Colour Modification of Wood by Dry Thermal Treatment between 90 ºC and 200 ºC

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Abstract – The colour modification effect of dry thermal treatment was studied in black locust (*Robinia pseudoacacia* L.), poplar (*Populus x euramericana cv. Pannonia*), Scots pine (*Pinus sylvestris* L.), spruce (*Picea abies* Mill.) and larch (*Larix decidua* L.) species in the temperature range 90-200 °C. Colour data were presented and evaluated in the CIE L*a*b* coordinate system. All thermal treatments applied altered the wood colour throughout the entire cross section regardless of the treatment temperature. At lower temperatures, wood extractives played a decisive role in colour change. The degradation products of hemicelluloses were the major determinant of the change in lightness at 200°C. Redness change in percentage showed much greater alteration than the yellowness and the lightness change. Spruce presented the greatest chromaticity coordinate (a* and b*) alteration among the investigated species. Changes in redness and yellowness followed the Arrhenius law during the investigated dry thermal treatments confirming that the temperature dependence of these colour parameters is exponential for wood material.

Arrhenius law / extractives / hemicelluloses / chroma

Kivonat – A faanyag színének változása száraz hőkezelés hatására 90 ºC és 200 ºC között. Száraz körülmények között végrehajtott termikus kezelés színváltoztató hatását vizsgáltuk akác (*Robinia pseudoacacia* L.), nyár (*Populus x euramericana cv. Pannonia*), erdei fenyő (*Pinus sylvestris* L.), lucfenyő (*Picea abies* Mill) és vörösfenyő (*Larix decidua* L.) faanyag esetében 90 – 200 °C hőmérséklet tartományban. A szín adatokat a CIE L*a*b* koordináta rendszerben adtuk meg és értékeltük. Az alkalmazott hőkezelések a faanyag színét, függetlenül az alkalmazott hőmérséklettől, a próbatestek teljes keresztmetszetében megváltoztatták. Alacsony hőmérsékleten az extrakt anyagtartalom volt meghatározó a színváltozásban. A hemicellulózok degradációs termékei határozták meg döntő mértékben a világosság változását 200 °C-on. A százalékosan megadott vörös színezetváltozás sokkal nagyobb mértékű volt, mint a sárga színezet és a világosság változása. A luc faanyaga mutatta a legnagyobb színezeti koordináta-változást (a*és b*) a vizsgált faanyagok közül. A vörös és a sárga színezet változása követte az Arrhenius törvényt a száraz termikus kezelések során mutatva, hogy ezeknek a paramétereknek a hőmérséklet függése exponenciális faanyag esetében.

Arrhenius törvény / extrahálható anyagok / hemicellulózok / színezett dústság

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1 INTRODUCTION

Extractives are the thermally most sensitive molecules in wood. Among the main chemical components, hemicelluloses are the most susceptible and can undergo thermal degradation above 100 °C. The colour modifying effect of thermal treatment is highly temperature dependent. Colour change is a slow process below 100 °C under dry conditions. Therefore, dry thermal treatment is not of industrial interest below 100 °C, but is of theoretical importance. The colour change of wooden surfaces at room temperature is an extremely slow process. Nevertheless, an indoor wooden structure will darken within a few years, even in total darkness, due to thermal degradation.

The colour modifying effect of dry thermal treatment below 150 °C is a scarcely investigated phenomenon because of the long treatment time required. Tolvaj – Faix (1996) compared the colour modification effect of thermal treatment at 90 °C under dry and wet (steaming) conditions. The treatment period was 36 days because of the slow changes under dry conditions. The results showed that the dry treatment resulted in a much smaller colour alteration compared to the wet (steam) treatment. Chen et al. (2012) studied the colour change of black locust flour caused by thermal treatment in oxygen and nitrogen atmospheric dry and wet conditions. Results showed that the samples suffered greater colour change in terms of all colour parameters (L*, a*, b*) in the presence of oxygen than in nitrogen atmosphere during dry thermal treatment at 120 °C. The decrease in lightness value was, for example, twofold in the presence of oxygen. Popescu et al. (2013) studied the chemical modifications of lime (*Tilia cordata* Mill.) wood during heat treatment at low temperature (about 140 °C) and 10% relative humidity, by infrared spectroscopy. The treatment resulted in the formation of acetic acid, which catalyse the hydrolysis reactions of hemicelluloses and amorphous cellulose. The cleavage of the β-*O*-4 linkages and splitting of the aliphatic methoxyl chains from the aromatic lignin ring was also observed.

Higher temperatures (above 150 °C) produce rapid colour change. These treatments have industrial importance not only for the quick and intensive colour change but because of some positive property change of wood. The industrially used temperature interval is 160-260 °C. Stamm – Hansen (1937) heated wood with different gases and reported the decrease of equilibrium moisture content, swelling and shrinkage values. During the next almost 100 years plenty of papers reported more and more results regarding dry thermal treatments at high temperatures. A review paper (Esteves – Pereira 2009) summarised the results of 160 papers. Also, there are several patented processes to produce thermally treated wood material. The most successful in Europe is the ThermoWood patented by Viitaniemi et al. (1997).

There are two distinct groups of thermal treatments based on the presence of oxygen. One group of treatments is where the heating medium contains oxygen while procedures in the other group exclude oxygen. Chemical reactions are quite different within these two groups since the presence of oxygen allows oxidation. Heat treatments create free radicals which can then react with oxygen forming oxidation products such as quinines (Bekhta – Niemz 2003). Such oxidative reactions are inherently impossible in the absence of oxygen. The most common option to exclude oxygen is the application of oils as heating medium. A review article (Lee et al. 2018) presents 139 papers dealing with the thermal treatment of wood in vegetable oils.

Most of the experiments in the literature were carried out in the presence of oxygen above 160 °C (Bekhta – Niemz 2003, Nuopponen et al. 2003, Sundqvist et al. 2006, Windeisen et al. 2007, Kaciková et al. 2013, Kamperidou et al. 2013, Zanuncio et al. 2015, Miklecic – Jirous-Rajkovic 2016, Griebeler et al. 2018, Sikora et al. 2018, Lo Monaco et al. 2020).

The aim of this study was to give wide insight on the temperature dependence of the colour modification effects of dry thermal treatment applying broad temperature range between 90 and 200 °C.

2 MATERIALS AND METHODS

The colour modification effect of dry thermal treatment was studied applying wide range of temperatures (90 - 200 °C) in a drying chamber. Duration of the heat-up period to reach the desired treatment temperature was 10, 15, 15, 20 and 30 minutes at 90, 110, 130, 160 and 200 °C, respectively. There was no cooling time. Samples were removed from the chamber right after finishing the treatment. Test samples for the different temperatures were cut from the same board to minimize the effect of inhomogeneity. The sample size was 100 x 20 x 10 (mm^3) having planed surfaces. Deciduous and conifer species with different extractive content were involved in the test to determine the temperature dependence of dry thermal treatment.

Black locust heartwood (*Robinia pseudoacacia* L.) was chosen because of its high extractive content. On the contrary, Poplar (*Populus x euramericana cv. Pannonia*) was tested due to its low extractive content. Larch heartwood (*Larix decidua* L.), Scots pine heartwood (*Pinus sylvestris* L.) and spruce (*Picea abies* Mill.) were chosen because of their high, medium and low extractive content, respectively. Previous results showed that the colour change generated by thermal treatment is highly temperature dependent. Longer treatment times are required at lower temperatures. Below 150 °C, several days are necessary to obtain a substantial colour change. The treatment time above this temperature limit is rather short, some hours are enough to induce remarkable colour change. That is why the treatments were caried out in two different series. In the first series (at 90, 110 and 130 °C) the chosen treatment period was 18 days. In contrast, only 6-hour treatment was applied in the second series at all investigated temperatures (at 90, 110, 130, 160 and 200 °C). Temperatures less than 90 °C generates extremely slow colour change. Therefore, this temperature value was chosen as the lower limit.

Colour parameters were measured on the radial surface of the samples to determine the average colour of earlywood and latewood. Colour measurement was carried out with a Konica-Minolta 2600d colorimeter. The CIE L* (Lightness), a* (Redness), b* (Yellowness) colour space data were calculated based on the D65 illuminant and 10° standard observer with the aperture diameter of 8 mm.

The intensity of a thermally activated process increases usually exponentially by rising temperature according to the Arrhenius law. The Arrhenius equation relates the rate of a chemical reaction k to temperature T and it includes the activation energy. The equation is simple if the activation energy is constant. The Arrhenius equation can be given in the logarithmic form:

$$\ln k = \frac{-E_a}{R}\frac{1}{T} + \ln A \tag{1}$$

Where:

R :	the universal gas constant
A:	the pre-exponential constant
k:	kinetic constant
T:	temperature
E_a	activation energy

An Arrhenius plot displays the logarithm of a kinetic constant (k, ordinate) plotted against inverse temperature (1/T, abscissa). Arrhenius plots are often used to analyse the effect of temperature on the rates of chemical reactions. For a single rate-limited thermally activated process, an Arrhenius plot gives a straight line and presents that the temperature dependence is exponential.

3 RESULTS AND DISCUSSION

This study discusses solely the colour change aspects of thermal treatments. One of the main advantages of dry thermal treatment is that it alters wood colour in its whole cross section without using any harmful chemicals. This thermal process can also be applied to modify the colour of wood species having unattractive or highly inhomogeneous initial colour. Moreover, thermally modified dark wood may substitute tropical wood species as well (Banadics et al. 2016).

Plotting the L*, a* and b* colour coordinates as a function of the treatment time could provide detailed information regarding the colour alteration. *Figure 1* shows the lightness alterations of certain investigated wood species. Dry thermal treatment at 90 °C generated almost no lightness decrease except for black locust where the total lightness decrease was 10 units at the 18th day of the treatment. Elevated temperatures amplified the lightness decreases. The investigated conifer species presented similar lightness decreases. Trendlines belonging to the same treatment temperature run close to parallel. Only spruce presented a little deviation as its lightness decrease was less intensive during the first 3 days of the treatment compared to the other two conifers. Poplar samples showed somewhat different lightness decrease character that the other species. All tree trendlines of poplar are close to straight lines and the measured lightness values generated by 90 and 110 °C are close to each other showing very little lightness decrease. In contrast, the treatment at 130 °C produced intensive lightness decrease. Poplar has low extractive content. Consequently, the darkening of the samples was generated mainly by the degradation products of hemicelluloses. 130 °C is high enough to degrade wood hemicelluloses. The missing intensive change at the beginning of the treatment procedure is due to the low extractive content of poplar and the same is valid partly for spruce as well. Black locust showed the most rapid lightness decrease during the first 3 days of treatment at all temperatures compared to the other investigated species. It is not surprising because black locust has the highest extractive content among the investigated species. Results show that the extractives are the most sensitive chemical components in wood during thermal treatment at low temperatures, followed by the hemicelluloses.

When investigating colour change procedures, a* and b* colour coordinates provide more detailed information than L* alone thus allow more subtle conclusions because the calculation of a* and b* use both X and Z tristimulus values beside Y. (Calculation of L* incorporates Y only.) Furthermore, it is important to know that the calculation of Y encompasses the reflection values of almost the whole visible wavelength interval. In contrast, determination of X utilizes two wavelength intervals around 600 and 450 nm, and the calculation of Z utilizes only one narrower wavelength interval around 450 nm. These properties of a* and b* show that they are more sensitive to the wavelength dependence of the reflectance changes than L*.

Figure 2 presents the redness change of the investigated species generated by dry thermal treatment. The initial redness values of the investigated species were for poplar, spruce, black locust, Scots pine and larch 2.7, 4.5, 5.9, 6.4 and 11.6, respectively. This order represents the extractive content values responsible for the redness of the species. Initial redness value of larch was more than twice as high as the initial a* values of the other species. The unequalled high durability of larch is lying in its special extractives responsible also for its high redness value. Gierlinger et al. (2004) investigated different larch species having large distribution of a* values (between 5.6 and 9) and found that the redness values were strongly correlated with phenolics content.

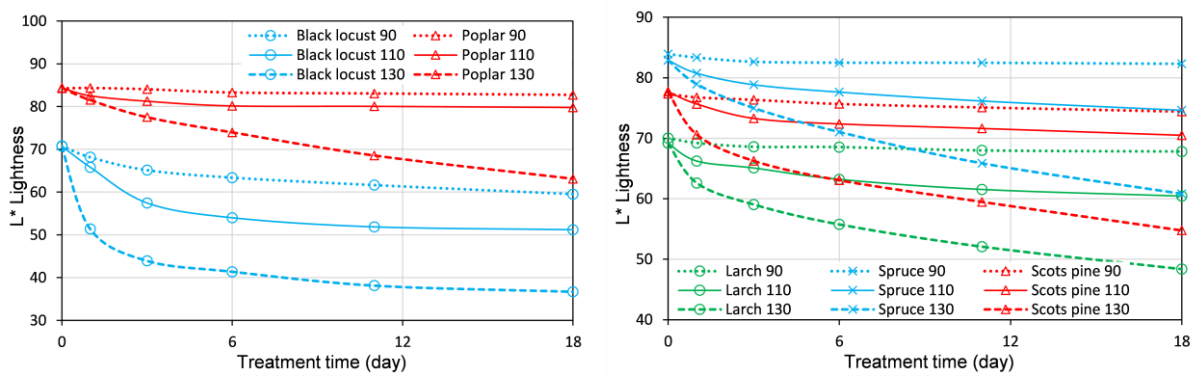

Figure 1. Lightness change of black locust, poplar, larch, spruce and Scots pine samples during dry thermal treatment

Heating at 90°C caused only a little redness increase during the 18-day treatment. There were two exceptions. The redness value of larch did not change at all while black locust produced considerable redness value increase (59 %) even at this temperature. Extractives in black locust responsible for redness increase are highly sensitive to thermal treatment. Treatment at 130 °C showed that the heat generated degradation products of extractives are chemically not stable enough. These degradation products underwent secondary degradation and reduced the redness values after 6-day treatment of black locust and after 11-day treatment of larch. Larch seems to be the thermally most stable species among the investigated ones. It has the highest initial redness value (12 units) and the maximum of redness increase was only 24 %. Spruce and Scots pine produced similar redness alterations. The redness change course of poplar was completely different to the other species. The intensive increase during the first 3 days of treatment (comparing to the whole change) was completely missing, the trendlines were close to linear. The redness increase was slow at 90 and 110 °C while at 130 °C a quite intensive and enormously high (323 %) increase could be observed during the 18-day treatment. Similar data for spruce, Scots pine, black locust and larch are 161, 106, 73 and 21 %, respectively. The results show that the degradation of hemicelluloses was dominant at 130 °C and it generated the main portion of redness increase. It seems that the extractive content could partly protect the hemicelluloses against thermal degradation. Black locust and larch with the highest extractive content showed the smallest red hue increase while poplar featuring the smallest extractive content produced the greatest red hue increase. In terms of protective effect, the type of the extractives can also play an important role. These assumptions need further chemical investigations.

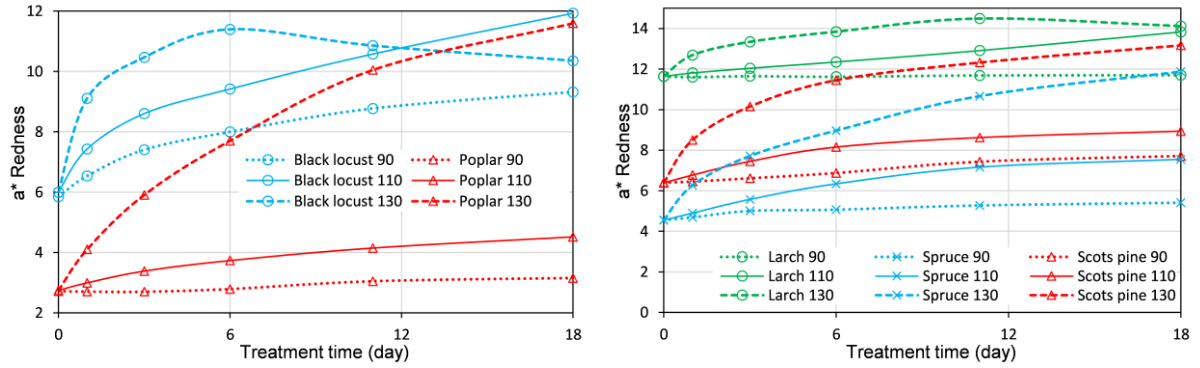

Figure 2. Redness change of black locust, poplar, larch, spruce and Scots pine samples during dry thermal treatment

Figure 3 shows the yellowness change of the investigated species induced by different dry thermal treatments. The initial values of the b* co-ordinate were 17.4, 22.3, 26.6, 27.7 and 28.1 for poplar, spruce, larch, black locust and Scots pine, respectively. The initial yellowness values were higher than the initial redness values, and the dispersion of the colour co-ordinates was smaller for yellowness than for redness. Most of species showed yellowness increase due to the thermal treatments. The only exception was black locust. Its yellowness decreased during the applied thermal treatments. The high robinetin content of black locust (covering 2-5 % of its total mass) causes its unattractive yellow colour. The robinetin type extractives are highly sensitive to thermal treatment. Heat induced degradation of these extractives led to a reduce in the yellowness of black locust even at 90 °C and caused an extremely rapid decrease during the first day already at 130 °C.

The investigated species have two types of extractives that are responsible for yellow colour. One of them generated yellowness increases while the other type produced yellowness decrease during thermal treatment. The second type of extractives were found in black locust and in larch. Larch samples produced mainly yellowness decrease at 130 °C similarly to black locust. The final yellowness value at the end of the 130 °C treatment was considerably smaller than the initial one meaning that substantial part of the decrease was induced by the degradation of extractives being originally in larch wood. Treatment at 90 °C generated slow but continuous yellowness change in all investigated species. These changes were relatively fast during the first 3 days of all treatments, then the shift slowed down afterward showing that extractives were involved in the degradation processes of all species. Treatment at 110 °C resulted in considerably greater redness change than that at 90 °C. Conifers produced both yellowness increase and decrease during the treatment at 130 °C. Maximum b* values were reached on the 11th, 6th and 1st day of the treatment of spruce, Scots pine and larch, respectively. This phenomenon shows that the heat generated degradation products underwent secondary degradation at 130 °C.

Poplar and spruce (having the smallest extractive content) presented the greatest yellowness increase on the 11th day of the treatment. These increases were 73 and 42 % for poplar and spruce at 130 °C. These data show that hemicelluloses play an important role in the yellowness change during dry thermal treatment.

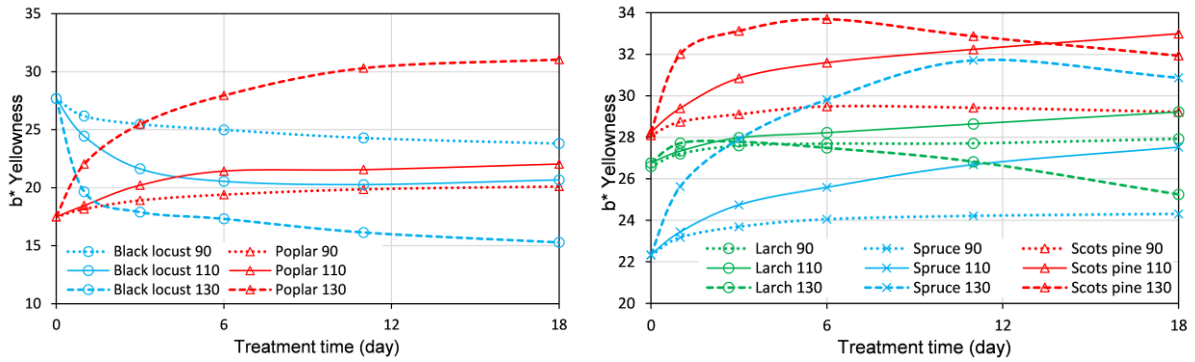

Figure 3. Yellowness change of black locust, poplar, larch, spruce and Scots pine samples during dry thermal treatment

Figure 4 present the locations of colour dots of black locust, poplar, larch, spruce and Scots pine species generated by dry treatments at 90, 110 and 130 °C. The left end of the trendlines represents the colour dots of untreated samples followed by the dots of treated samples with increasing treatment time. These figures show the change of hue and of chroma. Black locust is a great exception. It suffered great hue value decrease from 78° to 55° at 130 °C changing the greyish-yellow colour to brown tint. The treatment at 90 °C also caused considerable hue

value decrease for black locust comparing to the other species. The other species hardly changed their hue at this temperature. Treatment at 110 °C produced moderate chroma increase and small hue decrease. Exception was the hue decrease of black locust. This decrease was 18 units. The chroma values (distance between the colour dot and the origin of the coordinate system) of black locust slightly increased by dry thermal treatment only. The behaviour of larch was partly exceptional as well. It showed small chroma increase and moderate hue value decrease. The other species (poplar, spruce and Scots pine) produced great chroma increase and moderate hue value decrease. The maximum chroma increase of poplar and spruce were 16 and 12 units generated by thermal treatment at 130 °C.

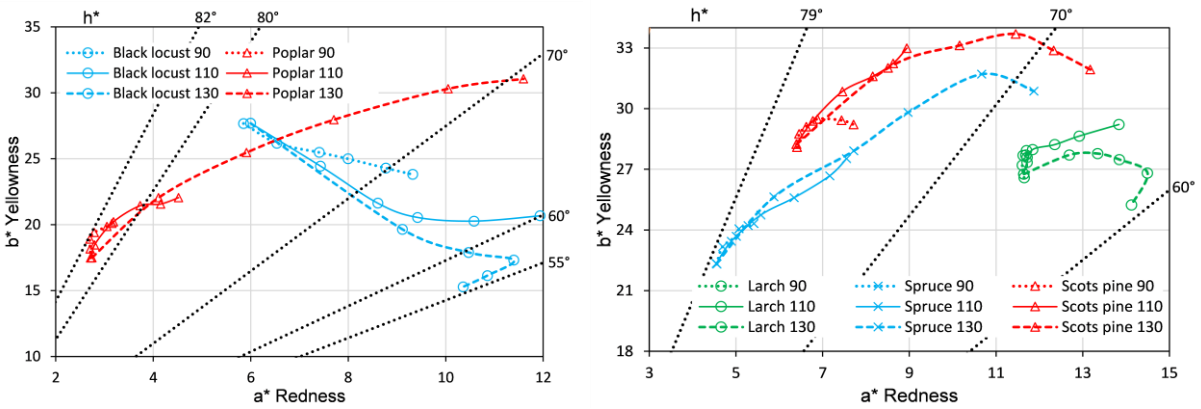

Figure 4. Colour dots of black locust, poplar, larch, spruce and Scots pine samples on the a-b* plane during dry thermal treatment. (Dots at the left end of the lines represent the untreated samples.) Constant hue (h*) lines are also indicated*

The investigated temperature range (90-130 °C) has only theoretical importance because of the long treatment time. The results show that dry thermal treatment below 100 °C causes extremely slow colour alteration. Similar results were found by Liu et al. (2016) at 100°C under semidry conditions (air RH=55 %).

Higher temperatures (above 150 °C) produce rapid colour change. These treatments have industrial importance not only for the quick and intensive colour change but because of some positive property change of wood. These temperatures generate intensive colour change after some hours of treatment already. Consequently, the treatments for the parallel investigation at 90, 130, 160 and 200 °C required short periods. The chosen time interval was 6 hours.

The lightness change data generated by the chosen four temperatures are presented in *Figure 5*. Lightness values decreased for all investigated species at all treatment temperatures. The decrease at 90°C was negligible (almost zero) for all species during the 6-hour treatment. At this temperature, the greatest lightness decrease was produced by black locust (0.7 unit). The next temperature stage, the treatment at 130 °C induced a relatively large lightness decrease (14 units) for black locust. In contrast, this data was only 0.2 unit for poplar. The elevated temperature (160 °C) doubled the lightness change of black locust compared to the effect of 130 °C. The same lightness decrease was 3, 6, 7 and 54-fold for Scots pine, spruce, larch and poplar, respectively. Comparing the behaviour of black locust and poplar, a highly different nature of changes can be observed. The dominant change of back locust was induced by its extractives. On the contrary, poplar hardly has extractives, thus its darkening was mainly generated by the degradation products of hemicelluloses. It is also visible that the degradation of extractives is rapid at the beginning of the treatment while the tendency of change is moderate later on. In contrast, the degradation progress of hemicelluloses is much more uniform during thermal treatment. Dry thermal treatment at 200 °C generated almost the same lightness decrease for all investigated species showing that the degradation of hemicelluloses is the

dominant alteration at this high temperature. Black locust presented the greatest relative lightness decrease at 200 °C compared to the initial value (82 %). Researchers demonstrated that most of the extractives disappeared during thermal treatment at such high temperatures. The thermal stability of extractives in Scots pine was studied by Nuopponen et al. (2003). It was found that fats and waxes were not detectable in the sapwood edges above 160 °C. At temperatures above 200 °C all resin acids disappeared from the wood.

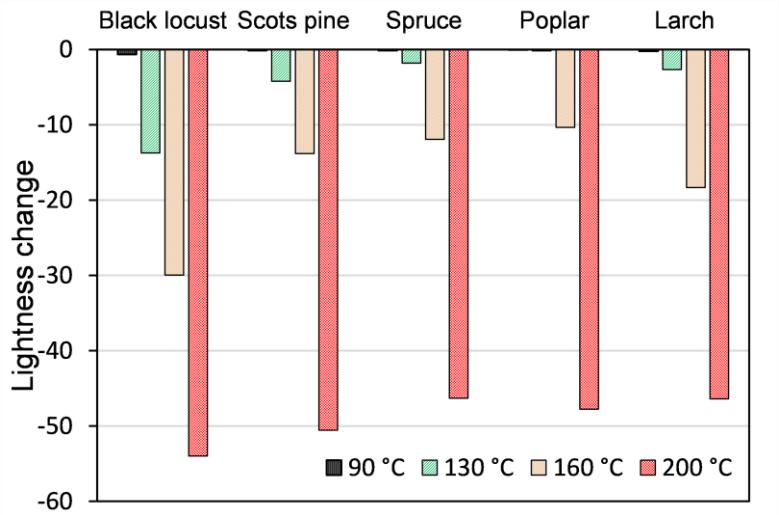

Figure 5. Lightness change of different wood species generated by 6-hour dry thermal treatment at different temperatures

Figure 6 shows the redness change of the species generated by 6-hour dry thermal treatment at various temperatures. The 6-hour treatment caused very little redness change at 90 °C. Only larch presented a slight redness decrease. It is important to mention that the redness of larch remained intact during 18-day treatment at 90 °C (see *Figure 2*). Larch showed very little redness increase at 130 °C as well. Black locust suffered the greatest redness increase at all applied temperatures except 200 °C because of the high thermal sensitivity of its extractives. The 6-hour treatment at 200 °C was too long for black locust. Its redness value started decreasing after 4-hour treatment. The other species showed small redness increase at 130 °C and medium increase at 160 °C. The only exception was poplar. Its redness increase was only 1.2 units at 160°C. These results confirm that the degradation of hemicelluloses is a relatively slow but continuous process. In contrast, the degradation of extractives is rapid at the beginning of thermal treatment and slows down afterward. Obviously, 6 hours at 160 °C was too short to induce such a strong modification of hemicelluloses that would be able to lead to a considerable redness increase. However, this short period was enough for the alteration of extractives to generate considerable redness increase in the other investigated species (except poplar). The treatment at 200°C produced great redness increase for all species. The rate of the redness change was more species dependent than in the case of lightness change. Spruce presented the greatest relative redness increase at 200 °C compared to the initial value (306 %).

Figure 7 shows the yellowness change of the species generated by 6-hour dry thermal treatment at various temperatures. Black locust is an exception in terms of yellowness change as well; b* colour coordinate of all investigated species increased except that of black locust. Although, the yellowness decreases of black locust showed similar temperature dependence than that of the other species but in the opposite direction.

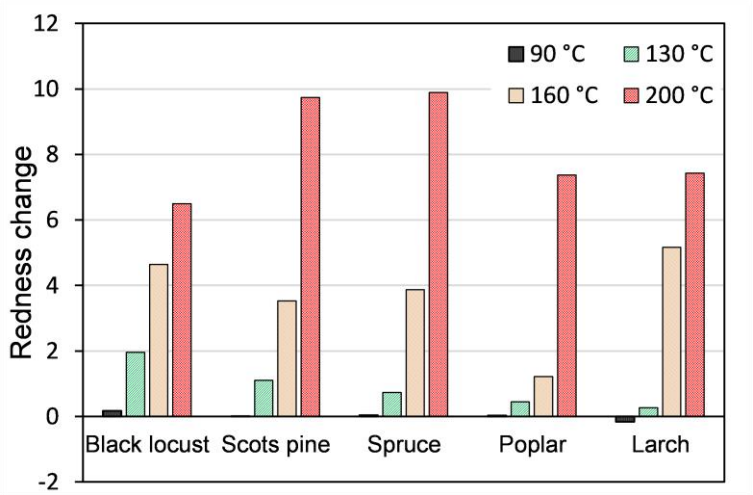

Figure 6. Redness change of different wood species generated by 6-hour dry thermal treatment at different temperatures

This behaviour can be interpreted by the high robinetin type extractive content of black locust. These extractives are responsible for the greyish-yellow initial colour of black locust. Robinetin type extractives are highly sensitive to thermal degradation and this degradation reduces the yellowness value considerably. The effect of the treatment at 90 °C seems to be neglectable in terms of b* shift similarly to the redness change, for all species. Yellowness change at high temperatures (above 130 °C) was highly species dependent. This phenomenon shows the complexity of the yellowness change. Redistribution and degradation of lignin may cause yellowness change beside the degradation of extractives and hemicelluloses. This kind of lignin related, high temperature induced processes were published in many papers (Tjeerdsma et al. 1998, Wikberg – Maunu 2004, Boonstra – Tjeerdsma 2006, Esteves – Pereira 2009, Kaciková et al. 2013, Esteves et al. 2013, Sikora et al. 2018). As it was introduced in *Figure 6*, spruce and Scots pine presented similar redness change at all temperatures. Yellowness change of these species was also similar except at 200°C. Spruce produced the greatest yellowness increase at 200 °C, which was 80 % compared to the initial value. This value was almost twice as high as the yellowness increases of Scots pine. This huge difference was probably generated by the different degradation properties of lignin within these species.

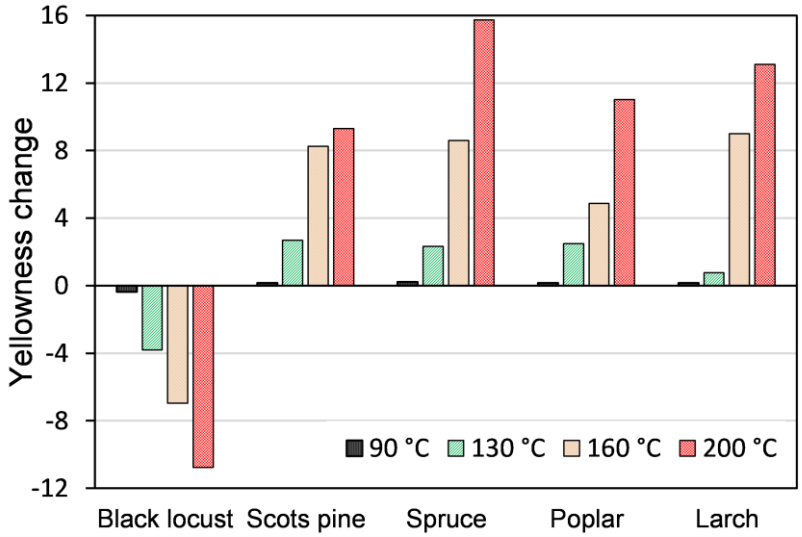

Figure 7. Yellowness change of different wood species generated by 6-hour dry thermal treatment at different temperatures

Figures 8 and 9 present the Arrhenius plots of redness and yellowness values of black locust, poplar, larch, spruce and Scots pine species thermally treated for 6 hours at different temperatures. The Arrhenius law declares that if the Arrhenius plot is a straight line, the temperature dependence of the studied process is exponential. In our case, all Arrhenius plots are straight lines presenting that both redness and yellowness changes are exponential functions of the dry thermal treatment temperature. The coefficients of determination values are quite high for all investigated wood species. R^2 values are above 0.9 for redness and above 0.92 for yellowness. The slope of the trendline for yellowness of black locust is opposite of that of the other lines showing that the changing tendency is also opposite in this case. Some colour dots are relatively fare from the trendline. The reason is laying in the colour inhomogeneity of wood. It is important to mention that all presented dots in the figures belong to different samples having slightly different initial colour. The other inhomogeneity problem is that the radial surface of the specimens was used for colour measurement. It is impossible to guarantee that all individual colour measurement covers the same earlywood-latewood ratio within the measured area.

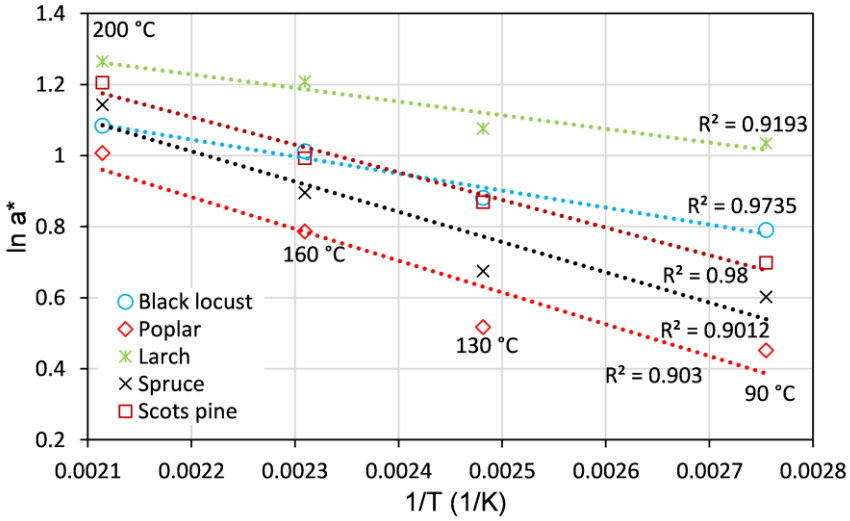

Figure 8. Arrhenius plots of redness values for black locust, poplar, larch, spruce and Scots pine species thermally treated for 6 hours at different temperatures

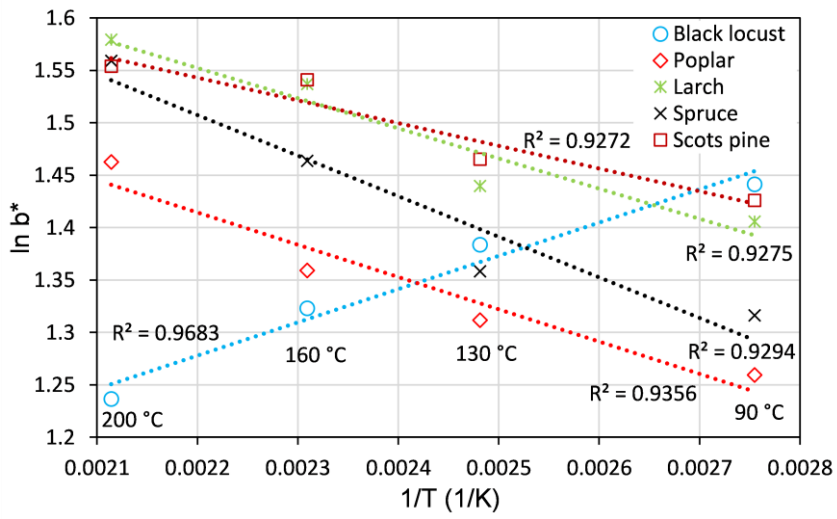

Figure 9. Arrhenius plots of yellowness values for black locust, poplar, larch, spruce and Scots pine species thermally treated for 6 hours at different temperatures

Arrhenius plots of the lightness values do not determine exactly straight lines. Fitting straight lines on the lightness dots the coefficient of determination values are much smaller (between 0.6 and 0.8) than in the case of redness and yellowness suggesting that the lightness change is determined by multiple chemical alterations generating absorption in the visible light region and the individual chemical changes are marked with different temperature dependence.

4 CONCLUSIONS

The colour modification effect of dry thermal treatment was studied in the temperature range 90-200 °C. The results showed that the 18-day treatment at 90 °C was short, while 6 hours at 200 °C was sufficient. The applied thermal treatments altered the wood colour throughout the whole cross section independently on the treatment temperature. In terms of the colour change, the extractive content of the species was dominant at low temperatures. The degradation products of hemicelluloses were the major determinant of the change in lightness at 200 °C. Redness change in percentage showed much greater alteration than the yellowness and the lightness change. Spruce presented the greatest chromaticity coordinate (a* and b*) alteration. Redness and yellowness changes followed the Arrhenius law during dry thermal treatment confirming that the temperature dependence of these colour parameters is exponential in the case of wood material.

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REFERENCES

BANADICS, E.A. – GÁLOS, B. – TOLVAJ, L. (2016): A sötét egzóta faanyagok helyettesítése gőzölt akác faanyaggal. [Substitution of dark exotic wood species by steamed black locust.] (in Hungarian) Faipar 64(1): 22–28.

BEKHTA, P. – NIEMZ, P. (2003): Effect of high temperature on the change in color, dimensional stability and mechanical properties of spruce wood. Holzforschung 57: 539–546. https://doi.org/10.1515/HF.2003.080

BOONSTARA, M.J. – TJEERDSMA, B. (2006): Chemical analysis of heat treated softwoods. Holz als Roh- und Werkstoff 64: 204–211. http://dx.doi.org/10.1007/s00107-005-0078-4

CHEN, Y. – FAN, Y. – GAO, J. – STARK, M.S. (2012): The effect of heat treatment on the chemical and color change of black locust (*Robinia pseudoacacia*) wood flour. BioResources 7(1): 1157–1170.

ESTEVES, B.M. – PEREIRA, H.M. (2009): Wood modification by heat treatment: A review. BioResources 4(1): 371–404.

ESTEVES, B. – MARQUES, A.V. – DOMINGOS, I. – PEREIRA, H. (2013): Chemical changes of heat treated pine and eucalypt wood monitored by FTIR. Maderas, Sciencia Technology 15(2): 245–258. http://dx.doi.org/10.4067/S0718-221X2013005000020

GIERLINGER, N. – JACQUES, D. – GRABNER, M. – WIMMER, R. (2004): Colour of larch heartwood and relationships to extractives and brown-rot decay resistance. Trees 18(1): 102–108. https://doi.org/10.1007/s00468-003-0290-y

GRIEBELER, C. – TONDI, G. – SCHNABLE, T. – IGLESIAS, C. – RUIZ, S. (2018): Reduction of the surface colour variability of thermally modified Eucalyptus globulus wood by colour pre-grading and homogeneity thermal treatment. European Journal of Wood and Wood Product 76: 1495–1504. https://doi.org/10.1007/s00107-018-1310-3

KACIKOVA, D. – KACIK, F. – CABALOVÁ, I. – DURKOVIC, J. (2013): Effects of thermal treatment on chemical, mechanical and colour traits in Norway spruce wood. Bioresource Technology 144: 669–674. https://doi.org/10.1016/j.biortech.2013.06.110

KAMPERIDOU, V. – BARBOUTIS, I. – VASILEIOU, V. (2013): Response of colour and hygroscopic properties of Scots pine wood to thermal treatment. Journal of Forestry Research 24(3): 571–575. https://doi.org/10.1007/s11676-013-0389-y

LEE, S.H. – ASHAARI, Z. – LUM, W.C. – HALIP, J.A. – ANG, A.F. – TAN, L.P. – CHIN, .L. – TAHIR, P.M. (2018): Thermal treatment of wood using vegetable oils: A review. Construction and Building Materials 181: 408–419. https://doi.org/10.1016/j.conbuildmat.2018.06.058

LIU, X.Y. – TIMAR, M.C. – VARODI, A.M. – SAWYER, G. (2016): An investigation of accelerated temperature-induced ageing of four wood species: colour and FTIR. Wood Science and Technology 21: 357–378. https://doi.org/10.1007/s00226-016-0867-4

Lo MONACO, A. – PELOSI, C. – AGRESTI, G. – PICCHIO, R. – RUBINO, G. (2020): Influence of thermal treatment on selected properties of chestnut wood and full range of its visual features. Drewno-Wood 63: 5–24. https://doi.org/10.12841/wood.1644-3985.344.10

MIKLEČIĆ, J. – JIROUŠ-RAJKOVIĆ, V. (2016): Influence of thermal modification on surface properties and chemical composition of beech wood (*Fagus sylvatica* L.). Drvna Industrija 67(1): 65–71. https://doi.org/10.5552/drind.2016.1520

NUOPPONEN, M. – VUORINEN, T. – JAMSA, S. – VIITANIEMI, P. (2003): The effects of a heat treatment on the behaviour of extractives in softwood studied by FTIR spectroscopic methods. Wood Science and Technology 37: 109–115. https://doi.org/10.1007/s00226-003-0178-4

POPESCU, M.C. – FROIDEVAUX, J. – NAVI, P. – POPESCU, C.M. (2013): Structural modifications of *Tilia cordata* wood during heat treatment investigated by FT-IR and 2D IR correlation spectroscopy. Journal of Molecular Structure 1033: 176–186.

SIKORA, A. – KACIK, F. – GAFF, M. – VONGROVÁ, V. – BUDENIKOVÁ, T. – KUBOVSKY, I. (2018): Impact of thermal modification on color and chemical changes of spruce and oak wood. Journal of Wood Science 64: 406–416. https://doi.org/10.1007/s10086-018-1721-0

STAMM, A. – HANSEN, L. (1937): Minimizing wood shrinkage and swelling: Effect of heating in various gases. Industrial and Engineering Chemistry 29(7): 831–833.

SUNDQVIST, B. – KARLSSON, O. – WESTERMARK, U. (2006): Determination of formic-acid and acetic acid concentrations formed during hydrothermal treatment of birch wood and its relation to colour, strength and hardness. Wood Science and Technology 40: 549–561. https://doi.org/10.1007/s00226-006-0071-z

TJEERDSMA, M. – BOONSTRA, M. – PIZZI, A. – TEKELY, P. – MILLITZ, H. (1998): Characterisation of thermally modified wood: molecular reasons for wood performance improvement. Holz als Roh und Wekstoff 56: 149–153.

TOLVAJ, L. – FAIX, O. (1996): Modification of Wood Colour by Steaming. ICWSF '96 Conference, Sopron 10-19. pp.

VIITANIEMI, P. – JÄMSÄ, S. – VIITANEN, H. (1997): Method for improving biodegradation resistance and dimensional stability of cellulosic products. United States Patent Nº5678324.

WIKBERG, H. – MAUNU, S.L. (2004): Characterisation of thermally modified hard- and softwoods by CPMAS NMR. Carbohydrate Polymers 58: 461–466. https://doi.org/10.1016/j.carbpol.2004.08.008

WINDEISEN, E. – STROBLE, C. – WEGENER, G. (2007): Chemical changes during the production of thermo-treated beech wood. Wood Science and Technology 41: 523–536. https://doi.org/10.1007/s00226-007-0146-5

ZANUNCIO, A.J.V. – CARVALHO, A.G. – de SOUZA, M.T. – JARDIM, C.M. – CARNEIRO, A.C.O. – COLODETTE, J.L. (2015): Effect of extractives on wood color of heat treated Pinus radiata and Eucalyptus pellita. Maderas, Ciencia Technoligy 17(4): 857–864. http://dx.doi.org/10.4067/S0718-221X2015005000074