WOOD RESEARCH doi.org/10.37763/wr.1336-4561/66.4.544555
e-ISSN 2729-8906
66(4): 2021 544-555 pp.

COLOUR STABILITY OF STEAMED BLACK LOCUST, BEECH AND SPRUCE TIMBERS DURING SHORT-TERM PHOTODEGRADATION

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(RECEIVED JANUARY 2021)

ABSTRACT

Black locust (*Robinia pseudoacacia* L.), beech red heartwood (*Fagus sylvatica* L.) and spruce (*Picea abies* Karst.) wood samples were treated in saturated steam at 100, 110 and 120°C then irradiated using a UV emitter mercury lamp in order to test their colour stability. Colour change was evaluated and presented in the CIE Lab colour coordinate system. Untreated black locust, beech and spruce specimens as control samples were irradiated using the same mercury lamp. Results revealed that beech produced the greatest colour stability during both steam treatment and the following UV treatment while spruce was the most sensitive species to photodegradation. Steaming reduced the colour change intensity only for black locust during photodegradation. Both redness and yellowness change demonstrate this colour stability increase. Steaming at 120°C resulted in the greatest protection against the colour alteration of black locust caused by photodegradation. The investigated thermal treatments did not change the photodegradation properties of beech and spruce specimens. A considerable increase in colour saturation of the specimens was generated by steaming, and the saturation value further increased during the UV treatment.

KEYWORDS: Steaming, UV irradiation, colour change, extractives.

INTRODUCTION

Industrial scale steam treatment of wood with the aim of colour alteration was started as early as the second half of the last century. Nevertheless, systematic research to discover the specific effects of steaming parameters has been carried out only for the last three decades. In the frame of industrial scale steam treatments, black locust and beech timbers are involved in the largest quantities. Steaming practice demonstrated that steaming of beech is uncomplicated while steaming of black locust is rather difficult due to its peculiar mechanical and anatomical properties such as the extremely high extractive content. This observation determined that steaming of black locust was the most deeply investigated process (Horváth-Szováti 2000, Tolvaj et al. 2000, 2010, Tolvaj and Molnar 2006, Varga and Van der Zee 2008, Dzurena 2018a). At the same time, steaming behaviour of beech was also widely investigated (Tolvaj et al. 2009, Dzurenda 1013, Milić et al. 2015, Geffert et al. 2017 Dzurenda and Dudiak 2020).

There are few publications regarding the steaming properties of wood species other than beech and black locust. The possible colour variations of cherry wood generated by steam treatment were investigated by Straze and Gorisek (2008) and by Dianiskova et al. (2008). The steaming can reduce the great colour difference between sapwood and heartwood of Turkey oak (Tolvaj and Molnar 2006, Todaro et al. 2012, Csanady et al. 2015). Recently the colour change of poplar (*Populus x euramericana cv. pannonia*) by steaming was investigated to obtain attractive colour suitable for various indoor applications (Banadics and Tolvaj 2019). Steaming was found to be a proper technique to turn the naturally unattractive colour of poplar wood to a pleasant brown colour. Steam was able to double the colour saturation, which is a significant result from the perspective of industrial applications. The treatment increased both redness and yellowness values and reduced the lightness. Steaming behaviour of oak and maple wood was investigated by Dzurenda (2017, 2018b, Dzurenda et al. 2020) for getting attractive brown colour. Geffert et al. (2020) studied the colour change and chemical changes of birch wood during steaming in the 105-135°C temperature interval. Steaming properties of conifers (Scots pine, spruce, larch, sugi) were investigated in the last decade (Tolvaj et al. 2012, 2019, Preklet et al. 2019). Although, they are characterized with a rather low extractive content, depending on the applied steaming parameters, a variety of colours could be created from the initial yellowish to the modified light brown colour. These new colours were similar to the colours of aged indoor wooden constructions and furniture thus providing a fully natural alternative to chemical surface colour modification.

Objective colour measurement helps the researchers to perform exact and detailed investigation in the field of colour modification of wood. This measurement method has widely been applied in the recent past in wood research (Mitsui et al. 2001, 2004, Oltean et al. 2010, Zivkovic et al. 2014). Objective colour co-ordinates allow us to describe the behaviour of different wood species during steam treatment.

Natural wood is sensitive to weathering. Sun radiation is the main factor that induces wood photodegradation primarily through the decomposition of lignin. Although, photodegradation of natural wood is a widely studied phenomenon (Pandey 2005, Popescu et al. 2011, Persze and Tolvaj 2012, Denes and Lang 2013, Zivkovic 2013, Timar et al. 2016, Varga et al. 2017), only two papers were found dealing with the photodegradation behaviour of steamed wood. The effect of UV radiation on native and steamed maple wood was presented in a conference paper (Dzurenda et al. 2020). The change in the lightness coordinate L* was opposite for native and steamed wood. While native maple wood darkened, steamed maple wood faded. Other researchers claimed that steaming enhanced the redness stability of poplar wood against UV irradiation and slightly reduced the yellow colour sensitivity to photodegradation (Banadics et al. 2019).

Steam treated timber with an attractive brown colour is favourable used for outdoor applications. Therefore, it is considered to be important to gain information on the stability of this modified colour. The aim of this study was to discover the colour stability of steamed wood species with different extractive content during photodegradation.

MATERIALS AND METHODS

Black locust (*Robinia pseudoacacia* L.), beech red heartwood (*Fagus sylvatica* L.) and spruce (*Picea abies* Karst.) specimens were used for the tests. The specimen size was 150 × 25 × 15 mm (L × R × T). The radial surface was used for colour measurement. Initial moisture content of the specimens was between 9 and 10% before steaming. The steaming was carried out in an autoclave to keep the pressure generated by the steam. Wood specimens were placed in the autoclave with distilled water at the bottom for conditioning the air to maintain maximum relative humidity. The chosen steaming temperatures were 100°C, 110°C, and 120°C, the steaming time was 2 days.

Steamed samples were dried at room temperature up to the initial weight to generate equal moisture content for all colour measurements. Steam treated specimens were subjected to photodegradation together with the thermally untreated control specimens. UV light emitter mercury vapour lamp provided the light irradiation. The UV radiation was 80% of the total emissions (31% UV-A, 24% UV-B, and 25% UV-C). The total electric power of the applied double mercury lamps was 800 W, and the distance between the specimens and the light source was 64 cm. The light power density was 76 $J \cdot m^{-2}s^{-1}$ on the surface of the specimens. The irradiation chamber set for 60°C guaranteed ambient temperature conditions. The total irradiation time was 90 h. The irradiation was interrupted after 7, 16, 36, 60, and 90 h to measure the colour change. Colour measurements were carried out with a colorimeter (Konica-Minolta 2600d). The CIE L*, a*, and b* colour coordinates were calculated by applying the D65 illuminant and 10° standard observer with a test-window diameter of 8 mm. The colour of ten randomly chosen dots were measured and averaged for each specimen.

RESULTS AND DISCUSSION

The investigated species were chosen because of their diverse extractive content. Black locust has high extractive content covering 5-9% The main component is dihydrorobinetin covering 2-5% (Molnar and Bariska 2002). The main extractive components of black locust heartwood are the flavonoids which give 89% of the total extractive content. Within flavonoids, dihydrorobinetin is the main component covering 58% of total flavonoid content. Robinetin content covers 14% of the total flavonoid content (Sanz et al. 2011). Extractive content of beech is between 3 and 5%, while that of spruce is 2-3%. Extractive content is important because the colour of wood species is determined mainly by this chemical constituent. Most of the extractives and the hemicelluloses are sensitive to thermal treatment. Results of the thermal treatments are the darkening and the shift of the hue towards brown. The lightness change of black locust is presented in Fig. 1 generated by the applied steaming and UV treatment. The steaming caused great lightness intensity decrease which was determined by the steaming temperature. The two-day steaming produced 20%, 38% and 54% lightness decrease generated by 100°C, 110°C and 120°C steaming temperatures, respectively. The same data for spruce (Fig. 2) were 14%, 19% and 24%. The beech showed different behaviour. The lightness change was slightly temperature dependent and the maximum lightness decrease was 15%. The reason might be that the two-day steaming was too long for beech. In industrial practice, the steaming time for beech is maximum one day. Steam generated chromophore molecules may suffer further degradation during the relatively long steaming period. The two-day steaming period was chosen as an average. (It was long for beech and short for spruce.)

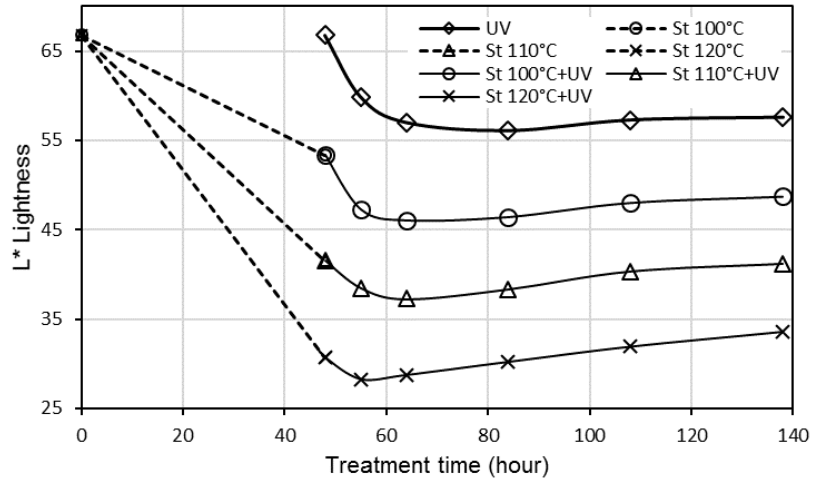

Fig. 1: Lightness change of unsteamed (UV) and steamed (St) black locust specimens caused by steaming and UV irradiation.

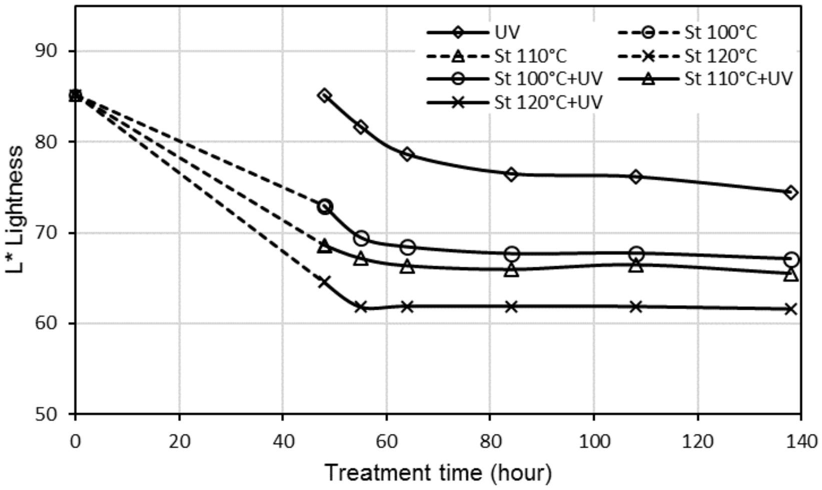

Fig. 2: Lightness change of unsteamed (UV) and steamed (St) spruce specimens caused by steaming and UV irradiation.

All of the unsteamed control specimens showed intensive lightness decrease during the first 16 hours of the UV treatment. The lightness hardly changed after this irradiation period except in the case of spruce specimens where a slow but continuous lightness decrease was observed in the 64-138-hour treatment period (Fig. 2). Steam treated specimens showed similar lightness change as the unsteamed specimens during UV irradiation. The decrease was smaller at the beginning of the UV irradiation and also the period of the decrease became shorter. The lightness value of steamed black locust increased after 10-hour UV irradiation. The UV irradiation caused small lightening for the specimens steamed at 120°C compared to the lightness generated by the steaming. The lightness of beech and spruce samples remained constant in the 16-90-hour UV irradiation period. Results demonstrated that the steamed wood specimens were less sensitive to the darkening effect of photodegradation than the unsteamed ones.

The applied steam treatments resulted in substantial redness increase in the case of back locust and spruce specimens (Figs. 3-4). The two-day steaming produced 71%, 52% and 63% redness value increase generated by 100°C, 110°C and 120°C steaming temperatures, respectively. The same data for spruce were 153%, 172% and 181%. The redness increase of spruce followed the trend of temperature increase while no such rule could be observed in the case of black locust. Most probably this is because steam generated chromophore chemical groups in this wood species are highly sensitive to thermal degradation (Tolvaj et al. 2010). It was found that the redness of black locust timber tends to decrease after one-day steaming if the temperature is above 100°C. That is why the steaming at 100°C caused bigger redness increase than at 110°C and 120°C. The beech showed moderate redness increase during steaming and the change was hardly temperature dependent. The largest redness increase was 22% produced by steaming at 100°C.

The UV irradiation generated rapid redness increase in unsteamed black locust specimens during the first 7 hours of irradiation and the increase was moderate afterward (Fig. 3).

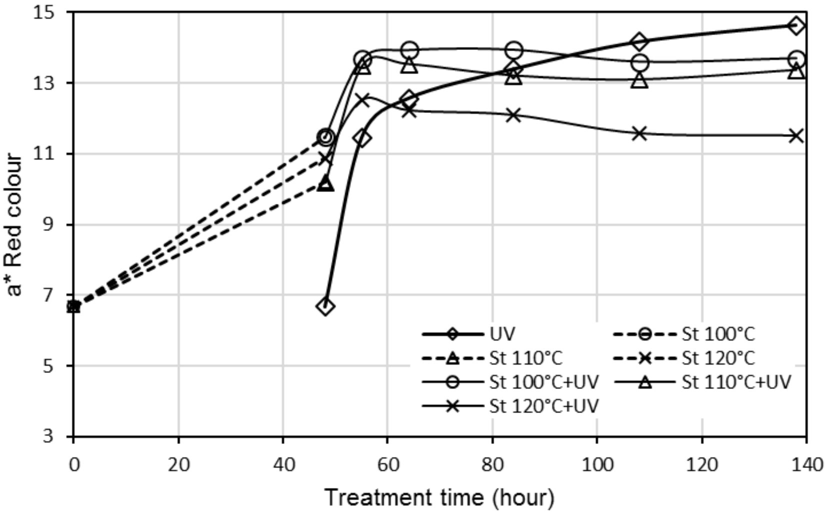

Fig. 3: Redness change of unsteamed (UV) and steamed (St) black locust specimens caused by steaming and UV irradiation.

Unsteamed beech and spruce specimens showed almost linear red colour increase during the whole 90-day UV irradiation period. Red colour values of steamed beech specimens were equal in the 84-138-hour treatment interval (UV) independently of the previous steaming process. Steamed spruce specimens presented similar photodegradation behaviour as beech regarding redness increase, with the difference that the dots representing the red colour of spruce were far from each other after steaming. Consequently, the functions of redness values were parallel (Fig. 4). The only exception was the redness change of specimens steamed at 120°C. The redness barely changed in the 48-84-hour treatment period and moved parallel to the other lines afterwards. It can be concluded that steaming improved the redness stability only in the case of black locust. Steaming at 120°C provided the most stable red colour against UV irradiation.

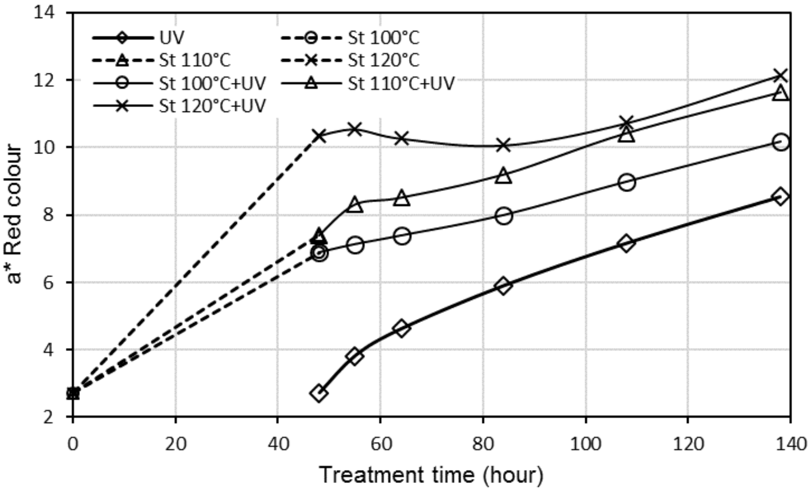

Fig. 4: Redness change of unsteamed (UV) and steamed (St) spruce specimens caused by steaming and UV irradiation.

The applied steam treatments reduced slightly the yellowness of black locust (Fig. 5). Beech specimens also suffered moderate yellowness intensity change but unlike black locust it was a temperature-independent increase (Fig. 6). Spruce specimens suffered considerable yellowness increase. The two-day steaming induced 40%, 70% and 76% yellowness value increase at 100°C, 110°C and 120°C steaming temperatures, respectively.

The UV irradiation caused rapid yellowness intensity increase during the first 7 hours of irradiation, followed by slow and linear increase afterward on the surface of all unsteamed control specimens. Comparing steamed and unsteamed black locust samples, the applied steaming reduced considerably the yellow colour coordinate increase generated by UV irradiation (Fig. 5). Moreover, steam treatment at 120°C was able to hinder further yellow colour change, so that the UV irradiation hardly influenced the yellow colour of black locust. The results showed that steaming improved the yellowness stability of black locust against UV irradiation. The yellowing effect of UV irradiation was somewhat smaller for steamed beech specimens than for unsteamed ones (Fig. 6). UV light induced yellow colour change of steamed spruce was similar to that of beech with the only difference in the starting point distribution (generated by steaming). The biggest distance between the yellowness values of unsteamed and steamed specimens (before UV irradiation) was 34 and 4 units for spruce and beech, respectively. As a consequence, trend lines were parallel and were farther to each other for spruce than for beech. These results demonstrate that preliminary steam treatment does not influence the UV irradiation induced yellow colour shift of beech and spruce specimens. UV irradiation test results showed that steaming improved the yellow colour stability of black locust. The yellowing of wood during UV irradiation is generated mainly by the degradation of lignin. Previous study demonstrated that the high extractive content of black locust partly protected the lignin against UV degradation (Tolvaj and Varga 2012). Present results strengthen this finding as well. Unsteamed black locust, beech and spruce specimens produced 10, 14 and 23 units of yellowness increase during 90-hour UV irradiation, respectively. The order of the extractive content of the investigated species is the opposite of the order of yellowness increase. Fig. 5 demonstrates that the steaming modified new extractives are also able to protect the lignin against photodegradation. To verify this finding further chemical investigations need to be done.

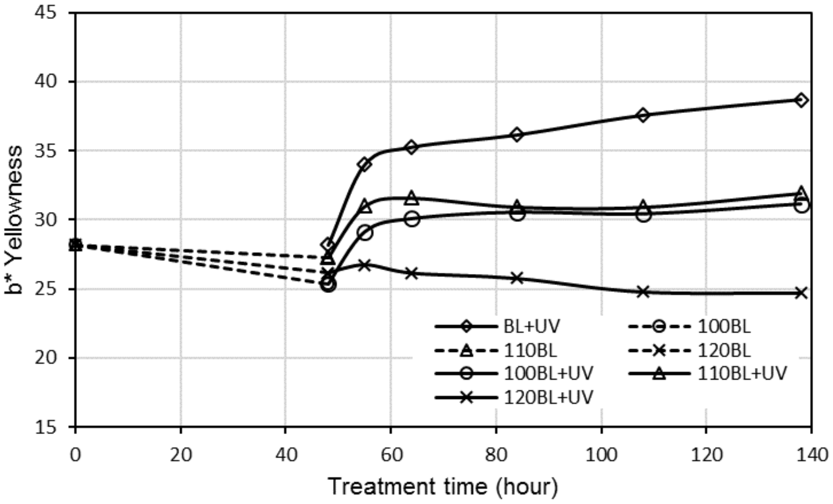

Fig. 5: Yellowness change of unsteamed (UV) and steamed (St) black locust specimens caused by steaming and UV irradiation.

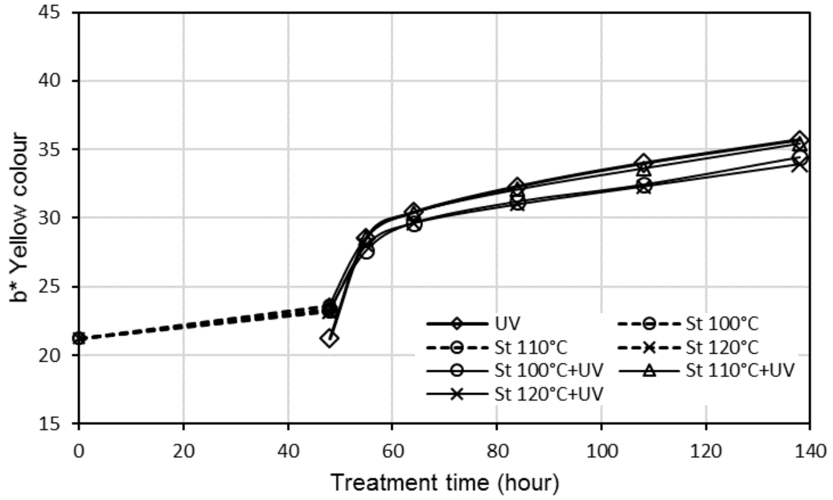

Fig. 6: Yellowness change of unsteamed (UV) and steamed (St) beech specimens caused by steaming and UV irradiation.

Shifting of colour coordinates on the a*-b* plane visualise the combined effect of steaming and UV irradiation (Figs. 7-9). This type of presentation gives the possibility to compare redness and yellowness changes. Dotted lines demonstrate the colour change caused by steaming (start and end points only), while solid lines represent the colour modification effects of the UV irradiation. The distance from the origin (0, 0) on the a*-b* plane provides the saturation value of the individual colour dots. Figs. 7-9 demonstrate that both steaming and UV irradiation increased the saturation value considerably. The hue angle altered between 77-65°, 73-67° and 83-73° for black locust, beech and spruce, respectively. The hue alteration of black locust was twice as large as that of beech. This large hue change was generated by the steaming. The visual colour changed from the greyish yellow to chocolate brown. This alteration reflects the high sensitivity of the extractives in black locust to thermal degradation. The size of the diagram area covered by the presented colour dots represents the colour stability of the tested specimens. Small occupied area shows good colour stability. Among the investigated species, beech presented the highest colour stability against both steaming and UV irradiation (Fig. 8). Steaming did not change the sensitivity of beech to UV irradiation. Trend lines of steamed and unsteamed specimens were parallel close to each other during UV irradiation.

Unsteamed black locust produced moderate yellowness change and intensive redness value increase during the treatments. Fig. 7 presents clearly that the steaming improved the colour stability of black locust against UV treatment.

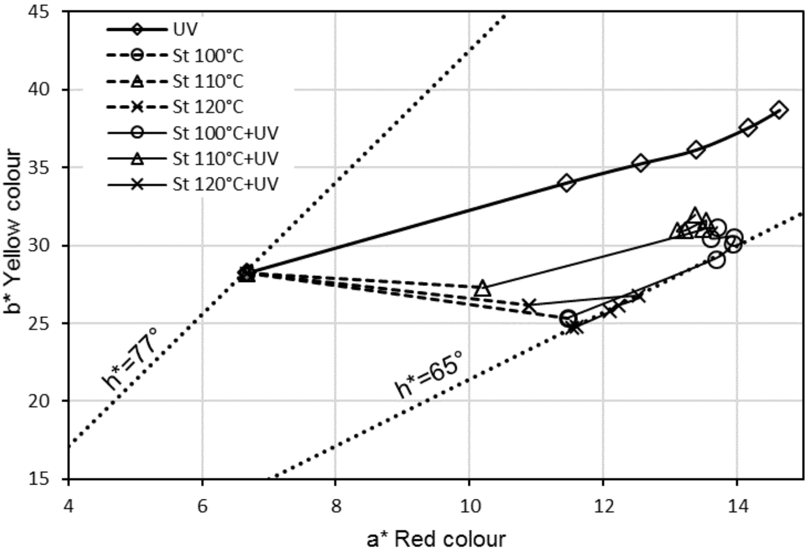

Fig. 7: Demonstration of the colour shift of black locust on the red colour (a) - yellow colour (b*) plane generated by steaming (St) and UV light irradiation.*

The colour parameters hardly changed after 7-hour UV irradiation. The colour of spruce specimens was sensitive to both steaming and UV irradiation (Fig. 9). Out of the three investigated wood species, spruce suffered the largest colour change generated by both steaming and UV irradiation. Unsteamed spruce specimens showed the highest sensitivity to UV irradiation. The low extractive content of this species was unable to reduce the effect of photodegradation. Steaming did not affect this sensitivity.

Results show that beech provided the highest colour stability during the treatments. Steaming reduced the photodegradation intensity only in the case of black locust. Spruce was the most sensitive species to photodegradation. Results strengthen the important role of extractives in photodegradation. The distance from the origin (0, 0) on the a*-b* plane provides the saturation value of the individual colour dots. Figs 7-9 demonstrate that both steaming and

UV irradiation elevated the saturation value considerably. Spruce produced the greatest and beech the smallest saturation increase.

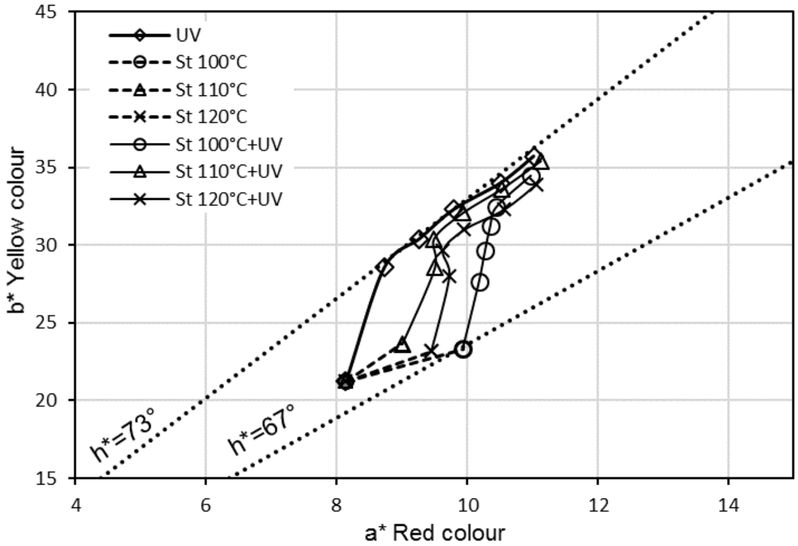

Fig. 8: Demonstration of the colour shift of beech on the red colour (a) - yellow colour (b*) plane generated by steaming (St) and UV light irradiation.*

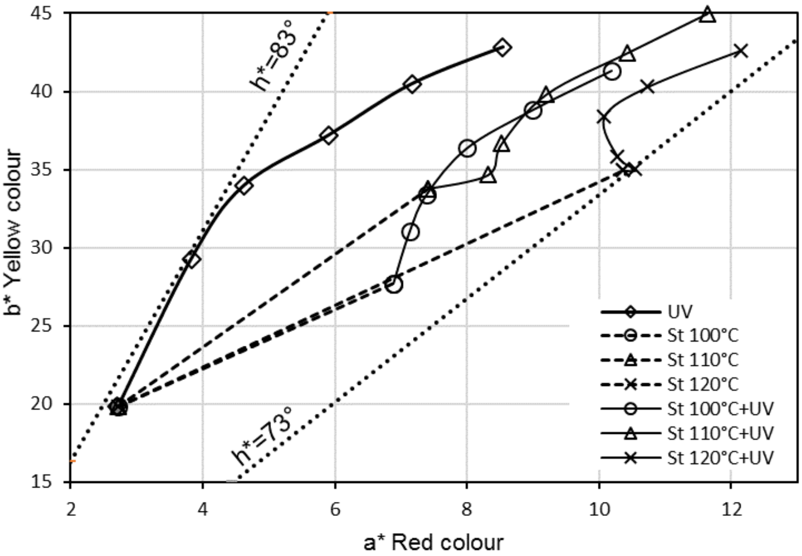

Fig. 9: Demonstration of the colour shift of spruce on the red colour (a) - yellow colour (b*) plane generated by steaming (St) and UV light irradiation.*

CONCLUSIONS

Untreated and steamed black locust, beech and spruce specimens were irradiated by UV emitter mercury lamp to test the colour stability of wood species featuring different extractive content. Colour changes were monitored by objective colour measurements and the colour data were presented in the CIE Lab system. Results revealed that beech had the greatest colour stability during the treatments while spruce was the most sensitive species to photodegradation. Steaming reduced the UV light induced colour change intensity only in the case of black locust imparting a higher colour stability thanks to its high extractive content. Results also demonstrated that the steamed wood specimens were less sensitive to the darkening effect of photodegradation than the unsteamed ones.

Steaming reduced the sensitivity of black locust against UV irradiation concerning red colour shift. In contrast, the redness change was similar for both unsteamed and steamed beech and spruce specimens.

High extractive content of black locust was able to reduce the intensity of lignin degradation during UV irradiation monitored by the yellowness change. Steam generated extractives also showed this protecting effect in treated black locust specimens. Steaming at 120°C provided the most efficient protection.

Steaming generated considerable increases in colour saturation of the specimens, and the saturation value increased further during the UV treatment.

ACKNOWLEDGEMENT

This article was made in frame of the „EFOP-3.6.1-16-2016-00018 – Improving the role of research+development+innovation in the higher education through institutional developments assisting intelligent specialization in Sopron and Szombathely”.

REFERENCES

1. Banadics, E.A., Tolvaj, L., 2019: Colour modification of poplar wood by steaming for brown colour. European Journal of Wood and Wood Products 77: 717–719.
2. Csanady, E., Magoss, E., Tolvaj, L., 2015: Quality of machined wood surfaces. Pp 171-173, Springer.
3. Denes, L., Lang, E.M., 2013: Photodegradation of heat treated hardwood veneers. Journal of Photochemistry and Photobiology B: Biology 118: 9-15.
4. Dianisková, M., Babiak, M., Tolvaj, L., 2008: Colour homogenisation of cherrywood (*Cerasus avium* L.) and black locust (*Robinia pseudoacacia* L.) during steaming. Wood Research 53(4): 45-58.
5. Dzurenda, L., 2013: Modification of wood colour of *Fagus sylvatica* L to a brown-pink shade caused by thermal treatment. Wood Research 58(3): 475-482.
6. Dzurenda, L., 2017: Modification of wood colour of *Acer platanoides* L. to a brown-red shade caused by thermal treatment. Forestry and Wood Technology 98: 26-32.
7. Dzurenda, L., 2018a: Colour modification of *Robinia pseudoacacia* L. during the processes of heat treatment with saturated water steam. Acta Facultatis Xylologiae Zvolen 60(1): 61−70.
8. Dzurenda, L., 2018b: The shades of color of *Quercus robur* L. wood obtained through the processes of thermal treatment with saturated water vapour. BioResources 13(1): 1525-1533.
9. Dzurenda, L., Geffert, A., Geffertova, J., Dudiak, M., 2020: Evaluation of the process thermal treatment of maple wood saturated water steam in terms of change of pH and color of wood. BioResources 15(2): 2550-2559.

10. Dzurenda, L., Dudiak, M., 2020: Changes in wood tree species *Fagus sylvatica* L. and characteristics of the thermal process of modifying its color with saturated water steam. Advances in Ecological and Environmental Research 5(04): 142-156.
11. Dzurenda, L., Dudiak, M., Banski, A., 2020: Influence of UV radiation on color stability of natural and thermally treated maple wood with saturated water steam. In: Tenth international scientific and technical conference "Innovations in Forest Industry and Engineering Design" – INNO 2020, 1–3 October 2020. Pp 219-224, Vitosha park hotel, Sofia, Bulgaria.
12. Geffert, A., Vybohová, E., Geffertová, J., 2017: Characterization of the changes of colour and some wood components on the surface of steamed beech wood. Acta Facultatis Xylologiae Zvolen 59(1): 49−57.
13. Geffert, A., Geffertova, J., Vybohova, E., Dudiak, M., 2020: Impact of steaming mode on chemical characteristics and colour of birch wood. Forests 11(4): 478, 12 pp.
14. Horváth-Szováti, E., 2000: A gőzölt akác világosság-változásának hőmérséklet- és időfüggése (Temperature and time dependence of lightness change of steamed black locust) (in Hungarian with English and German abstracts). SE Tudományos Közleményei 46: 179-189.
15. Milić, G., Todorović, N., Popadić, R., 2015: Influence of steaming on drying quality and colour of beech timber. Glasnik Šumarskog Fakulteta 83-96.
16. Mitsui, K., Takada, H., Sugiyama, M., Hasegawa, R., 2001: Changes in the properties of light-irradiated wood with heat treatment. Part 1. Effect of treatment conditions on the change in color. Holzforschung 55: 601-605.
17. Mitsui, K., Murata, A., Tsuchikawa, S., Kohara, M., 2004: Wood photography using light irradiation and heat treatment. Color Research Applications 29: 312-316.
18. Molnar, S., Bariska, M., 2002: Wood species of Hungary. Szaktudás Kiadó Ház, Budapest.
19. Oltean, L., Hansmann, C., Nemeth, R., Teischinger, A., 2010: Wood surface discolouration of three Hungarian hardwood species due to simulated indoor sunlight exposure. Wood Research 55: 49-58.
20. Pandey, K. K., 2005: Study of the effect of photo-irradiation on the surface chemistry of wood. Polymer Degradation and Stability 90: 9-20.
21. Persze, L., Tolvaj, L., 2012: Photodegradation of wood at elevated temperature: Colour change. Journal of Photochemistry and Photobiology B: Biology 108: 44-47.
22. Popescu, C.M., Popescu, M.C., Vasile, C., 2011: Structural analysis of photodegraded lime wood by means of FT-IR and 2D IR correlation spectroscopy. International Journal of Biological Macromolecules 48: 667-675.
23. Preklet, E., Tolvaj, L., Banadics, E.A., Alpar, T., Varga, D., 2019: Colour modification and homogenisation of larch wood by steaming. Wood Research 64(5): 811-820.
24. Sanz, M., Fernández de Simon, B., Esteruelas, E., Munoz, A.M., Cadahia, E., Hernández, T., Estrella, I., Pinto, E., 2011: Effect of toasting intensity at cooperage on phenolic compounds in acacia (*Robinia pseudoacacia*) heartwood. Journal of Agricultural and Food Chemistry 59: 3135–3145.

25. Straze, A., Gorisek, Z., 2008: Research on colour variation of steamed cherry wood (*Prunus avium* L.). Wood Research 52(2): 77-90.
26. Todaro, L., Zuccaro, L., Marra, M., Basso, B., Scopa, A., 2012: Steaming effects on selected wood properties of Turkey oak by spectral analysis. Wood Science Technology 46(1-3): 89-100.
27. Tolvaj, L., Horváth-Szováti, E., Safar, C., 2000: Colour modification of black locust by steaming. Wood Research 45(2): 25-32.
28. Tolvaj, L., Molnár S., 2006: Colour homogenisation of hardwood species by steaming, Acta Silvatica et Lignaria Hungarica 2: 105-112.
29. Tolvaj, L., Nemeth, R., Varga, D., Molnar S., 2009: Colour homogenisation of beech wood by steam treatment. Drewno 52: 5-17.
30. Tolvaj, L., Molnár, S., Németh, R., Varga, D., 2010: Colour modification of black locust depending on the steaming parameters. Wood Research 55(2): 81-88.
31. Tolvaj, L., Papp, G., Varga, D., Lang, E., 2012: Effect of steaming on the colour change of softwoods. BioResources 7(3): 2799-2808.
32. Tolvaj, L., Banadics, E.A., Tsuchikawa, S., Mitsui, K., Preklet, E., 2019: Color modification and homogenization of sugi wood by steaming. Asian Journal of Forestry 3(1): 20-24.
33. Varga, D., Van der Zee, M.E., 2008: Influence of steaming on selected wood properties of four hardwood species. Holz als Roh-und Werkstoff 66(1): 11-18.
34. Zivkovic, V., Arnold, M., Radmanovic, K., Richter, K., Turkulin, H., 2014: Spectral sensitivity in the photodegradation of fir wood (*Abies alba* Mill.) surfaces: colour changes in natural weathering. Wood Science and Technology 48: 239–252.

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