

INVESTIGATION OF HYDROTHERMAL PLASTICIZATION PROCESS WITH CONVECTIVE AND DIELECTRIC HEATING

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Abstract:

*In this paper time needed for convectional and dielectric heating of wood samples to the desired plasticizing temperature was experimentally determined. The temperature was measured using a contactless infrared thermometer on the surface of samples and after their splitting in the spatial center of gravity. The same temperature of the samples was achieved by dielectric heating using microwave radiation six times faster than steaming within convection. Both methods of heating have caused moisture reduction of samples. In the case of microwave heating, the decrease in moisture content was twice as much as steaming. In the second part of this study mechanical properties of samples in a plastic and reference state were measured. The samples were tested in a static three-point bend. The results did not confirm statistically significant effect of heating methods on the degree of wood plasticity. Comparison of the mechanical properties of plastic and reference samples showed the expected large drop due to increased temperature and high moisture content. Test samples were made from beech (*Fagus sylvatica* L.).*

Key words: *hydrothermal plasticization; convectional heating; dielectric heating; time consumption; wood plasticity.*

INTRODUCTION

Convective heating

According to Younsi *et al.* (2006) the plasticization heat enters the wood internal structure from the surroundings starting at the surface and penetrating to the middle layers mainly by conduction. The velocity of heating in the superficial layers is considerably higher, corresponding to a steeper growth curve and a higher temperature of the surface during heating (Zielonka and Gierlik 1999). After a time, both curves approach the same horizontal asymptote that expresses the final temperature of heating within the entire wood volume.

The driving force of the plasticization heat movement, both for convection and conduction is the temperature gradient between two areas of wood structure. The direction of the heat flow is determined by Newton's law of cooling. The amount of the transmitted plasticization energy between the areas is in direct proportion to the size of the gradient and material constants that mediate the effect of wood material properties on thermal conduction (Plumb *et al.* 1985). Desh and Dinwoodie (1996) state that wood material properties can be expressed as a weighted sum of specific values of properties valid for individual basic wood components, i.e. cellulose, hemicelluloses, lignin, air, or gaseous and liquid moisture. Thus, the velocity of convective heating is determined by the size of the gradient of the wood internal structure temperature and the temperature of its surroundings, velocity of air flow, moisture content (MC) in wood, wood density, and the direction of heat distribution in relation to the orientation of wood anatomical elements. The values of all the above mentioned material constants of proportionality and thus also the velocity of wood heating change in dependence on the current wood temperature, which is a function of time – non-stationary diffusion of heat (Deliiski *et al.* 2010).

Dielectric heating

In the case of dielectric heating, the plasticization heat originates directly in the structure of the material. If the substance exposed to electromagnetic (EMG) radiation contains polar particles (in wood they are H₂O, -OH, -COOH, -CH₂OH), the polar particles react to the EMG field by a series of processes, which lead to a production of heat (Hansson and Antti 2008). First, the polar molecules start rotating fast, which is caused by the polarity of the EMG field usually alternating about 10⁹ times a second (Hansson 2007). The binding of polar particles with the wood internal structure causes friction, which finishes the process of the transformation of the EMG energy absorbed by the heated material to plasticization heat (Antti and Perré 1999). If the polar particles, mainly water molecules, were equally distributed within wood structure and the EMG field had the same intensity within the entire volume of the element, the wooden elements would be heated at the same time in entire cross-section (Hansson 2007).

Zielonka and Gierlik (1999) as well as Antti and Perré (1999) agree that after some time, the centre of the profile has the lowest temperature and the temperature increases in the direction towards the element surface, the maximum being achieved a few millimetres under the surface (Brodie 2007). The values of material constants and thus also the speed of dielectric heating are determined by the amount of polar molecules contained in the wood structure, the level of their polarity and their mobility, i.e. wood density, MC, and temperature; they also depend on the intensity of the EMG field and the position of the field related to the direction of wood fibres (Sahin and Ay 2004).

Influence of moisture and temperature on bonds in wood structure

At a nano-scale, the structure of the basic chemical components of wood and their connecting into higher units are provided through chemical (primary) and physical (secondary) bonds, whose complex is characterized by Fengel and Wegener (1984) as lignin-saccharide (LS) net. The new considerably more stable electron configuration of binding atoms that is created through the primary bonds brings about a strength of bonds that is one order higher than in the case of secondary bonds. The change in mechanical properties of wood is related to thinning of LS net. As Sjostrom (1993) described while during plasticization the original covalent and hydrogen bonds are reduced, during stabilization of the new shape their number increases; the process is reversible and the changes are temporary. The creation of both types of bonds is related to a release of the excess energy, which has to be supplied again to fission a created bond. Due to the real volume of water molecules, the macromolecules move further apart and the cohesion of the originally connected macromolecules is gradually lost. According to Sjostrom (1993) the disruption of covalent bonds is only possible with cooperation of heat. As demonstrated Bouajila *et al.* (2006) the increased temperature also causes a change of lignin as a thermoplastic hydrophobic substance to a viscoelastic up to a plastic state by exceeding the glass transition temperature.

The effect of moisture and temperature on modulus of rupture (MOR) and modulus of elasticity (MOE) of wood in static bending

The effect of MC on wood MOR in static bending, whose change is analyzed in this study, can be to a great extent derived, according to Okuyama (1975), from the effect of moisture content on wood strength in compression, tension and shear along the grain. As demonstrated Katsuragi (2002) the effect of MC on tensile strength along the grain is about a quarter of the value of strength in compression along the grain. It follows that under stress in static bending an increase in wood MC will lead to a faster wood damage on the concave side of bending in the form of fibre deflection. The concurrent effect of MC and temperature on the wood internal structure is synergic. A typical example of the multiplied effect of the concurrence of temperature and MC on wood structure is described by Bouajila *et al.* (2006).

OBJECTIVES

The aim of this study is the experimental determination of time consumption convectional and dielectric heating of wood for the purpose of plasticization. The question also is whether heating method significantly affects the degree of plasticity of wood.

EXPERIMENTAL

Material

For the experiment the wood of the European Beech (*Fagus sylvatica* L.) with a MC of 15 – 20% was used. The samples were made as rectangular special orthotropic blocks with dimensions 20x20x300mm. Their total number was 400 pieces, out of which 100 were used for time-consumption study, 300 for plasticity study. In case of time-consumption study 50 samples for determination time of steaming and other 50 samples for determination time of microwave (MW) heating were used. In case of plasticity study 100 samples for convectional heating (i.e. steaming), 100 for dielectric heating (i.e. MW heating), and 100 for reference values of mechanical properties were used.

Methods

First the samples were dried to 0% MC in a laboratory drying kiln SANYO MOV 112 at a temperature of $103\pm 2^{\circ}\text{C}$. Afterwards, each sample was weighed and measured. The observed data were used to calculate the mean density of each sample. The procedure for measuring density of absolutely dry samples included drying of the samples to 0% MC using a laboratory drying kiln SANYO MOV 112 at a temperature of $103\pm 2^{\circ}\text{C}$. An initial wood moisture content for both ways of heating was determined 75% and for reference samples 12%. The samples for heating were macerated in water with a temperature of 20°C for 306h. The reference samples were conditioned in the SANYO MTH 2400 conditioning chamber at a temperature of $20\pm 3^{\circ}\text{C}$, a relative air humidity of $65\pm 5\%$ for 595h. The actual moisture content was checked gravimetrically each 24h using 20 representative samples for each way of heating and reference samples. After these treatments all samples were weighed and their moisture was calculated.

The plasticization process time consumption was determined as the shortest time necessary to heat the samples to the demanded temperature within their entire cross-section. The optimum wood temperature value (i.e. 75°C) was derived from the glass transition temperature of lignin, which is substantially affected by wood moisture (Bouajila *et al.* 2006). The temperature was measured by contactless infrared thermometer IR – 380 on the surface of the samples and after split apart along the longitudinal axis, also in its spatial centre of gravity. The temperature was measured gradually with the lengthening time → the first sample was heated by steaming for 1min, the second for 2min etc. The temperatures at each time were measured always using two samples. Plasticization was conducted for each sample individually. The samples were weighed after temperature measuring and their output moisture was calculated. The data on steaming and dielectric heating of the samples are presented in Table 1.

The quantification of the plasticization effect of both wood plasticization methods was performed by comparing the values of MOR, MOE and the maximum deflection in static bending (three point bend) at a plastic state (i.e. temperature $\approx 70^{\circ}\text{C}$ and MC = above fibre saturation point) with the values of these parameters measured in reference samples (i.e. temperature $\approx 20^{\circ}\text{C}$ and MC = 12%). The span of supports was 240mm. For the calculation of MOE a linear part of stress-strain curve was used, limit values were $0,1 \cdot F_{\max}$ and $0,4 \cdot F_{\max}$ with corresponding deflections. Input data for testing device software (TestXpert) application were obtained by weighing and measuring of each sample dimensions. Each sample was placed between bending grips of the

ZWICK Z 050 universal testing device and the loading was launched. The mean density of each sample during the bending test was calculated by using its weight and dimensions. The conditions of steaming and microwave plasticization of the samples are presented in Table 1.

Table 1

Directive data of steaming and microwave heating of samples

PARAMETER	STEAMING	MW HEATING
Input moisture content of specimens	≈ 75%	≈ 75%
Input temperature of specimens	≈ 15°C	≈ 15°C
Required output temperature of specimen	≈ 75°C	≈ 75°C
Relative humidity	99,99%	70 – 75%
Vapor/air temperature	100°C	35 – 40°C
Vapor/air pressure	101,325KPa	101,325KPa
Vapor/air velocity	≈ 1,5m.min ⁻¹	≈ 0,2m.min ⁻¹
Frequency of electromagnetic radiation	-	2 450MHz
Mode of magnetrons	-	Continuous wave
High-frequency power	-	1 × 900W
Power frequency	-	36KWm ⁻³
Measuring temperature interval	after 1min	up to 1min after 5s over 1min after 10s
Time since removing from device after temperature measuring	surface up to 2s center up to 20s	surface up to 2s center up to 20s

RESULTS

Density in an absolutely dry state of samples before heating

The mean density of samples for determination time of steaming in an absolutely dry state was 643kgm⁻³, and it was 644kgm⁻³ for samples for determination time of MW heating. The variation coefficients ranged around 4%. The variation coefficient of both sets of samples was nearly 13%. In case of plasticity study the mean value of density at a MC of 0% was 647kgm⁻³ in the samples for steaming, 648kgm⁻³ in the samples for MW heating and 646kgm⁻³ in the reference samples. The data variability ranged up to 6% for all sets, this agrees with Gryc *et al.* (2008).

Wood density is significant factor that affects the speed of convectional and mainly dielectric heating; therefore, the resemblance of the mean values of density between the sets of samples was verified by statistical tests. Due to the non-normal distribution of the measured data, the non-parametric Mann-Whitney U test and Kruskal-Wallis ANOVA were used. For the same reasons, the values of the descriptive statistics were transformed by the Box-Cox transformation function before presentation. The results of these tests show that the effect of the density difference in an absolutely dry state on the variable measured later can be considered insignificant with 95% reliability.

The heating curves of samples during steaming and MW heating

For the specific conditions of this experiment, the minimum time of steaming was established to 15min and the minimum time of MW heating to 150s, i.e. 2.5min. This time is shown by the vertical green dotted line in Fig. 1. Thus, the heating of samples by convectional heating takes 6 times more than the microwave heating. In the case of steam, the heat will spread the fastest in the structure with minimum distances between particles. Wood is a very slow heat conductor. The reason for the higher time consumption of steaming is the slow distribution of heat, which is caused by the considerably inhomogeneous wood structure at all levels of observation. Due to the inhomogeneity of wood at microscopic level, the paths for heat spread consist of short sections with a large (cell wall) and small (cell lumens) density of particles, which slows down the speed of

plasticization heat spread along the cross-section of samples. Partially, the speed of heat spread is increased by the moisture contained in wood, which overpasses the free spaces, but its speeding-up effect is slowed down by the high specific temperature of water.

The shape of temperature curves recorded at steaming (Fig. 1) is typical of convectional heating and shows the faster heating up of surface layers as compared to central layers, confirming the gradual penetration of heat from the surface of samples to their internal structures, as presented by other authors, e.g. Younsi *et al.* (2006), Deliiski *et al.* (2010), Zielonka and Gierlik (1999). The speed of heating decreases with the decreasing gradient between temperature of samples and steam.

At the beginning of MW heating (until about 70th second of heating) the central layers were heated up faster than the surface layers. This can potentially be related to the decrease in MC in the central layers caused by the higher temperature that push the moisture to the surface layers. The higher temperature it brings about a higher intensity of heat development.

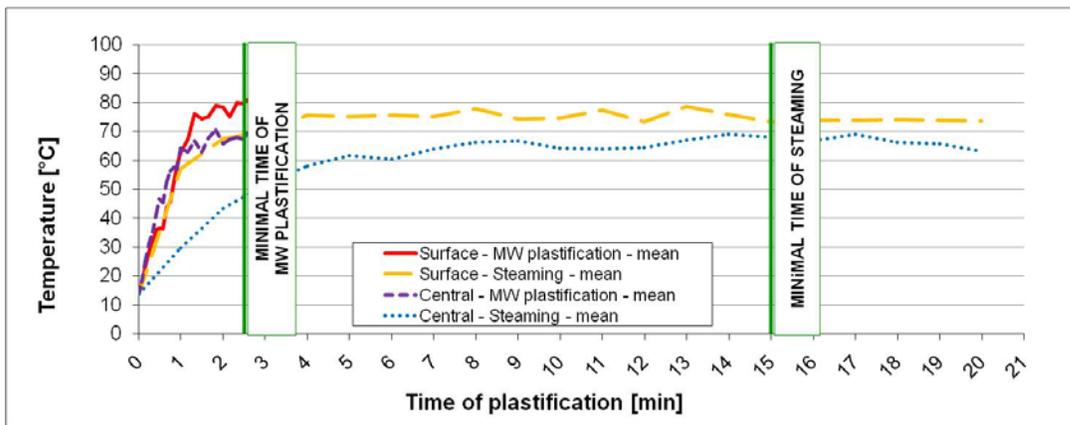


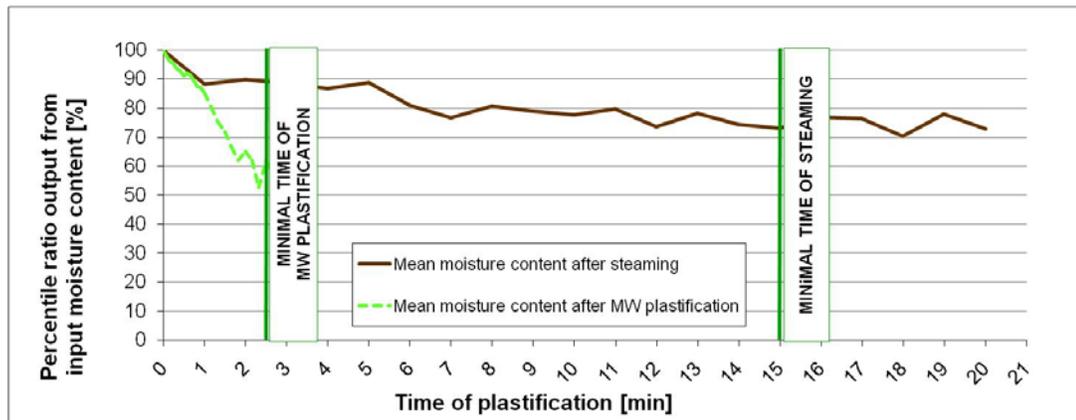
Fig. 1

Direct confrontation primary heating curves for central and surface layers of specimens plasticized by steaming and microwave heating.

With time, the curves of convectional and dielectric heating (Fig. 1) approach horizontal asymptotes, which in the case of convectional heating represent the temperature of saturated steam and in the case of dielectric heating the maximum temperature to which the water present in the structure of samples can be heated. However, the temperatures to which the heating curves of steaming and MW heating converged were lower than the theoretical temperature asymptotes, i.e. temperatures closely below 100°C. This is probably caused by the temperature decrease during the time delay between taking samples out of the plasticization device and measuring their temperature. The approach of curves to the presumed temperature asymptotes has probably occurred but a recording of this state could not be taken using the available measuring equipment. The time delay and the related temperature decrease can also clarify the gradient found between the temperatures of central and surface layers of samples in both ways of plasticization, which occurs at a certain moment.

The moisture loss curves of samples during steaming and MW heating

The curve of moisture loss during steaming shows that the MC of the samples decreased to about 75% of the original content after the process had finished, while MW heating decreased the moisture content by about a half, i.e. twice as much as steaming. Reduction of moisture content during MW heating was ten times faster than during steaming. The direct comparison of moisture loss from the structure of samples (Fig. 2) is possible due to the equal input MC of the samples.



Direct comparison of the primary curves of percentile ratio output moisture content from input moisture content at convective and dielectric heating (steaming and microwave heating).

The differing speed of moisture loss from the structure of wood can be explained by its hygroscopic properties, which mediate the effect of very different climatic conditions in the steaming chamber and cavity resonator on the resulting MC of the samples. The equilibrium moisture content (EMC) for the steaming conditions ($T = 100^{\circ}\text{C}$, relative air humidity = 99.99%) ranges around the saturation point (i.e. around 30%); the conditions in the cavity resonator during heating ($T = 35$ to 40°C , relative air humidity = 70 to 75%) approximately correspond to 12% EMC. The velocity of moisture loss during MW heating is considerably increased by the intensive development of heat in the structure of the samples. The considerably faster loss in moisture, i.e. also plasticity during MW heating of the samples, brings about the necessity to meet the established optimum time for plasticization. As the MC during steaming can only decrease to the saturation point, a longer time of steaming does not cause any undesirable changes in moisture. However, it will cause irreversible chemical changes in the wood structure that can, according to Kúdela (2009), be manifested by a darker shade, a slight increase in EMC, a partial deterioration of strength properties even after stabilization etc. Therefore, both plasticization technologies can be characterized by plasticity curves with a maximum, the MW heating having a markedly sharper maximum.

Plasticity parameters of samples measured in static bending

Through both methods of plasticization of the samples, the MOR decreased to about 25% of the solid wood MOR (Table 2). The MOR variability increased by about 4% after plasticization both by steaming and by microwaves (Table 2). The difference between the MOR of plasticized sets and the reference samples was assessed by the parametric 1-F ANOVA and Tukey HSD test as statistically very significant at a level of significance $\alpha = 0.05$ (Fig. 3). The difference between steamed and MW heated samples is statistically insignificant (Fig. 3).

The MOE, or the stiffness of the samples, decreased even more than strength – to about 20% of the value measured in reference samples, i.e. by about 80% in both the analysed ways of plasticization (Table 2). The variability of MOE increased three times after plasticization (Table 2). The non-parametric Kruskal-Wallis ANOVA and the Tukey modified HSD test assessed the fivefold decrease in MOE after modification as a statistically significant difference (Fig 4a). The difference between the mean values of steamed and MW heated samples is statistically insignificant at a level of significance $\alpha = 0.05$ (Fig 4a).

The maximum deflection of the samples after plasticization did not change so considerably as the previous parameters. In the case of steaming, it increased by about 42%. In the case of MW heated samples, the increase was about 46% (Table 2). The box plot in Fig 4b shows a marked variability of the measured data. The non-parametric tests assessed the increase in the maximum deflection of samples after plasticization as statistically significant for both methods of heating (Fig 4b). The difference between the methods was not proved at a level of significance $\alpha = 0.05$.

Table 3

Basic descriptive statistics for measured criteria of plasticity of specimens

MODIFICATION	NUMBER OF SPECIMENS	MEAN VALUE	LOWER CONFIDENCE INTERVAL	UPPER CONFIDENCE INTERVAL	MEDIAN	STANDARD DEVIATION	VARIATION COEFFICIENT	NORMALITY
MODULUS OF RUPTURE OF SPECIMENS [MPa]								
Steaming	100	30,2	29,5	30,8	30,4	3,5	11,5	YES
Microwave heating	100	30,5	29,7	31,2	30,2	3,6	11,9	YES
Reference	100	118,2	116,4	120,1	118,7	9,4	8,0	YES
MODULUS OF ELASTICITY OF SPECIMENS [MPa]								
Steaming	100	2215,9	2099,9	2330,8	2287,7	587,3	26,7	NO
Microwave heating	100	2175,3	2067,6	2281,5	2209,2	547,3	25,4	NO
Reference	100	11028,8	10844,7	11212,9	11110,5	927,8	8,4	YES
MAXIMAL DEFLECTION OF SPECIMENS IN STATIC BEND [mm]								
Steaming	100	15,5	14,5	16,5	15,8	5,3	32,3	NO
Microwave heating	100	16,7	15,8	17,8	17,1	5,3	30,9	NO
Reference	100	9,0	6,2	13,6	7,5	1,3	17,1	NO

▨ Values gained by Box-Cox transformation from abnormal values

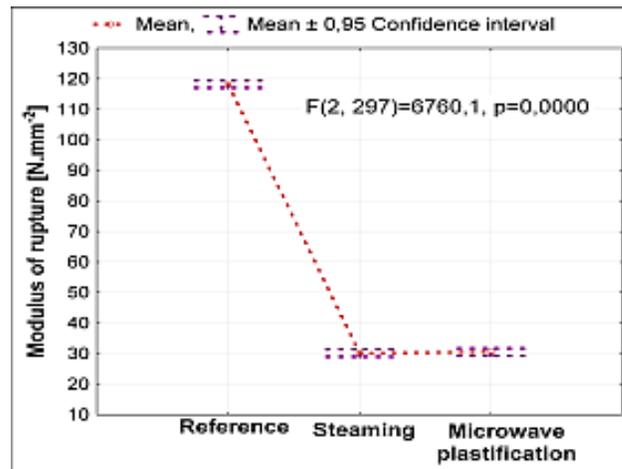


Fig. 3
Parametric 1 - F ANOVA – Modulus of rupture.

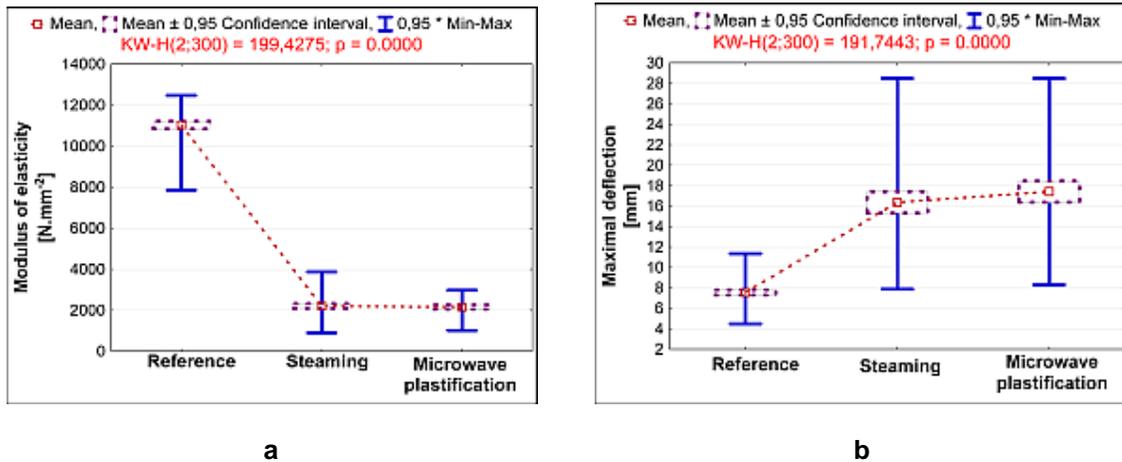


Fig. 4
Kruskal - Wallis ANOVA
a - Modulus of elasticity; b - Maximal deflection of specimens.

The available basic information on material plasticity indicates that an increase in plasticity should be accompanied by an increase in the amount of energy the sample is able to absorb for its plastic transformation. For this reason, the amount of absorbed energy needs to be assessed together with the work diagram analysis (Fig. 5), where the part of the absorbed energy used for elastic and plastic transformation is well distinguishable. In this context, we can say that in spite of the nearly same total amount of energy absorbed by the plasticized samples (the area below tension-deformation graph), their plasticity increased. The proof is the minimization of the linear part of the work diagram.

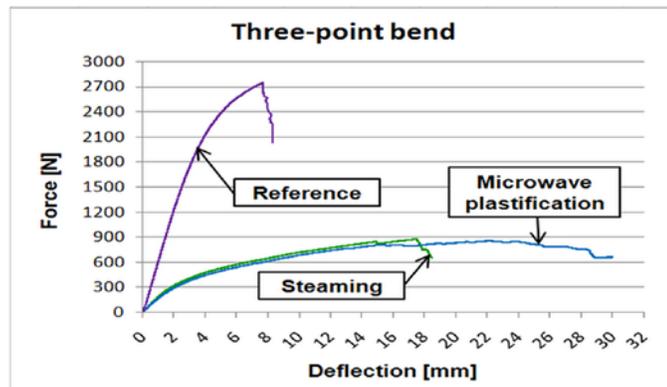


Fig. 5
Confrontation of work diagrams for three - point bend variously modified specimens.

CONCLUSION

1. The results of the presented experiment confirmed the assumed lower time consumption of dielectric heating of wooden elements when compared with convectional heating. The time for the specific conditions of sample steaming was set to 15min. The optimum time for MW heating of the samples was found to be 2.5min, which is 6 times shorter than the time for steaming. The main reason for this very different heating time can be found in the completely different principle of heat origination and distribution within the internal structure of the samples.

2. In both cases, the output moisture content was lower than the input moisture content. The moisture content of the samples dropped to a half of the original content in the case of MW heating, which is a double moisture loss when compared with steaming.

3. The plasticization of the samples caused practically and statistically significant changes in the wood structure. The difference between the results of the two analyzed plasticization technologies is always insignificant. This can be caused by the insignificant differences in the plasticity indicator parameters (temperature and moisture content) of samples after plasticization. Only in the case of the maximum deflection, the samples plasticized by MW heating have a slightly higher plasticity. The cause can be the faster cooling down of surface layers of the steamed samples due to the immense evaporation of moisture from slightly oversaturated surface layers.

4. Besides the shorter time needed for plasticization, the MW heating also saves energy necessary for the heating of wooden elements, which follows from the principle of MW heating, when the EMG energy is consumed, or rather transformed to thermal energy only within the heated elements, while in steaming also all the plasticization space is heated up, including the walls of the steaming chamber. Last but not least, the MW heating contributes to better working conditions for workers. On the other hand, the devices for microwave usage for plasticization still have many drawbacks when compared to the devices for steaming.

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