MATHEMATICAL DESCRIPTION OF THE LATENT HEAT OF BOUND WATER IN WOOD DURING FREEZING AND DEFROSTING

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ABSTRACT

Some basic characteristics and terms of the specific latent heat of crystallization of the water in wood and of melting the ice formed by it are presented in the paper. The change in the temperature of ice and water during their heating and cooling occurred in the phase transitions were described and analyzed. Using data from the specialized literature a mathematical description of the specific latent heat of crystallization and melting the bound water in wood were suggested. Based on that description, an update of an available equation for the specific heat capacity of the frozen bound water in wood above the hygroscopic range was carried out. The information about the specific latent heat and specific heat capacity of the water in wood materials is needed for computing the non-stationary temperature distribution and energy consumption during their thermal treatment, and also for the model based automatic control of that treatment.

Key words: latent heat, mathematical description, specific heat capacity, wood, bound water, freezing, defrosting.

INTRODUCTION

It is known that latent heat is the thermal energy absorbed or released by a body or a thermodynamic system during a constant-temperature process of phase transition of a given substance (https://www.britannica.com/science/latent-heat, https://www.engineeringtoolbox.com/latent-heat-melting-solids-d_96.html). During such a process, the temperature of the system stays constant as heat is added: the system is in a mixed-phase regime in which some parts of the system have completed the transition and others have not. A familiar example is the melting of ice, which does not convert suddenly into a liquid but for a definite period of time there is a mixture of crystals and liquid water.

The term "latent" was introduced around 1762 by British chemist Joseph Black. It is derived from the Latin *latere*, which means *to lie hidden* (https://en. wikipedia.org/ wiki/Water/data_page). Consequently, the latent heat represents a thermal energy in hidden form, which is supplied or extracted to change the aggregate state of a given substance without changing its temperature.

During the calculation of the duration and energy consumption of the defrosting processes of frozen wood materials, the heat needed to carry out the phase transition, i.e. for the melting of the ice, formed both from the free water, as well as from the frozen part of the

bound water in the wood must be taken into account. When relating these heat consumptions to 1 K they can be expressed by their corresponding specific heat capacities, i.e. through the specific heat capacities of the frozen free water, c_{ice-fw}, and of the frozen bound water, c_{ice-bw}, in the wood.

In the expressions to determine c_{ice-fw} and c_{ice-bw} in the specialized scientific literature the latent heat of the free and of the bound water in wood participate respectively (CHUDINOV 1966, STEINHAGEN 1986, 1991, STEINHAGEN – LEE 1988, GOSS – MILLER 1989, SHUBIN 1990, KHATTABI – STEINHAGEN 1992, 1993, 1995, POŽGAJ et al. 1997, SIMPSON – TENWOLDE 1999, TREBULA – KLEMENT 2002, VIDELOV 2003, DELIISKI 2004, 2009, 2011, 2013, PERVAN 2009, DELIISKI – DZURENDA 2010, DZURENDA – DELIISKI 2011, DELIISKI et al. 2013, 2015, 2019, HADJISKI – DELIISKI 2015, 2016, DELIISKI – TUMBARKOVA 2017a, 2017b, 2018, 2019). According to these expressions, there is widely accepted that the latent heat of both the free and bound water in the wood is the same and it is equal to $3.34 \cdot 10^5$ J·kg^{-1}.

The research on freezing water and melting the ice formed from it in different capillary-porous materials shows that the latent heats of the free and bound water in these materials differ from one another quantitatively (EFIMOV 1985, LANGE et al. 1994, CHO et al. 1996, OLIEN – LIVINGSTON 2006, SZEDLAK et al. 2009, KIANI – SUN 2011, KRANIOTIS – KRISTINE 2017). That gives us reason to investigate the difference between the latent heat of free and of bound water in wood, which are typical representatives of capillary-porous materials.

The aim of the present paper is to suggest a mathematical description of the latent heat of the bound water in wood and to incorporate it in an updated version of the equation to determine the specific heat capacity of the frozen bound water in wood.

MATERIAL AND METHODS

Basic characteristics and terms of the latent heat

When a substance changes phase, i.e. it goes from either a solid to a liquid or a liquid to a gas state, it requires energy to do so. The potential energy stored in the interatomic forces and between molecules needs to be overcome by the kinetic energy of the motion of the particles before the substance can change phase.

In Figure 1, example schemes of the change in the temperature T of the ice during its heating and also of water during its freezing are given.

Starting at point A (see Fig. 1-left), the ice was in its solid phase, heating it by the energy Q brings the temperature up to its melting point but the ice was still in a solid state at point B. As it was heated further, the energy from the heat source Q went into breaking the bonds holding the atoms in place. This took place from B to C where the water absorbed the energy Q_{Lat} without a change of its temperature. At point C the whole amount of the ice was transformed into water. The further heat addition from C to D went into the kinetic energy of the water, which caused an increase in its temperature.

During cooling the water (see Fig. 1-right) its temperature decreased from A to B. When the melting point was reached in B, a crystallization of the water started. From B to C, the freezing of the whole amount of the water into ice occurred without change in its temperature. During that process a release of the absorbed energy Q_{Lat} into the surrounding was carried out. The further cooling of the ice from C to D causes a decrease in its temperature.

In the International System of measuring units (SI) the latent heat is represented by the parameter of specific heat of phase transition, which is also called by the term specific latent heat L. It is expressed by the amount of energy in the form of heat Q (in J), which is required to completely effect a phase change of mass m (1 mol or 1 kg) of a given substance, i.e.
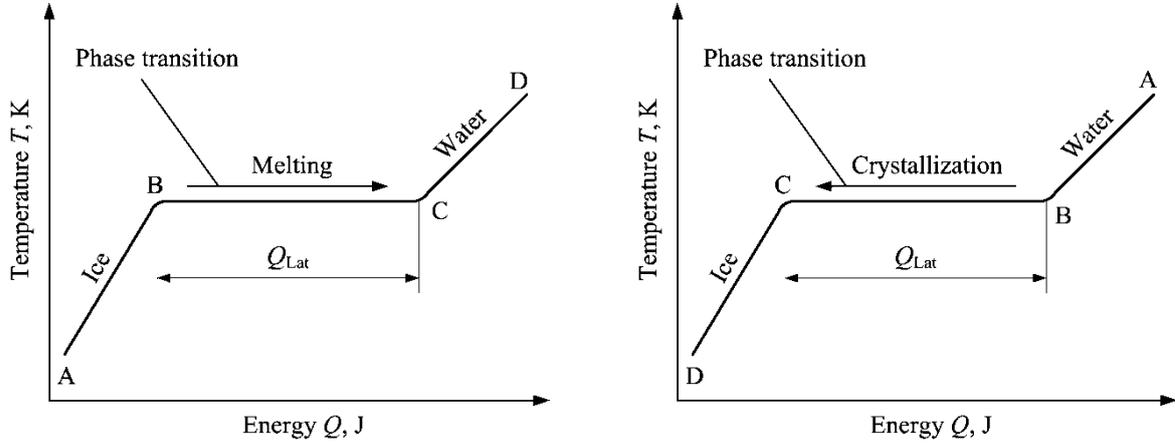
Fig. 1 Change in T of ice during its heating (left) and change in T of water during its freezing (right).

$$L = \frac{Q}{m} \qquad (1)$$

Using eq. (1), the thermal energy denoted as Q_{Lat} can be determined, as follows:

$$Q_{Lat} = m \cdot L \qquad (2)$$

where Q_{Lat} is the amount of the energy absorbed or released during the change of phase of the substance, J; m – mass of the substance, mol or kg; L – specific latent heat for a particular substance, J·mol^{-1} or J·kg^{-1}.

In the specialized literature the following indexed options of L are mostly used:

• L_f – specific latent heat of fusion or melting. It is used in models of processes with phase transitions from solid to liquid state of the substances;

• L_{cr} – specific latent heat of crystallization. It is used in models of processes with phase transitions from liquid to solid state of the substances.

• L_v – specific latent heat of vaporization. It is used in models of processes with phase transitions from liquid to gaseous state of the substances.

• L_c – specific latent heat of condensation or liquefaction. It is used in models of processes with phase transitions from gaseous to liquid state of the substances.

Dependence of the latent heat of water on the temperature

In the specialized literature it has been proven that the latent heat L of a given substance is equal to the difference ΔH between the enthalpy of the two phases of the substance (EFIMOV 1985, SZEDLAK et al. 2009, PAHI 2010, https://en.wikipedia.org/wiki/ Latent_heat). In our case of freezing of the water in wood and melting of the ice in wood this means that

$$L_f = L_{cr} = \Delta H \qquad (3)$$

where

$$\Delta H = H_{water} - H_{ice} \qquad (4)$$

is the difference between the enthalpies of the water, H_{water}, and the ice, H_{ice}, J·kg^{-1}.
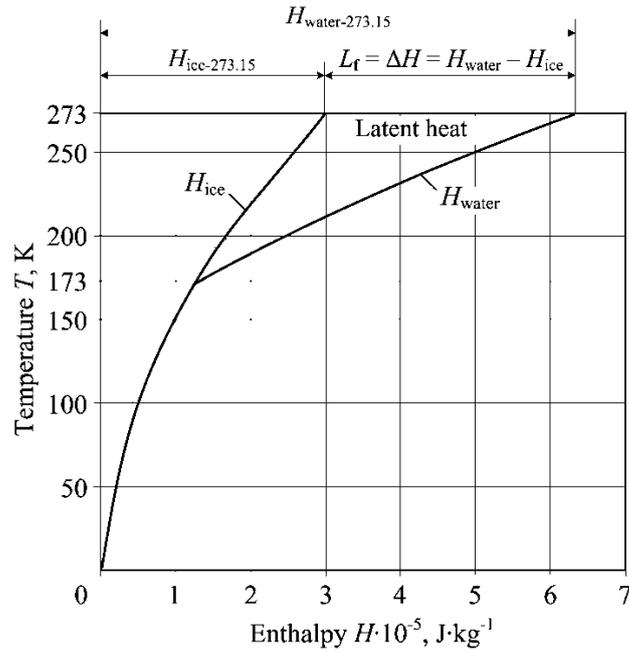
Fig. 2 Change in ΔH between water and ice, depending on *T* (acc. OLIEN and LIVINGSTON 2006).

The difference $L_f = \Delta H$ can be determined using the graphs given in Fig. 2.

The temperature dependence of the difference in enthalpy between water and ice at a constant pressure *p* can be expressed by the following equation of Kirchhoff (SZEDLAK *et al.* 2009, PAHI 2010, https://www.physics.info/heat-latent/):

$$\left(\frac{\partial \Delta H}{\partial T}\right)_p = \Delta c_p \tag{5}$$

where

$$\Delta c_p = c_{water} - c_{ice} \tag{6}$$

is the difference between the specific heat capacities of the liquid water, c_{water}, and the ice, c_{ice}, at atmospheric pressure, $J.kg^{-1}\cdot K^{-1}$.

Taking into account eqs. (4) and (6), for the case of the ice melting, the solution of eq. (5) is equal to (SZEDLAK *et al.* 2009)

$$L_f(T) = L_f(T_f) - \int\limits_{T}^{T_f} \left(c_{water} - c_{ice}\right) dT \tag{7}$$

where L_f is the specific latent heat of melting of the ice, $J \cdot kg^{-1}$; T_f – temperature at the point of the ice melting, K; *T* – temperature lower then T_f, K; c_{water} and c_{ice} – specific heat capacities of the liquid water and ice respectively, $J \cdot kg^{-1} \cdot K^{-1}$.

In Fig. 3, experimental data given in SZEDLAK *et al.* (2009) for L_f of overcooled water in the range from 236 to 263 K are presented. They were obtained by three scientific teams using different measuring methods and equipment. In that Figure, the change in L_f depending on *T* calculated by eq. (7) is also given.
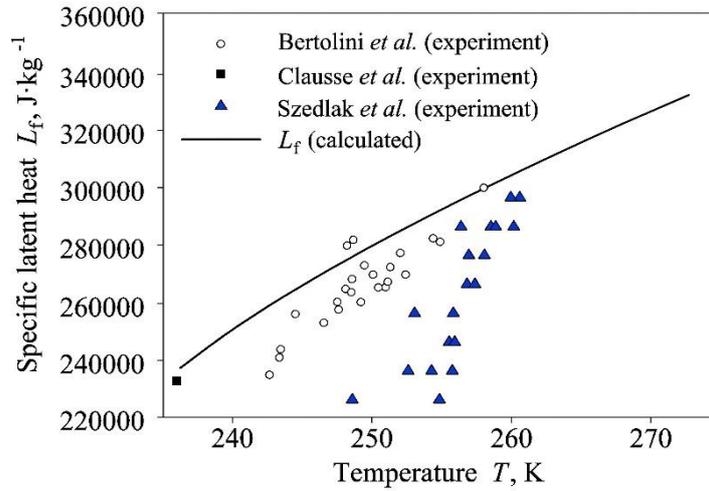
Fig. 3 Change in L_f of water, depending on T (acc. SZEDLAK *et al.* 2009).

Latent heat of water in wood during its freezing and defrosting

The information about the change in the specific latent heat of crystallization and melting of both the free and bound water in wood materials is of significant interest during the modeling and model-based automatic control of the freezing and defrosting processes of these materials.

As it was noted in the Introduction, in the specialized scientific literature it is widely accepted that the specific latent heat of crystallization of the free water and of melting of the ice formed by it in capillary porous materials, including wood, do not depend on the temperature and are equal to, as follows:

$$L_{cr-fw} = L_{f-fw} = 3.34 \cdot 10^5 \ \text{J} \cdot \text{kg}^{-1}. \tag{8}$$

The reason for the constant value of L_{cr-fw} and L_{f-fw} is the circumstance that the free water freezes at constant temperature, equal to 273.17 K, i.e. 0 °C or in very narrow temperature range below 273.15 K.

The temperature conditions for the freezing of the bound water differ significantly from these for the freezing of the free water. It is known that the bound water freezes gradually in a very wide range below 273.15 K and even at extremely low climatic temperatures on the earth part of it remains in a liquid state in the wood.

During studying of the impact of T on the latent heat of melting of the ice at constant pressure and temperatures below 273.15 K, EFIMOV (1985) derived the following equation, which represents a modification of the classical equation of Clausius-Klapeyron:

$$\frac{\partial L}{\partial T} = \Delta c_p + \frac{L}{T} \tag{9}$$

If the condition $\Delta c_p = \text{const}$ is fulfilled, the solution of eq. (9) is equal to

$$L_{f-bw} = L_{f-fw} \cdot \frac{T}{T_{f-fw}} + \Delta c_p \cdot T \cdot \ln \frac{T}{T_{f-fw}} \tag{10}$$

where $T_{f-fw} = 273.15$ K is the melting temperature of the ice formed from the free water at atmospheric pressure, K; T – temperature of overcooling of the water subjected to freezing: $T < T_{f-fw}$, K; L – latent heat of melting of the ice formed from bound water at temperature T,

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J·kg^{-1}; $L_{\text{f-fw}} = 3.34 \cdot 10^5$ J·kg^{-1} – latent heat of melting of the ice formed from the free water; Δc_{p} – difference between the specific heat capacities of the water and ice at atmospheric pressure, J·kg^{-1}·K^{-1}.

According to EFIMOV (1985), equation (10) can be used for the calculation of the latent heat of melting of the bound water, $L_{\text{f-bw}}$, in capillary-porous materials.

The results obtained by the equation (10) show that a decrease in T causes a decrease in $L_{\text{f-bw}}$, which could be explained with the circumstance that the absorption of the bound water in capillary-porous materials including wood is accompanied with heat release and with an increase in the alignment of the water molecules in the materials.

RESULTS AND DISCUSSION

After substitution in eq. (6) of $c_{\text{water}} = 4218$ J·kg^{-1}·K^{-1} and $c_{\text{ice}} = 2116$ J·kg^{-1}·K^{-1} (https://en. wikipedia.org/wiki/Water/data_page), $\Delta c_{\text{p}} = 2102$ J·kg^{-1}·K^{-1} at atmospheric pressure and temperature $T = 273.15$ K (i.e. at 0 ℃) was obtained.

Replacing in eq. (10) the values of $T_{\text{f-fw}} = 273.15$ K, $L_{\text{f-fw}} = 3.34 \cdot 10^5$ J·kg^{-1}, and $\Delta c_{\text{p}} = 2102$ J·kg^{-1}·K^{-1}, the following equation was obtained for the calculation of the specific latent heat of melting the ice formed from the bound water in wood materials:

$$L_{\text{f}-\text{bw}} = 1.223 \cdot 10^3 T + 2.102 \cdot 10^3 T \cdot \ln \frac{T}{273.15} \qquad (11)$$

Update of the mathematical description of the specific heat capacity of frozen bound water in wood above the hygroscopic range

According to DELIISKI (2004, 2009, 2011, 2013) the suggested mathematical descriptions of the effective specific heat capacities of wood during its freezing and defrosting, the following equations for the calculations of the specific heat capacities of the frozen free and bound water in wood above the hygroscopic range, $c_{\text{ice-fw}}$ and $c_{\text{ice-bwm}}$ respectively, were given in DELIISKI et al. (2019):

$$c_{\text{ice-fw}} = 3.34 \cdot 10^5 \frac{u - u_{\text{fsp}}^{272.15}}{1 + u} \qquad (12)$$

$$c_{\text{ice-bwm}} = 1.8938 \cdot 10^4 \left(u_{\text{fsp}}^{272.15} - 0.12 \right) \cdot \frac{\exp\left[0.0567 \left(T - 272.15 \right) \right]}{1 + u} \qquad (13)$$

$$u_{\text{fsp}}^{272.15} = u_{\text{fsp}}^{293.15} + 0.021 \qquad (14)$$

where $c_{\text{ice-fw}}$ is the specific heat capacity of the frozen free water in wood, J·kg^{-1}·K^{-1}; $c_{\text{ice-bwm}}$ – specific heat capacity of the frozen bound water in wood when the maximum possible amount of bound water in the wood species is present, J·kg^{-1}·K^{-1} (that condition is always fulfilled above the hygroscopic range); u – wood moisture content, kg·kg^{-1}; $u_{\text{fsp}}^{293.15}$ – standardized value of the fiber saturation point at $T = 293.15$ K (i.e. at $t = 20$ ℃), and $u_{\text{fsp}}^{272.15}$ is the fiber saturation point at $T = 272.15$ K (i.e. at $t = -1$ ℃), kg·kg^{-1}. At that temperature, melting the frozen bound water in wood is fully completed and melting the free water in wood starts during heating the frozen wood (DELIISKI – TUMBARKOVA 2017a, 2017b, 2018, 2019).

The multiplier $3.34 \cdot 10^5$ in eq. (12) is the value of the specific latent heat of the free water in wood (refer to eq. (8)), which is needed for the phase transition of 1 kg liquid free water into ice.

The number $1.8938 \cdot 10^4$ in the right part of eq. (13) represents the result from multiplying of the number 0.0567 by $3.34 \cdot 10^5$, which was obtained in DELIISKI (2011, 2013) during the mathematical description of $c_{\text{ice-bwm}}$. That description was based on the widely accepted assumption that the specific latent heats of melting both the bound and free water in wood are the same and equal to $3.34 \cdot 10^5$ J.kg^{-1}.

Using eq. (11), the following update and more precise version of eq. (13) was suggested in the present paper:

$$c_{\text{ice-bwm}} = \left(69.344\,T + 119.183\,T \cdot \ln \frac{T}{273.15} \right) \cdot \left(u_{\text{fsp}}^{272.15} - 0.12 \right) \cdot \frac{\exp\left[0.0567 \left(T - 272.15 \right) \right]}{1+u} \quad (15)$$

The expression given in the first brackets in eq. (15) was obtained after multiplying of eq. (11) by the number of 0.0567.

In order to solve the eqs. (11), (12), (14), and (15) with different values of the variables in them, a software program was prepared, which was an input in the calculation environment of Visual FORTRAN Professional. With the help of that program, calculations were carried out for the determination of L_f, $L_{f\text{-bw}}$, $c_{\text{ice-fw}}$, and $c_{\text{ice-bwm}}$.

In Fig. 4 the change in the specific latent heat L_f in the range from 233.15 to 273.15 K (i.e. from –40 to 0 $^{\circ}$C) was given, which was calculated according to 3 different approaches.

The calculated by SZEDLAK *et al.* (2009) according to eq. (7) change in L_f is shown in Fig. 4 through the top line "Szedlak et al.".

Through the middle line „Acc. eq. (11): $L_{f\text{-bw}}$" the calculated by us according to eq. (11) change in $L_{f\text{-bw}}$ is given.

The bottom line "Olien & Livingston" shows the change in L_f, which was obtained according to eqs.(3) and (4) using the graphs of $H_{\text{water}} = f(T)$ and $H_{\text{ice}} = f(T)$ given in Fig. 2.

The comparison of the graph of $L_{f\text{-bw}}$ in Fig. 4 with the two graphs of L_f shows well their compliance with each other. The slope of $L_{f\text{-bw}}$ occupied an intermediate place between the slopes of L_f determined by SZEDLAK *et al.* (2009) and OLIEN – LIVINGSTON (2006). The differences between the analogous values of $L_{f\text{-bw}}$ and L_f in the studied temperature range were as follows: $\pm$ 9 at $T = 243.14$ K; $\pm$ 6% at $T = 253.15$ K, ± 3 % at $T = 263.15$ K and 0% at $T = 273.15$ K.

It can be noted, that the graph of $L_{f\text{-bw}}$ in Fig. 4 is more relevant to the experimental data in Fig. 2 in comparison to both graphs of L_f in Fig. 4. This fact gave reason to recommend eq. (11) for implementation as a mathematical description of $L_{f\text{-bw}}$ in the models of wood thermal treatment processes, which include freezing and defrosting of materials from different wood species.

In Figure 5 the calculated according to eq. (12) change in the specific heat capacity of the frozen free water, $c_{\text{ice-fw}}$, in beech and poplar wood, depending on the wood moisture content u in the range from $u = u_{\text{fsp}}^{272.15}$ to $u = 1.0$ kg·kg^{-1} is presented. For the determination of $u_{\text{fsp}}^{272.15}$ according to eq. (14) the standardized values of the fiber saturation point $u_{\text{fsp}}^{293.15} = 0.31$ kg·kg^{-1} for the beech wood and $u_{\text{fsp}}^{293.15} = 0.35$ kg·kg^{-1} for the poplar wood was used (VIDELOV 2003, DELIISKI – DZURENDA 2010).
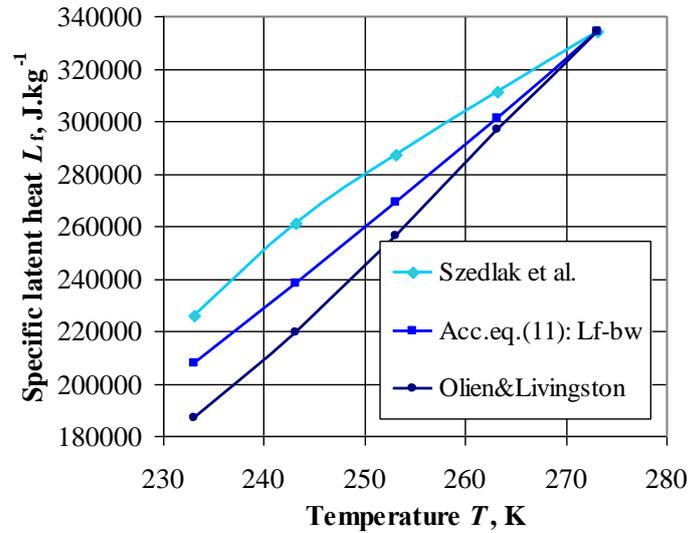
Fig. 4 Change in L_f at P = const, depending on T
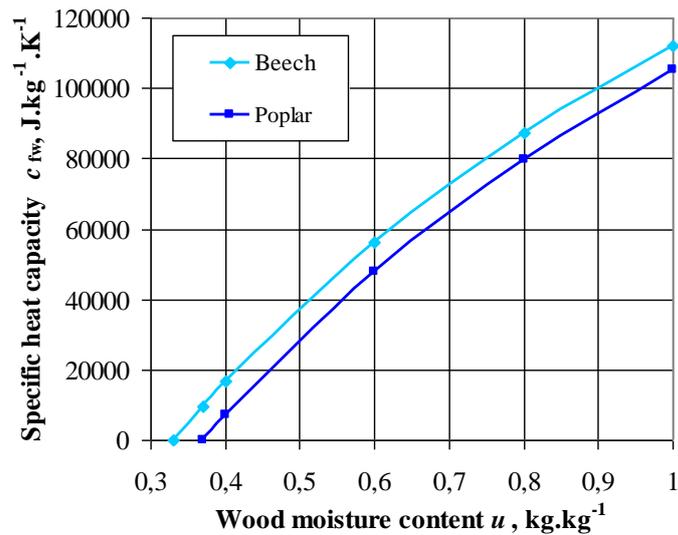
Fig. 5 Change in $c_{\text{ice-fw}}$ of beech and poplar wood, depending on u.

In Figures 6 and 7, the calculated according to eqs. (13) and (15) change in the specific heat capacity of the frozen bound water, $c_{\text{ice-bw}}$, in beech and poplar wood respectively, depending on the temperature in the range from 213.15 to 272.15 K (i.e. from –60 °C to –1 °C) is shown. The derived by eqs. (13) and (15) lines of $c_{\text{ice-bw}}$ have labels "old" and "updated" respectively in the legend of these figures.

The analysis of the obtained simulation results, part of which are presented in Fig. 5 to Fig. 7 lead to the following statements:

1. The specific heat capacity of the frozen free water, $c_{\text{ice-fw}}$, increases curvilinearly from 0 to 111890 J·kg^{-1}·K^{-1} for the beech wood and from 0 to 105210 J·kg^{-1}·K^{-1} for the poplar wood when u increases from $u = u_{\text{fsp}}^{272.15}$ to $u = 1.0$ kg·kg^{-1}.
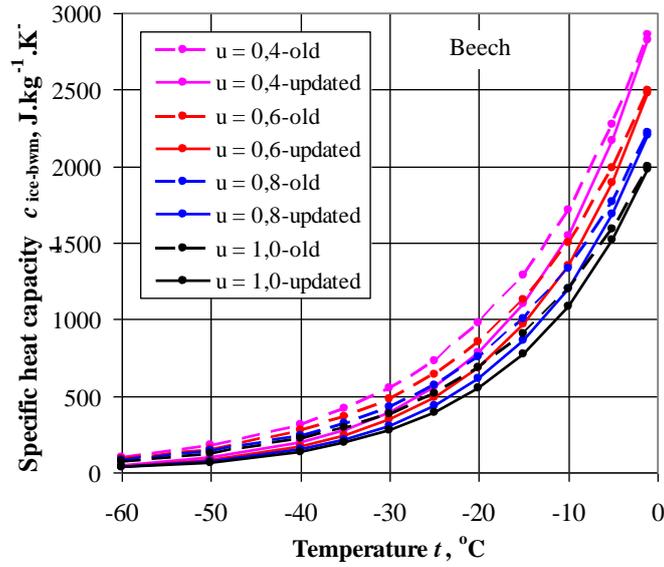
Fig. 6 Change in $c_{\text{ice-bw}}$ of beech wood, depending on t.
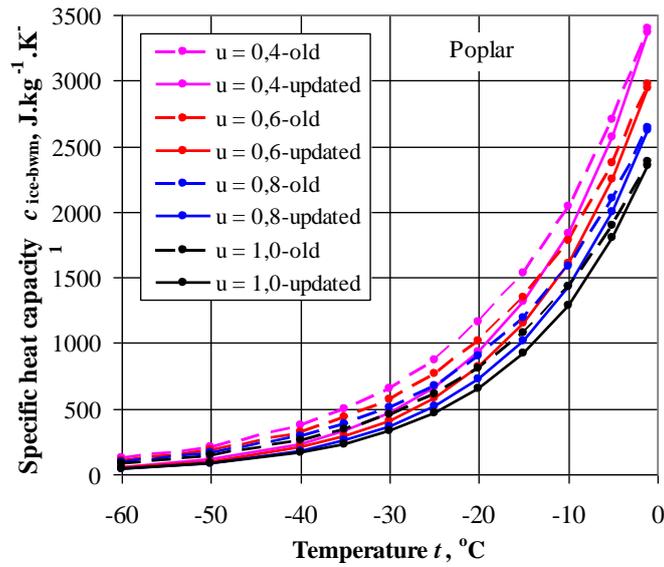
Fig. 7 Change in $c_{\text{ice-bw}}$ of poplar wood, depending on t.

2. An increase in the standardized value of the fibre saturation point of the wood $u_{\text{fsp}}^{293.15}$ causes a decrease in $c_{\text{ice-fw}}$ because of the circumstance that with an increase in $u_{\text{fsp}}^{293.15}$ the amount of the free water in the wood equal to $u - u_{\text{fsp}}^{272.15}$ decreases (see eqs. (12) and (14)).

Each increase in $u_{\text{fsp}}^{293.15}$ by 0.01 kg·kg^{-1} causes an exponential increase in the specific heat capacity of the frozen bound water $c_{\text{ice-bwm}}$. When t increases from –60 ºC to –1 ºC, $c_{\text{ice-bwm}}$ increases several tens of times at the given value of the wood moisture content u. At $t = -1$ ºC the capacity $c_{\text{ice-bwm}}$ reaches its maximum values, which according to eq. (15) are equal to the following:

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• at $u = 0.4$ kg·kg^{-1}: $c_{\text{ice-bw}} = 2826$ J·kg^{-1}·K^{-1} for beech and $c_{\text{ice-bw}} = 3362$ J·kg^{-1}·K^{-1} for poplar wood;

• at $u = 0.6$ kg·kg^{-1}: $c_{\text{ice-bw}} = 2473$ J·kg^{-1}·K^{-1} for beech and $c_{\text{ice-bw}} = 2942$ J·kg^{-1}·K^{-1} for poplar wood;

• at $u = 0.8$ kg·kg^{-1}: $c_{\text{ice-bw}} = 2198$ J·kg^{-1}·K^{-1} for beech and $c_{\text{ice-bw}} = 2615$ J·kg^{-1}·K^{-1} for poplar wood;

• at $u = 1.0$ kg·kg^{-1}: $c_{\text{ice-bw}} = 1978$ J·kg^{-1}·K^{-1} for beech and $c_{\text{ice-bw}} = 2354$ J·kg^{-1}·K^{-1} for poplar wood.

4. The increase of u causes a decrease in $c_{\text{ice-bwm}}$. When u at a given t increases from 0.4 to 1.0 kg·kg^{-1}, $c_{\text{ice-bwm}}$ decreases by 30.0% for both the beech and poplar wood. This means that each increase of u by 0.01 kg·kg^{-1} reduces $c_{\text{ice-bwm}}$ by approximately 0.5%.

5. An increase in $u_{\text{fsp}}^{293.15}$ for separate tree species causes a linear increase in $c_{\text{ice-bw}}$. Each increase in $u_{\text{fsp}}^{293.15}$ by 0.01 kg·kg^{-1} causes an increase in $c_{\text{ice-bwm}}$ by approximately 4%.

6. The calculated by eq. (15) updated values of $c_{\text{ice-bwm}}$ are found to be smaller in the range of up to 20% compared to the analogous old values of $c_{\text{ice-bwm}}$, which are calculated by eq. (13) (see Fig. 6 and Fig. 7). This means that the application of the more precise eq. (15) instead of eq. (13) will lead to smaller calculated values of the energy, which is needed for the melting of the frozen bound water in wood materials during their thermal treatment.

CONCLUSIONS

Some basic characteristics and terms of the specific latent heat of crystallization of the water in wood and of melting of the ice are being considered in this work. Based on the analysis of data in the specialized literature and in the Internet sources the following were determined:

• the specific latent heat of fusion and crystallization of free water in wood does not depend on the temperature and is equal to $L_{\text{f-fw}} = L_{\text{cr-fw}} = 3.34 \cdot 10^5$ J·kg^{-1};

• the specific latent heat of fusion and crystallization of the bound water in wood, $L_{\text{f-bw}} = L_{\text{cr-bw}}$, decreases with a decrease in the temperature T. This can be explained with the circumstance that the absorption of the bound water in capillary-porous materials, including wood, is accompanied with heat release and with an increase in the alignment of the water molecules in the materials.

• an equation was derived, which can be used as a mathematical description of the specific latent heat $L_{\text{f-bw}} = L_{\text{cr-bw}}$ in wood materials during the phase transition of the bound water in them. That equation is based on the classical equation of Clausius-Klapeyron, which has been modified by EFIMOV (1985).

Based on that description, an update of the available equation for determination of the specific heat capacity of the frozen bound water in wood above the hygroscopic range, $c_{\text{ice-bwm}}$, were carried out. The calculated by that equation updated values of $c_{\text{ice-bwm}}$ are found to be smaller in the range of up to 20% compared to the analogous values of $c_{\text{ice-bwm}}$, which are calculated by a previous version of that equation. The application of the more precise updated equation for $c_{\text{ice-bwm}}$ will lead to smaller calculated values of the energy, which is needed for the melting of the frozen bound water in wood materials during their defrosting.

The obtained results can be used for scientifically based determination of the specific latent heats $L_{\text{f-fw}} = L_{\text{cr-fw}}$, $L_{\text{f-bw}} = L_{\text{cr-bw}}$, and also of the specific heat capacity of the frozen bound water $c_{\text{ice-bwm}}$ in mathematical models of freezing and defrosting processes of wood

materials. They could support the development of improved software of systems for model-based automatic control of such processes (HADJISKI – DELIISKI 2016).

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