009; Viitanen and Ritschkoff, 2011; Hochmańska *et al.*, 2014; Reinprecht and Pánek, 2015; Kúdela *et al.*, 2017, Liu *et al.*, 2017; Vidholdová *et al.*, 2017; Cogulet *et al.*, 2018; Slabejová *et al.*, 2019; Jirouš-Rajković and Miklečić, 2021; Hasanagić *et al.*, 2024). These disturbed areas act as starting points for further degradation, mainly through moisture absorption, which induces swelling in the surface layers of the wood. The resulting tension at the wood-coating interface promotes crack propagation, accelerating the coating breakdown (Lagaňa *et al.*, 2021). This process impairs adhesion between the coating and the wood substrate, leading to peeling, and necessitates frequent

and costly maintenance of the damaged surface. Considerable research efforts are focused on mitigating these negative effects to extend the lifespan of surface coatings.

A significant advancement in this field is the development of self-healing coatings (White et al., 2001; Survanarayana et al., 2008; Cho et al., 2009; Samadzadeh et al., 2010; Zhao et al., 2012; Schreiner et al., 2017; Landry et al., 2023). Microencapsulation technologies offer a wide range of core and shell material combinations for producing selfhealing coatings (Ghosh, 2006). Recent reviews by Blanchet and Pepin (2021) and Chang et al. (2023) highlight the increasing use of microcapsules in wood surface treatments as a notable trend. A variety of organic and inorganic materials, such as melamine-formaldehyde resin, urea-formaldehyde resin, polyurethane, poly(methyl methacrylate), polystyrene, polyester, silica, calcium carbonate, titanium dioxide, and alumina, have been identified as suitable for the construction of microcapsule shells in phase-change materials (Maiti et al., 2023). These microcapsules, often filled with active agents such as low-viscosity polymers, release their contents when cracks form, filling and sealing the damaged areas. This selfrepair mechanism restores the integrity of the coating, enhancing its durability and reducing the frequency of required maintenance, thus providing a promising solution for more resilient wood surface treatments. These coatings are effective for microcrack repair but may not handle larger-scale damage effectively. However, the self-healing functionality of these capsules is typically single-use, as they cannot be refilled once the contents are released.

Research has explored several types of microcapsules for use in wood surface treatments. For instance, Queant *et al.* (2018) investigated microcapsules embedded with UV absorbers in calcium carbonate templates, coated with a light-responsive polymer, which enhanced UV protection by gradually releasing the absorbers, thereby extending the longevity of the wood coating. Yan and Peng (2020) demonstrated the use of urea-formaldehyde resin (UF) microcapsules in waterborne coatings intended to improve crack resistance on wood surfaces. Han *et al.* (2020) studied the influence of preparation processes on the performance of microcapsules, using paraffin as the core material and melamine-formaldehyde as the shell material. Furthermore, Xia *et al.* (2024) examined how mixed shellac microcapsules coated with melamine rice husk powder, on the aging resistance and self-healing properties of waterborne coatings for *Tilia europaea* L. Wang *et al.* (2022) prepared intelligent wood coatings with dual functions of self-healing and discolouration, integrating fluorane microcapsules for discolouration and shellac resin microcapsules for self-healing.

Several studies were conducted to examine the effects of microcapsule addition on the optical and mechanical properties of coats. For example, in the study by Yan et al. (2021), UF-coated waterborne acrylic resin microcapsules with varying core-shell ratios were tested. As microcapsule concentration increased, the colour difference and hardness of the coating film gradually increased while gloss and adhesion decreased. Impact resistance and elongation at break initially increased and then declined. Increasing the core-shell ratio also led to a similar trend: hardness and impact resistance rose initially and then decreased, while adhesion decreased consistently. In another study by Yan et al. (2022), melamine/rice husk powder-coated shellac microcapsules were prepared via in-situ polymerization, using melamine resin and rice husk powder as the wall material and shellac as the core. The authors found that these coatings had minimal impact on optical properties. Following a UV aging resistance test, changes in colour difference and gloss were minimal. Wu et al. (2023) found that the gloss and adhesion of waterborne wood paint coatings decreased with increasing microcapsule concentration. The hardness, impact resistance, and tensile properties initially increased and decreased as microcapsule content rose. This study utilized microcapsules containing multi-walled carbon nanotubes and carbonyl iron powder as core materials and a melamine-formaldehyde resin as the wall. Finally, in the study by Xia *et al.* (2024), it was found that waterborne coatings with mixed shellac microcapsules maintained their optimal optical and mechanical properties over time. According to aging resistance tests, these coatings demonstrated a prolonged effective lifespan.

To develop effective self-healing coatings, it is crucial to assess the impact of microcapsules on the properties of the coating and its interaction with other layers within the coating system. A study by Xia *et al.* (2022) further highlights that the impact resistance of a paint film with microcapsules added solely in the primer or topcoat was lower compared to the films with microcapsules incorporated into both layers. While microcapsules can enhance impact resistance to a certain extent, their agglomeration weakens the adhesion between the paint film and the wood surface, leading to reduced overall impact resistance. Consequently, the best impact resistance was observed in coatings containing microcapsules in both the primer and topcoat layers, as this configuration provided optimal attachment to the wood surface and improved durability.

The objective of this experimental study is to evaluate the effectiveness of self-healing coatings by investigating the influence of microcapsules on the selected coating's properties and its interaction with other layers in the coating system. These specific systems were designed for dimensionally stable wooden products such as windows, doors, and other exterior wooden construction. Specifically, it examines a self-healing surface treatment incorporating microcapsules within the intermediate layer. The research aims to evaluate the impact of this modified intermediate layer on colour stability during natural weathering, along with its effects on selected mechanical properties and adhesion of the surface treatment systems to oak wood.

# **MATERIALS AND METHODS**

### Wood Material

Oak wood (*Quercus petraea* L) was used in the experiment. The dimensions of the test specimens were 375 mm  $\times$  78 mm  $\times$  20 mm. Fourteen test specimens were used in experiment. The test specimens' surface was sanded according to the recommendations listed in technical sheets for the coating materials.

### **Coating System**

The following water-based white pigmented coatings were selected for the surface treatment of dimensionally stable components such as windows, exterior doors, and furniture for winter gardens and outdoor use:

- Base coat A protective impregnation based on a mixture of alkyd and acrylic resins, offering good coverage. It contains wood preservative 0.8% of 3-iodo-2-propynyl-N-butyl-carbamate (IPBC) and 0.4% of tebuconazole.
- Intermediate coat with microcapsules An acrylic dispersion-based coating with a low solvent content, enriched with microcapsules designed to seal cracks caused by weathering or mechanical damage.
- Top coat An acrylic dispersion-based coating with excellent weather resistance.

White pigmented coatings on oak wood often balance aesthetic goals with practical considerations, offering a clean, modern look while masking any natural imperfections or colour variations and ensuring consistency. They also enhance the wood's texture by highlighting its grain, adding depth and character to wooden constructions like windows and doors.

Two types of white pigmented coating system were created:

- Standard coating system Consisting of one coat of the base (average wet film thickness of 100 μm) and one coat of the top coat (150-180 μm wet film thickness).
- Self-healing coating system Consisting of one coat of the base (100 μm wet film thickness), one coat of the intermediate coat with microcapsules (150-180 μm wet film thickness), and one of the top coat (150-180 μm wet film thickness).

The test specimens were surface finished on all sides by low-pressure spraying.

## Natural Outdoor Weathering of Coated Wood Specimens

Exposure of all coated specimens in the exterior was carried out according to the standard STN EN 927–3 (2006) in frames facing south at an angle of 45° and placed approximately 1 m above the ground for 24 months at the Technical university in Zvolen.

## **Thickness of the Coating Film**

Non-destructive method was chosen to measure the film thickness using the ultrasonic instrument PosiTector 200. Thickness measurements were taken at 12 points on each coated specimen across the top surface before and after 2 years of weathering.

## **Colour Evaluation**

The colour characteristics of the wood surfaces were measured using a spectro-photometer Spectro-guide 45/0 gloss manufactured by BYK – GARDNER GmvH. The device was configured with a D65 light source and a 10° observation angle. The parameters L<sup>\*</sup> (= black/white), a<sup>\*</sup>(+a<sup>\*</sup>/ – a<sup>\*</sup> = red/green), and b<sup>\*</sup> (+b<sup>\*</sup>/ – b<sup>\*</sup> = blue/yellow) were measured at six points at the same position on each coated specimen over the tangential surface at the beginning of the experiment and after 2 years of weathering. The colourimetric parameters for each coated specimen were analysed according to the CIE Lab system (ISO 7724–3: 1984). A positive value for L<sup>\*</sup>, a<sup>\*</sup>, or b<sup>\*</sup> indicates a lighter, redder, or yellower colour, respectively.

The relative colour changes  $\Delta L^*$ ,  $\Delta a^*$ , and  $\Delta b^*$ , representing the differences between the chromaticity coordinates of the weathered coated specimens and the initially coated specimens, were used to calculate the total colour difference ( $\Delta E^*_{ab}$ ) according to Equation (1):

$$\Delta E_{ab}^* = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}} \tag{1}$$

### **Impact Resistance**

The impact resistance of the surface finishes was determined according to the standard STN EN ISO 6272–2 (2011). The intrusion (diameter of the intrusion) was measured and the surface finish was evaluated subjectively according to Tab. 1. For the purposes of microscopic analysis, a Keyence VHX-7000 digital microscope (Keyence Corporation, Osaka, Japan) was used (Adamčík *et al.* 2023).

Degree	Visual evaluation
1	No visible changes
2	No cracks on the surface and the intrusion were only slightly visible
3	Visible light cracks on the surface, typically one to two circular cracks around the intrusion
4	Visible large cracks at the intrusion
5	Visible cracks were also off-site of intrusion, peeling off the coating

Tab. 1 Impact resistance: degree and evaluation.

# **Adhesion Tests**

Adhesion of the coating films to wood was determined by the Pull-off test according to the standard STN EN ISO 4624 (2023) and by the Cross-cut test according to the standard STN EN ISO 2409 (2020).

The testing machine PosiTest AT-M (Qualitest, Canada) was used for the Pull-off test. Small 20 mm diameter dollies were glued to the coating using two-component epoxy resin (Pattex Repair Epoxy). After 24 h of curing at 20 °C and a relative air humidity of 60%, perimeters of glued dollies were carefully incised to prevent the propagation of failures outside the tested area. Pulling was carried at a rate of 1 mm/min up to separating the dolly from the surface. After each test, the fracture was evaluated visually using Tab. 2.

Classification	Nature of the violation
А	Cohesive failure in the substrate
A/B	Adhesion failure between the substrate and the first coating
В	Cohesive failure in the first coating
B/C	Adhesion failure between the first coating and the second coating
n	Cohesive failure of the n <sup>th</sup> coating in a multi-layer system
n/m	Adhesion failure between the n <sup>th</sup> coating and m <sup>th</sup> coating in a multi-layer system
-/Y	Adhesion failure between the top coating and the adhesive
Y	Cohesive failure in the adhesive
Y/Z	Adhesion failure between the adhesive and the metal dolly

Tab. 2 Evaluation of the Pull-off area.

The Cross-cut test was done as follows: a cross hatch pattern was cut through the coating film to the substrate. The adhesion of the coating film was classified according to the standard STN EN ISO 2409 (2020) (Tab. 3). The figures are examples of a cross-cut within each step of the classification. The percentages stated are based on the visual impression given by the pictures and the same percentages will not necessarily be reproduced with digital imaging.

Tab. 3 Evaluation of the Cross-cut area.

Classification	0	1	2	3	4	5
Surface of cross- cut area from which flaking has occurred. (Example for						
six parallel cuts)	none	< 5%	5% - 15%	15% - 35%	35% - 65%	> 65%

## **Statistical Evaluation**

The evaluation of colour changes and selected mechanical properties and adhesion of the coating systems during weathering was performed using an analysis of variance (ANOVA) and post hoc Duncan test with an  $\alpha$  significance value of 0.05. All the statistical analyses were carried out in the software STATISTICA 12 (StatSoft, USA) and MS Excel (Microsoft, USA).

# **RESULTS AND DISCUSSION**

## **Thickness of the Coating Film**

The thickness of the surface finishes was determined using non-destructive method (Tab. 4).

	Film Thickness of coating system (µm)										
	Unwea	thered	Weathered								
	x	SD	x	SD							
Self-healing	164	2	164	4							
Standard	101	3	101	4							

Tab. 4	4 Coating	films	thickness	before	and aft	er 2-ve	ars of r	atural	weathering.	
1 a	+ Coaimg	mins	thickness	DUIDIC	anu anu	u 2-yu	ars or n	aturar	weathering.	

Note: Average  $(\bar{x})$  and standard deviation (SD) were derived from a dataset of twelve values.

The thickness of the coating film plays a crucial role in determining the quality of the surface finish. Studies by Hundhausen *et al.* (2018) and Palija *et al.* (2018) investigated the factors influencing coating film thickness in industrial production, emphasizing the significant impact thickness has on overall surface quality. Furthermore, knowledge of the coating film thickness is essential for determining adhesion properties when applying the cross-cut method.

## **Effect of Weathering on Colour Changes**

The changes in the colour coordinates  $L^*$ ,  $a^*$ , and  $b^*$  of the self-healing and standard coating system due to natural weathering are shown in Tab. 5. The  $a^*$  and  $b^*$  parameters changed more moderately compared to lightness ( $L^*$ ), with a similar pattern of change across all coordinates. The self-healing coating system, containing specialized microcapsules, displayed a change trend comparable to that of the standard coating system, with only slight differences.

	CIE Lab Parameters (-)											
	Unweat	hered	Weath	ered	Statistical Significance							
	x	SD	x SD		p – level							
Self-healing	Self-healing											
$L^*$	96.78	0.21	94.72	0.27	0.000							
a*	-0.67	0.04	-0.58	0.08	0.000							
b*	1.60	0.16	3.18	0.22	0.000							
Standard	Standard											
$L^*$	96.52	0.28	94.09	0.40	0.000							
a*	-0.88	0.07	-0.60	0.09	0.000							
b*	2.07	0.23	3.18	0.27	0.000							

Tab. 5 Colour stability analysis: CIE Lab parameters  $L^*$ ,  $a^*$ , and  $b^*$  of coated systems before and after 2-years of natural weathering.

Note: Average  $(\bar{x})$  and standard deviation (SD) were derived from a dataset of forty-two values. <sup>A</sup> - Duncan's test compares the means between unweathered and weathered groups to identify statistically significant differences between them according to the p – level value.

After two years of weathering, the self-healing coating system showed a slight advantage in colour stability compared to the standard system, with average  $\Delta E^*_{ab}$  values of 2.6 and 2.7, respectively (Fig. 1). This colour difference did not exceed a value of 3.5 (degree 3); according to the five-level scale (Mokrzycki and Tatol, 2011), this indicates that even an unexperienced observer would notice the difference. The primary contributors to the colour

change were the alterations in lightness  $(L^*)$  and, to a lesser degree, in the b<sup>\*</sup> coordinate. These changes led to a slight darkening of the coatings and a very subtle yellowish tone.

The statistical analysis of colour changes  $\Delta L^*$ ,  $\Delta a^*$ ,  $\Delta b^*$  and colour difference  $\Delta E^*_{ab}$  between self-healing and standard coating system, conducted using Duncan's test, the type of coating system revealed a statistically significant effect on colour changes  $\Delta L^*$ ,  $\Delta a^*$ , and  $\Delta b^*$  during weathering (p = 0.000). However, the total colour difference value ( $\Delta E^*_{ab}$ ) was not significantly affected by weathering (p = 0.331). This finding suggests that the self-healing coating system exhibited colour stability due to having the same top coat as the standard coating system.



Fig. 1 Differences in colour coordinates and the total colour difference after 2-years of natural weathering.

The discolouration is among the first visible indicators of degradation, as supported by previous studies (Reinprecht and Pánek, 2015; Kúdela *et al.*, 2017; Jirouš-Rajković and Miklečić, 2021). The colour change of coatings during weathering is linked to physical and chemical changes on the surface of the coating, as well as within its interior and the underlying substrate (Van den Bulcke *et al.*, 2008). While the pigments in coatings serve to block UV radiation, thereby catalysing wood degradation, the resins as organic polymers undergo UV-induced degradation, leading to the breakdown of organic compound bonds. The degradation rate depends on the UV resistance of the polymer used (Williams, 2010). Based on the evaluation of total colour differences, both white coating systems demonstrated superior colour stability following exposure to natural weathering, aligning with the findings of Oberhofnerová *et al.*, (2018). In the study by Yan *et al.*, (2022), the authors found that the Dulux waterborne primer with melamine/rice husk powder-coated shellac microcapsules had also little effect on colour difference after UV aging. They explained this by noting that aging creates bubbles in the primer, and that the use of microcapsules helped reduce their size.

#### **Effect of Weathering on Impact Resistance**

The impact resistance, measured as the penetration diameter, and the subjective evaluation of damage to the self-healing and standard coating system are presented in Tab. 6. No improvement in impact resistance at a drop height of 10 and 20 mm was observed to the naked eye for the self-healing coating system compared to the standard system. The self-

healing coating exhibited limited impact resistance. Visible light cracks on the surface, typically one to two circular cracks around the intrusion was after the impact resistance test at a drop height of 200 mm. This indicates that the self-healing system is less durable, as it is more brittle and prone to cracking than the standard system.

Tab. 6 Degree of change on the surface and diameter of the intrusions before and after 2-years of natural weathering.

	Impact Resistance											
Drop height (mm)	Unweathered							Weathered				
	10	25	50	100	200	400	10	25	50	100	200	400
Self-healing coating sys	Self-healing coating system											
Degree	1(1)	1(1)	1(1)	2(3)	2(4)	3(5)	1(1)	2(2)	2(2)	2(3)	3(5)	3(5)
Diameter (mm)	0	0	1	2	4	4.5	0	2	2	3	6	7
Standard coating system												
Degree	1(1)	1(1)	2(2)	2(2)	2(5)	2(5)	1(1)	2(2)	2(3)	2(3)	3(5)	3(5)
Diameter (mm)	0	0	2	3	4	5	0	2	4	5	6	7

Note: For degree, two classifications of impact resistance on the surface is listed, noted as follows: X(Y). The first one is derived from the visual analyse observed to the naked eye. The second one is based on the microscopic analysis.

Microscopic analysis revealed predominantly circular cracks around the intrusion that were not visible to the naked eye. Cracks were also observed in the self-healing coating system prior to aging at a drop height of 100 mm (Tab. 6). Figure 2 shows the sample surface after the impact resistance test, with these cracks evident at a drop height of 200 mm on surface both before and after two years of natural weathering.



Fig. 2 Sample surface after the impact resistance test with typically circular cracks (marked as white arrows) around the intrusion at the drop height of 200 mm before and after 2-years of natural weathering.

Microscopic analysis revealed visible cracks were also off-site of intrusion. After two years of natural weathering, the number of cracks increased after the impact resistance test. However, the naked eye said they were less pronounced than in the standard coating system. These findings suggest that, while the self-healing coating system is designed to seal microcracks that develop during the aging of exterior treatments, it appears to be more susceptible to cracking under mechanical impacts, such as hail or small rocks. This highlights the potential vulnerability of the self-healing system to mechanical stress.

The impact resistance of coatings increases with coating thickness, but only up to a certain point (Slabejová, 2012; Slabejová *et al.*, 2018). In this study, the self-healing coating system exhibited greater thickness than the standard coating, which supports the cited observation. Hazir and Koc (2019) emphasize that the type of coating system significantly influences impact resistance, as it depends on all components within the paint film. Our findings align with this, showing that the intermediate layer containing microcapsules in the self-healing system affected the impact resistance at specific drop heights. Also, a study by Xia *et al.* (2022) found that microcapsules can enhance impact resistance to a certain extent.

### Effect of Weathering on Adhesion of the Coating Film (Pull-off Test)

As shown in Fig. 3, the both self-healing and standard coating system exhibited similar failure in adhesion to oak wood. Minimal fibre separation occurred on surface of wood, from 5% to10% of the area before and after weathering (failure was classified as A/B). We also confirmed that the interlayer with microcapsules had no effect on the coherence of the coating system, *i.e.*, relationship to the base coat as well as to the top coat. Adhesion was less strong than the cohesion of the coating system.



Fig. 3 Sample surface after the Pull-off test before and after 2-years of natural weathering.

The difference between the standard and self-healing coating systems concerning their adhesion to oak wood was evident (Tab. 7). The adhesion of the coating film prior to the exposure of the self-healing coating system was 23% lower than that of the standard surface treatment system. After weathering, the self-healing coating system demonstrated better adhesion to the substrate in comparison to the standard coating system, thus suggesting that the standard coating system exhibits a higher susceptibility to adhesion failure following prolonged exposure. In contrast, the adhesion of the standard surface treatment system experienced a decline of 32% following exposure. These findings were confirmed by statistical evaluation using variance analysis of the Duncan test (Tab. 7). Grüll et al. (2013) demonstrated that the high film thicknesses contributed to better moisture protection, higher coating durability, and longer maintenance intervals. We assume that the greater thickness of the self-healing coating system ensured comparable adhesion after weathering to before weathering. The effect of weathering of the standard surface treatment system caused a decrease in adhesion. Bilgen (2010) noted that a reduction of the adhesion between the varnish layer and sample layer occurred due to the expansion of the varnish layer due to the weathering.

	Adhesion of the Coating Film (Pull-off Test) (MPa)							
	Before W	eathering	After We	eathering	Statistical Significance*			
	x	SD	x	SD	p-value			
Self-healing	5.74	0.33	5.64	0.13	0.449			
Standard	7.45	0.59	5.06	0.18	0.000			

Tab. 7 Adhesion of the coating film before and after 2-years of natural weathering.

Note: Average  $(\bar{x})$  and standard deviation (SD) were derived from a dataset of twelve values. \* Duncan's test compares the means between self-healing and standard coating system groups to identify statistically significant differences between them according to the p - level value.

The significant impact of surface finish on adhesion was confirmed by Hazir and Koc (2019), Slabejová and Vidholdová (2019), Miklečić *et al.* (2017), and Delpech and Coutinho (2000). Hazir and Koc (2019) demonstrated that the type of coating significantly affects adhesion strength, surface hardness, layer thickness, and response to rapid deformation testing. Self-healing coatings typically use microcapsules that release healing agents upon damage to seal cracks. However, as demonstrated by studies such as Slabejová *et al.* (2020), the integration of these capsules into a coating can lead to reduced adhesion and impact resistance. The encapsulated healing agents may compromise the coating's structural integrity, especially under mechanical loading. This aligns with our findings where the self-healing coating system showed lower impact resistance and adhesion compared to the standard system.

### Effect of Weathering on Adhesion of the Coating Film (Cross-cut test)

Figure 4 shows the adhesion results evaluated by the Cross-cut test. The adhesion of both the unweathered self-healing and the standard coating system coatings on oak wood was rated as grade 2.

As noted in the studies by Yan *et al.* (2021); Xia *et al.* (2022), Wu *et al.* (2023), the coating's inability to encapsulate completely the microcapsules contained in high portions in coatings, may results in reduced adhesion. After two years of weathering, the adhesion of both the self-healing and standard coating on oak wood was rated as grade 2, with comparable performance.



Fig. 4 Adhesion of the coating film (Cross-cut test) before and after 2-years of natural weathering.

A comparison of the two adhesion test methods showed that the Cross-cut test provides less information than the Pull-off test. According to the Cross-cut test, the self-healing coating system did not influence adhesion. The results indicate that the intermediate layer with microcapsules reduced the adhesion of the coating film to the oak wood surface. Our observation that the Pull-off test provides more information than the Cross-cut test is consistent with the findings of Brzozowska *et al.* (2023). They report that the Cross-cut test is less sensitive to small-scale variations in adhesion, whereas the Pull-off test gives a more precise measure of bond strength across different layers. In contrast, the Cross-cut test is better suited for quick assessments of coating quality and adhesion uniformity. However, this test does not capture the more complex interactions between coating layers and substrates.

### CONCLUSION

The study on self-healing and standard coating system for oak wood, including a microcapsule-based intermediate layer in the self-healing coating system, provided essential insights into the effects of natural weathering on coatings' optical and mechanical properties. After weathering, it was observed that:

• The self-healing coating system showed a slight advantage in colour stability compared to the standard system, with average  $\Delta E^*_{ab}$  values of 2.6 and 2.7, respectively. The primary contributors to colour change were shifts in lightness (L<sup>\*</sup>) and, to a lesser extent, the b<sup>\*</sup> coordinate, resulting in subtle darkening and yellowing of the coatings.

- The self-healing coating system exhibited lower impact resistance than the standard system, being more brittle and more prone to cracking.
- The self-healing coating system showed no change in adhesion as measured by the Pull-off test, while the standard coating system exhibited a 32% decrease.
- The self-healing coating system also showed no change in adhesion according to the Cross-cut test.

These findings suggest that while self-healing coatings may offer advantages in colour retention, their use on exterior wood surfaces, especially those subject to mechanical stress, should be approached cautiously. For applications that prioritize both colour stability and structural integrity, further optimization of the microcapsule formulation is recommended to improve adhesion and impact resistance.

### REFERENCES

- Adamčík, L., Kminiak, R., Schmidtová, J., 2023. Measurement of the roughness of the sanded surface of beech wood with the profile measurement software of the Keyence VHX-7000 microscope. Acta Facultatis Xylologiae Zvolen, 65(1), 73–86. https://doi.org/10.17423/afx.2023.65.1.07
- Bilgen, S., 2010. The Effects of Outside Conditions Over Some Characteristics of Varnished Juniper Wood. Institute of Science and Technology, Karabuk University, Karabuk, 48–52.
- Blanchet, P., Pepin, S., 2021. Trends in Chemical Wood Surface Improvements and Modifications: A Review of the Last Five Years. Coatings 11(12), 1514. https://doi.org/10.3390/coatings11121514
- Brzozowska, K., Chowaniec-Michalak, A., Niewiadomski, P., Sadowski, Ł., 2023, Adhesive Properties of Polyurethane Paint Coatings Modified with Multi Walled Carbon Nanotubes for Hardwood Protection. In International Conference on Adhesive Bonding (pp. 41-51). Cham: Springer Nature Switzerland. https://doi.org/10.1007/978-3-031-48363-9\_4
- Bulian, F., Graystone, J., 2009. Wood coatings: Theory and practice. Elsevier.
- Cogulet, A., Blanchet, P., Landry, V., 2018. The multifactorial aspect of wood weathering: a review based on a holistic approach of wood degradation protected by clear coating. BioResources 13(1), 23 p. https://doi.org/10.15376/biores.13.1.Cogulet
- Delpech, M.C., Coutinho, F.M.B., 2000. Waterborne anionic polyurethane and poly(urethaneurea)s: influence of the chain extender on mechanical and adhesive properties. Polymer Testing 19(8), 939–952. https://doi.org/10.1016/S0142-9418(99)00066-5
- Ghosh, S.K., 2006. Functional coatings and microencapsulation: a general perspective. In Functional Coatings: by polymer microencapsulation, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, 1-28. https://doi.org/10.1002/3527608478.ch1
- Grüll, G., Truskaller, M., Podgorski, L., Bollmus, S., De Windt, I., Suttie, E., 2013. Moisture conditions in coated wood panels during 24 months natural weathering at five sites in Europe. Wood Material Science & Engineering 8(2), 95–110. https://doi/full/10.1080/17480272.2013.771212
- Hasanagić, R., Šljivo, U., Fathi, L., Gautam, P., Bahmani, M., Humar, M., 2024. Evaluation of Mechanical Properties and Surface Quality of Wood from Bosnia and Herzegovina Exposed to Outdoor Conditions. Journal of Renewable Materials 12(8), 1417–1431. https://doi.org/10.32604/jrm.2024.052826
- Hazir, E., Koc, K.H., 2019. Evaluation of wood surface coating performance using water based, solvent based and powder coating. Maderas. Ciencia y tecnología 21(4), 467–480. http://dx.doi.org/10.4067/S0718-221X2019005000404
- Hochmańska, P., Mazela, B., Krystofiak, T., 2014. Hydrophobicity and weathering resistance of wood treated with silane-modified protective systems. Drewno: prace naukowe, doniesienia, komunikaty 57(191), 99–110. https://doi.org/10.12841/wood.1644-3985.060.07

- Hundhausen, U., Slabohm, M., Meinlschmidt, P., 2018. Industrial coating of wood cladding: In line control of board temperature, film thickness, and microfoam. Conference: PRA's 11<sup>th</sup> International Wood Caotings Congress. 10 p.
- Chang, Y., Yan, X., Wu, Z., 2023. Application and prospect of self-healing microcapsules in surface coating of wood. Colloid and Interface Science Communications 56, 100736. https://doi.org/10.1016/j.colcom.2023.100736
- Cho, S.H., White, S.R., Braun, P.V., 2009. Self-healing polymer coatings. Advanced Materials 21(6), 645–649. https://doi.org/10.1002/adma.200802008
- ISO 7724–3, 1984. Paints and Varnishes—Colorimetry—Part 3: Calculation of Colour Differences; International Organization for Standardization: Geneva, Switzerland.
- Jirouš-Rajković, V., Miklečić, J., 2021. Enhancing weathering resistance of wood A review. Polymers 13(12), 1980. https://doi.org/10.3390/polym13121980
- Kúdela, J., Štrbová, M., Jas, F., 2017. Influence of accelerated ageing on morphology and wetting of wood surface treated with a modified water-based coating system. Acta Facultatis Xylologiae Zvolen res Publica Slovaca 59(1), 27.
- Lagaňa, R., Svocák, J., Kúdela, J., 2021. A study of the surface properties of a solid wood coating system with self-healing microcapsules. In 9<sup>th</sup> Hardwood Proceedings-Part II; University of Sopron: Sopron, Hungary, 9, 61–67.
- Landry, V., Boivin, G., Schorr, D., Mottoul, M., Mary, A., Abid, L., Carrère, M., Laratte, B., 2023. Recent Developments and Trends in Sustainable and Functional Wood Coatings. Current Forestry Reports 9(5), 319–331. https://doi.org/10.1007/s40725-023-00195-0
- Liu, X.Y., Timar, M.C., Varodi, A.M., Sawyer, G., 2017. An investigation of accelerated temperature-induced ageing of four wood species: colour and FTIR. Wood science and technology 51, 357–378. https://doi.org/10.1007/s00226-016-0867-4
- Maiti, T. K., Dixit, P., Suhag, A., Bhushan, S., Yadav, A., Talapatra, N., Chattopadhyay, S., 2024. Advancements in organic and inorganic shell materials for the preparation of microencapsulated phase change materials for thermal energy storage applications. RSC Sustainability 1(4), 665–697. https://doi.org/10.1039/D2SU00116K
- Miklečić, J., Turkulin, H., Jirouš-Rajković, V., 2017. Weathering performance of surface of thermally modified wood finished with nanoparticles-modified waterborne polyacrylate coatings. Applied Surface Science 408, 103–109. https://doi.org/10.1016/j.apsusc.2017.03.011
- Mokrzycki, W., Tatol, M., 2011. Color difference Delta E A survey. Machine Graphics and Vision 20(4), 383–411.
- Oberhofnerová, E., Hýsek, Š., Pánek, M., Böhm, M., 2018. Effect of artificial weathering and temperature cycling on the performance of coating systems used for wooden windows. Journal of Coatings Technology and Research 15, 851–865.https://doi.org/10.1007/s11998-017-0033-4
- Palija, T., Jaić, M., Džinčić, I., Šućur, A., Dobić, J., 2018. Variability of dry film thickness of a coating applied by roller coater on wood in a real industrial process. Drewno 61(201), 153–164. https://doi.org/10.12841/wood.1644-3985.251.13
- Reinprecht, L., Pánek, M., 2015. Effects of wood roughness, light pigments, and water repellent on the color stability of painted spruce subjected to natural and accelerated weathering. BioResources 10(4), 7203–7219.
- Queant, C., Blanchet, P., Landry, V., Schorr, D., 2018. Effect of Adding UV Absorbers Embedded in Carbonate Calcium Templates Covered with Light Responsive Polymer into a Clear Wood Coating. Coatings 8, 265. https://doi.org/10.3390/coatings8080265
- Samadzadeh, M., Boura, S.H., Peikari, M., Kasiriha, S.M., Ashrafi, A., 2010. A review on selfhealing coatings based on micro/nanocapsules. Progress in Organic Coatings 68(3), 159–164. https://doi.org/10.1016/j.porgcoat.2010.01.006
- Schreiner, C., Scharf, S., Stenzel, V., Rössler, A., 2017. Self-healing through microencapsulated agents for protective coatings. Journal of Coatings Technology and Research 14, 809–816. https://doi.org/10.1007/s11998-017-9921-x

- Slabejová, G., 2012. Vplyv vybraných faktorov na stabilitu systému drevo tuhý náterový film [Influence of selected factors on the stability of the wood-solid paint film system]. Acta Facultatis Xylologiae Zvolen res Publica Slovaca 54(2), 57–65.
- Slabejová, G., Šmidriaková, M., Pánis, D., 2018. Quality of silicone coating on the veneer surfaces. BioResources (13)1, 776–788. https://doi.org/10.15376/biores.13.1.776-788
- Slabejová, G., Vidholdová, Z., Šmidriaková, M., 2019. Surface finishes for thermally modified beech wood. Acta Facultatis Xylologiae Zvolen res Publica Slovaca 61(2), 41–50. https://doi.org/10.17423/afx.2019.61.2.04
- Slabejová, G., Šmidriaková, M., Svocák, J., 2020. Interlayer with microcapsules and its influence on the surface finish quality of wood. Acta Facultatis Xylologiae Zvolen res Publica Slovaca 62(2), 61–74. https://doi.org/10.17423/afx.2020.62.2.06
- STN EN 927–3, 2013. Paints and varnishes Coating materials and coating systems for exterior wood Part 3: Natural weathering test. Slovak Office of Standards, Metrology and Testing, Bratislava, Slovakia.
- STN EN ISO 4624, 2023. Paints and varnishes. Pull-off test for adhesion. Slovak Office of Standards, Metrology and Testing, Bratislava, Slovakia.
- STN EN ISO 2409, 2020. Paints and varnishes. Cross-cut test. Slovak Office of Standards, Metrology and Testing, Bratislava, Slovakia.
- STN EN ISO 6272–2, 2011. Paints and varnishes Rapid-deformation (Impact resistance) tests Part 2: Falling-weight test, small-area indenter. Slovak Office of Standards, Metrology and Testing, Bratislava, Slovakia.
- Suryanarayana, C., Rao, K.C., Kumar, D., 2008. Preparation and characterization of microcapsules containing linseed oil and its use in self-healing coatings. Progress in organic coatings 63(1), 72–78. https://doi.org/10.1016/j.porgcoat.2008.04.008
- Yan, X., Peng, W., 2020. Preparation of Microcapsules of Urea Formaldehyde Resin Coated Waterborne Coatings and Their Effect on Properties of Wood Crackle Coating. Coatings 10, 764. https://doi.org/10.3390/coatings10080764
- Yan, X., Zhao, W., Wang, L., Qian, X. 2021. Effect of Microcapsule Concentration with Different Core-Shell Ratios on Waterborne Topcoat Film Properties for *Tilia europaea*. Coatings 11, 1013. https://doi.org/10.3390/coatings11091013
- Yan, X., Li, W., Han, Y., Yin, T., 2022. Preparation of Melamine/Rice Husk Powder Coated Shellac Microcapsules and Effect of Different Rice Husk Powder Content in Wall Material on Properties of Wood Waterborne Primer. Polymers 14, 72. https://doi.org/10.3390/polym14010072
- Van den Bulcke, J., Van Acker, J., Stevens, M., 2008. Experimental and theoretical behavior of exterior wood coatings subjected to artificial weathering. Journal of Coatings Technology and Research 5, 221-231. https://doi.org/10.1007/s11998-007-9074-4
- Vidholdová, Z., Slabejová, G., Kaloč, J., 2017. Influence of wood pre-weathering on selected surface properties of the system wood–coating film. Acta Facultatis Xylologiae Zvolen res Publica Slovaca 59(2), 67–77.
- Viitanen, H., Ritschkoff, A.C., 2011. Coating and surface treatment of wood. In Fundamentals of mold growth in indoor environments and strategies for healthy living, Wageningen Academic. 463–488. https://doi.org/10.3920/9789086867226\_018
- Wang, L., Han, Y., Yan, X., 2022. Effects of adding methods of fluorane microcapsules and shellac resin microcapsules on the preparation and properties of bifunctional waterborne coatings for basswood. Polymers 14(18), 3919. https://doi.org/10.3390/polym14183919
- Williams, R.S., 2010. Finishing of Wood. In: Wood Handbook-Wood as an Engineering Material. General Technical Report FPL-GTR-190, US Department of Agriculture, Forest Service, Forest Products Laboratory, Madison.
- White, S.R., Sottos, N.R., Geubelle, P.H., Moore, J.S., Kessler, M.R., Sriram, S.R., Brown, E.N, Viswanathan, S., 2001. Autonomic healing of polymer composites. Nature 409, 794–797. https://doi.org/10.1038/35057232
- Wu, Q., Li, W., Yan, X., 2023. Effect of Microcapsules on Mechanical, Optical, Self-Healing and Electromagnetic Wave Absorption in Waterborne Wood Paint Coatings. Coatings 13, 1478. https://doi.org/10.3390/coatings13091478

- Xia, L., Han, Y., Yin, T., Zhu, Y., Yan, X., Li, J., 2024. Effects of Mixed Microcapsules in Different Proportions on Aging Resistance and Self-Healing Properties of Waterborne Coatings for *Tilia europaea* L. Coatings 14, 1042. https://doi.org/10.3390/coatings14081042
- Xia, Y., Yan, X., Peng, W., 2022. Preparation of Cellulose Modified Wall Material Microcapsules and Its Effect on the Properties of Wood Paint Coating. Polymers 14, 3534. https://doi.org/10.3390/polym14173534
- Zhao, J., Zhang, W., Liao, L., Wang, S., Li, W., 2012. Self-healing coatings containing microcapsule. Applied Surface Science 258(6), 1915–1918. https://doi.org/10.1016/j.apsusc.2011.06.154

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## **AUTHORS' ADDRESSES**

Ing. Zuzana Vidholdová, PhD. Technical University in Zvolen Faculty of Wood Sciences and Technology, Department of Wood Technology T.G. Masaryka 24, 960 01 Zvolen zuzana.vidholdova@tuzvo.sk

Ing. Gabriela Slabejová, PhD. Technical University in Zvolen Faculty of Wood Sciences and Technology, Department of Furniture and Wood Products T.G. Masaryka 24, 960 01 Zvolen slabejova@tuzvo.sk

prof. Ing. Jozef Kúdela, CSc. Ing. Ján Svocák, PhD. Technical University in Zvolen Faculty of Wood Sciences and Technology, Depatment of Wood Science T.G. Masaryka 24, 960 01 Zvolen kudela@tuzvo.sk jsvocak@yahoo.com