ON THERMAL PROPERTIES OF A PYROLYSING WOOD PARTICLE

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ABSTRACT

Among the physical properties, density and thermal conductivity cause the highest sensitivity of pyrolysis model predictions. The limits of detailed models lie in the use of questionable kinetics or arbitrary values of physical properties. Hence, a better knowledge of decomposition kinetics and the physical properties of biomass fuels can improve the predictive capabilities of transport models.

The present paper clarifies thermal properties of wood and wood char. These properties are thermal conductivity and specific heat capacity and focus is on their dependence on temperature. The main (though not exclusive) emphasis is given to Finnish softwood, namely pine, although some properties of other wood species are also presented.

Experiments were conducted on the thermal conductivity of pine and pine-derived char. The results of the experiments are reported and they are compared to values found in the literature. Finally a draft correlation for the thermal conductivity of a pyrolysing wood particle is suggested.

Keywords: wood, pine, pyrolysis, thermal conductivity, specific heat capacity

INTRODUCTION

Pyrolysis is the thermal decomposition of a solid fuel in an inert atmosphere. It is a preceding step for gasification and combustion although it can also be an independent conversion process. Biomass pyrolysis products are tarry liquid (bio oil), char and non–condensable gases. The amounts and compositions of the products depend on temperature, pressure and heating rate. Bio oil yield maximization requires a high heating rate, a temperature of approximately 773 K, and a rapid cooling of the gases and aerosols that are formed in pyrolysis. (Bridgewater and Peacocke, 2000)

The pyrolysis rate is controlled by chemical kinetics, heat transfer or mass transfer. As particle size increases, the time required for heat and mass transfer increases until these processes become rate controlling processes instead of chemical kinetics. Isothermal assumption is no longer valid with biomass particles exceeding 250 µm (Yang et al., 2008), and in practice the heating rate of wood is limited by its low thermal conductivity (Di Blasi, 2008). Mass transfer can account for some variations in the volatile yields of pyrolysis due to possible secondary reactions. However, it does not normally control the rate of pyrolysis of small or porous particles. (Solomon et al. 1992) The importance of heat transfer is supported by Janse et al. (2000). Their work on particles in the size range of 200 – 1000 µm shows that an extensive description of internal mass transport phenomena in flash-pyrolysis modeling is not necessary, while accurate knowledge of the reaction kinetics and heat transfer parameters is crucial.

Heat transfer during pyrolysis can be described by the following one–dimensional energy balance equation given for a volume element in a pyrolysing spherical fuel particle (Raiko, 1986).

$$\rho_p(T)c_p(T)\frac{\partial T}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(\lambda_p(T)r^2\frac{\partial T}{\partial r}\right) + \Delta h_{vol}\frac{\partial \rho_{vol}}{\partial t} - c_{vol}(T)\dot{m}''_{vol}\frac{\partial T}{\partial r} \qquad (1)$$

The first term in the equation depicts the heat conservation, the second the heat conduction, the third the heat source and the last term describes the heat convection caused by the released volatiles. The heat conduction inside the fuel particle is assumed to obey the Fourier-conduction equation.

Di Blasi (2008) reports in her extensive review on wood and biomass pyrolysis modeling that among the physical properties, density and thermal conductivity cause the highest sensitivity of the model predictions. She continues by stating that, in general, the limits of detailed models lie in the use of questionable kinetics or arbitrary values of physical properties, and, in numerous cases, in scarce experimental validation. In other words, only a better knowledge about decomposition kinetics and the physical properties of biomass fuels can improve the predictive capabilities of transport models. Hence, extensive model validation in relation to both product yields and conversion times is required. (Di Blasi, 2008)

A comprehensive review is given by Grønli (1996) of the correlations and values of the thermal conductivities, gas permeability, specific heats and other properties of several wood species and char. This present paper focuses on clarifying wood and wood char thermal conductivity and specific heat capacity. A literature review presents the most interesting findings of Grønli and clarifies research done on the properties since Grønli's thesis. Also the results of thermal conductivity experiments conducted at Tampere University of Technology are reported. Finally a draft correlation is proposed in order to evaluate the thermal conductivity of a pyrolysing wood particle.

WOOD AND WOOD CHAR PROPERTIES

As wood is pyrolysed its physical, chemical and structural properties change. The physical properties are thermal conductivity and specific heat, and structural properties particle size, density and porosity. (Grønli, 1996) The correlations presented in this paper are for dry wood and wood moisture is ignored. However, one needs to bear in mind that moisture has a significant effect both on thermal conductivity and specific heat.

Thermal conductivity

Wood is an anisotropic material, whose thermal conductivity is a function of heating direction, temperature, density and moisture. Based on his literature review, Grønli (1996) reports values for effective ***thermal conductivities for wood*** at room temperature, parallel with the grain, between 0.158 and 0.419 W/mK, and perpendicular to the grain, 0.081 – 0.209 W/mK. Thermal conductivity in the grain direction is 1.5 – 2.7 times the conductivity perpendicular to the grain direction. He concluded that effective thermal conductivity for wood at room temperature is 0.25 W/mK, although his own pyrolysis model converged best with experiments using a value of 0.35 W/mK at a temperature level of 293 – 523 K. Simpson and TenWolde (1999) report values from 0.09 to 0.14 W/mK for varying ovendry pine species.

Suleiman et al. (1999) measured thermal conductivities for Swedish birch of 0.291 – 0.323 W/mK in parallel, and 0.177 – 0.214 W/mK perpendicular to the grain direction at room temperature. They observed an increase of 14 % in thermal conductivity along the grain and an increase of 24 % perpendicular to the grain, on average, as the temperature increased from room temperature to 373 K.

Based on experiments on 10 Mexican softwoods, Leon et al. (2000) derived thermal conductivities of 0.156 – 0.278 W/mK for grain direction, 0.112 – 0.176 W/mK for radial direction and 0.074 – 0.133 W/mK for tangential direction at room temperature. They concluded that thermal conductivity along the grains is 2.16 times greater than the conductivity in a tangential direction and 1.36 times greater than in a radial direction. Gupta et al. (2003) conducted thermal conductivity tests for a North American softwood with a Fitch type device. They measured 0.0986 W/mK perpendicular to the softwood grains, and 0.2050 W/mK for the softwood bark. They conclude that the thermal conductivities of the wood and the bark increase linearly by 13 % as the temperature increases from 310 to 341 K.

Harada et al. (1998) developed the following correlation for wood thermal conductivity based on their flash method experiments at a temperature range of 293 – 513 K.

$$\lambda_w = 0.00249 + 0.000145\rho_w + 0.000184(T - 273) \qquad \text{(W/mK)} \qquad (2)$$

where λ_w is wood thermal conductivity, ρ_w density and T temperature. The correlation indicates that thermal conductivity is linearly proportional to temperature and it increases 50 % at a temperature range of 293 – 513 K.

The following correlation by Koufopanos et al. (1989) is used, for example, by Babu and Chaurasia (2003; 2004a; 2004b) in their pyrolysis models.

$$\lambda_w = 0.13 + 0.0003(T - 273) \qquad \text{(W/mK)} \qquad (3)$$

Although thermal conductivity is a function of temperature, constant values are still widely used in pyrolysis models, see, for example, (Kersten et al., 2005), (Wang et al., 2005), (Saastamoinen, 2006), (Younsi, 2006) and (Sand et al., 2008). Also more complex models have been developed, for example, by Saastamoinen and Richards (1996).

Determining the ***thermal conductivity of wood char*** is challenging due to its dependency on feedstock and pyrolysis condition. However, wood keeps its cellular structure relatively well in pyrolysis if moderate temperatures are used. This enables the determining of conductivity as a function of density and grain direction as for wood. (Grønli, 1996; Larfeldt et al., 2000)

Maple char thermal conductivity has been reported to be 0.1045 W/mK in parallel to the grains and 0.071 W/mK perpendicular to the grains at a temperature and density of 363 K and 200 kg/m^3, respectively (Grønli, 1996). According to the experiments by Gupta at al. (2003), the thermal conductivity of char derived from North American softwood bark increases linearly by 22 % from 0.0946 W/mK to 0.1156 W/mK as the temperature increases from 310 K to 341 K. Brown (1972) measured that wood char thermal conductivity at 823 – 873 K was three times greater than that at room temperature, and it was nearly twice as high as that of virgin wood (see Grønli, 1996).

More studies have been conducted on coal char thermal conductivity than on wood char thermal conductivity. However, comparing these studies is difficult due to the influences of feedstock and pyrolysis conditions, which are usually not described in detail. Kantorovich and Bar–Ziv (1999) conclude in their review on heat transfer within highly porous chars that, for the majority of the carbonaceous materials, the radiation contribution can be neglected at temperatures lower than 1000 K. Larfeldt at al. (2000) postulate that radiative heat transfer has insignificant effect when the pore size is smaller than 50 μm.

Specific Heat Capacity

The specific heat of wood changes as volatiles leave the particle and the particle temperature increases. The ***specific heat capacity of wood*** depends on the temperature and moisture content of the wood, but is practically independent of density and species (Simpson and TenWolde, 1999). Also the specific heat of water has a great influence on particle specific heat. According to a literature survey by Grønli (1996), specific heat capacities used for unreacted wood in pyrolysis models vary from 1.50 to 2.51 kJ/kgK, whereas Gupta et al. (2003) report that values from 0.67 to 2.5 kJ/kgK have been used. Kärkkäinen (2007) states that the specific heat capacity for an absolutely dry wood is 1.34 kJ/kgK at temperature range of 273 – 373 K.

Simpson and TenWolde (1999) report that the heat capacity of dry wood is approximately related to temperature by Equation 4. The temperature range for the correlation is not mentioned.

$$c_{p,w} = 0.1031 + 0.003867T \quad \text{(kJ/kgK)} \quad (4)$$

Harada et al. (1998) present a correlation for the wood specific heat capacity based on their experiments at 393 – 513 K, as follows.

$$c_{p,w} = 1{,}2 \ + 2{,}45 * 10^{-3}(T - 273) \quad \text{(kJ/kgK)} \quad (5)$$

Gupta et al. (2003) conclude, based on their experiments on the North American softwood, that in the temperature range of 313 – 413 K, specific heat capacity can be presented using the following linear equation.

$$c_{p,w} = 231{,}6 * 10^{-3} + 3{,}69 * 10^{-3} T \quad \text{(kJ/kgK)} \quad (6)$$

Gupta et al. report that they preheated samples at 423 K before they conducted specific heat capacity experiments to guarantee that the samples were dry. Grønli (1996) developed a correlation for a Norwegian spruce specific heat capacity at a temperature range of 350 – 500 K, based on his DSC experiments.

$$c_{p,w} = 1{,}5 + \frac{T}{1000} \quad \text{(kJ/kgK)} \quad (7)$$

Grønli (1996) also reports that Koch (1969) has measured pine specific heat capacities and that he wrote the following correction, which is valid at 298 – 413 K.

$$c_{p,w} = -9{,}12 * 10^{-2} + 4{,}4 * 10^{-3} T \quad \text{(kJ/kgK)} \quad (8)$$

All the correlations found in the literature and reported above present specific heat capacity as a linear function of temperature. These correlations are plotted in Figure 1.

Their values vary between 1.2 and 2.0 kJ/kgK at a temperature range of 293 – 513 K and differences between the correlations decrease as the temperature increases.

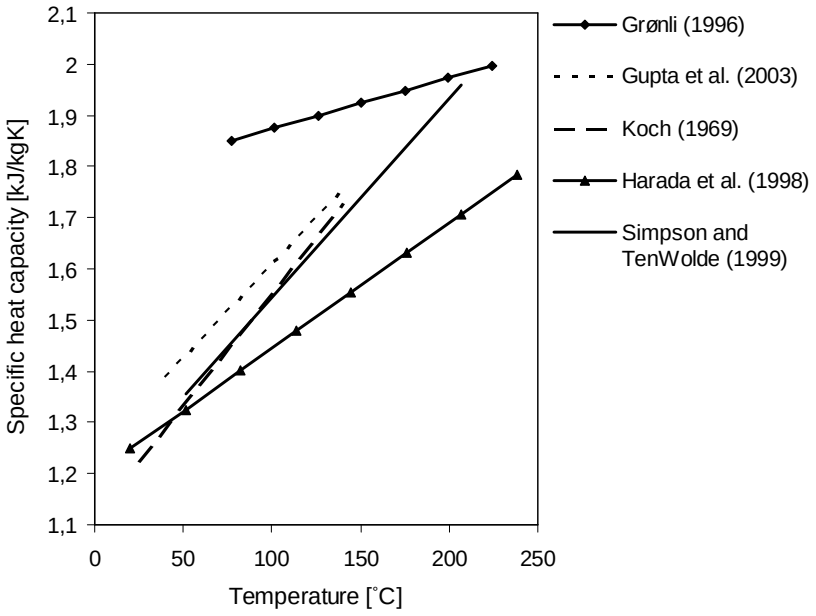

Figure 1. Specific heat capacities for wood reported in the literature.

The reported values for the ***specific heat of wood char*** at room temperature range from 0.67 to 1.35 kJ/kgK, according to the review by Grønli (1996). Grønli's correlation is based on specific heat capacity measurements conducted by Raznjevic (1976) at individual temperatures from 273 to 1273 K.

$$c_{p,c} = 0{,}42 + 2{,}09 * 10^{-3} T + 6{,}85 * 10^{-3} T^2 \qquad \text{(kJ/kgK)} \qquad (9)$$

A correlation for the specific heat of wood char by Fredlund (1998) (see Larfeldt et al., 2000) is

$$c_{p,c} = 1{,}43 + 0{,}355 * 10^{-3} T - \frac{7{,}32 * 10^4}{T^2}. \qquad \text{(kJ/kgK)} \qquad (10)$$

Koufopanos et al. (1989) have presented a linear correlation for biomass char specific heat capacity (see Babu & Chaurasia, 2003), as follows.

$$c_{p,c} = 1{,}0032 + 2{,}09 * 10^{-3} (T - 273) \qquad \text{(kJ/kgK)} \qquad (11)$$

Gupta et al. (2003) formed a correlation for specific heat capacity of the bark-derived char based on their DSC experiments on North American softwoods, as follows.

$$c_{p,c} = -795{,}28 * 10^{-3} + 5{,}98 * 10^{-3} T - 3{,}8 * 10^{-6} T^2 \qquad \text{(kJ/kgK)} \qquad (12)$$

The correlations found in the literature for the specific heat capacities of wood char are plotted in Figure 2. The correlations give relatively similar values, excluding the linear correlation by Koufopanos et al. (1989). Hence, it can be concluded that char specific heat capacity is relatively well known.

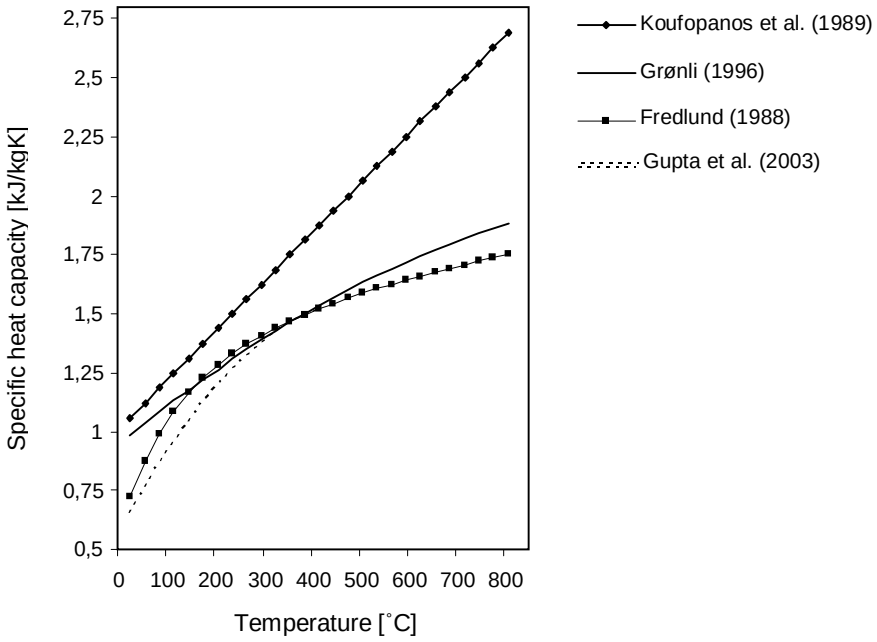

Figure 2. Specific heat capacities for wood char.

By comparing Figures 1 and 2, one can observe that the specific heat capacity of a pyrolysing wood particle is approximately 1.25 kJ/kgK at 293 K, increasing approximately to 1.9 kJ/kgK as the temperature increases to 523 K. Wood converts to char at temperature range of 523 – 673 K and the specific heat capacity of the pyrolysing particle decreases to 1.5 kJ/kgK by 673 K. As the temperature increases further from 673 K the specific heat capacity increases slightly to 1.75 kJ/kgK at 1083 K.

MEASUREMENTS ON THERMAL CONDUCTIVITY

The thermal conductivities were determined in Tampere University of Technology (TUT) with a simple modified Fitch-type apparatus. The apparatus is illustrated in Figure 3 below. The experiments were conducted by Mr. Tuukka Ahonen and the results are reported more extensively in his Master of Science Thesis (Ahonen, 2008). To the authors' knowledge, at least also Gupta et al. (2003) have measured wood, bark and bark derived char thermal conductivities applying a similar type of device.

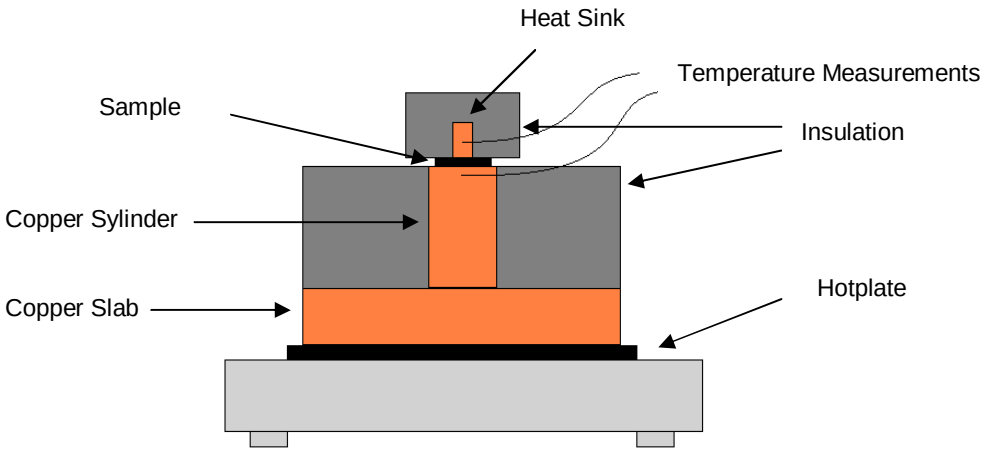

Figure 3. Fitch type device used for thermal conductivity measurements at Tampere University of Technology.

The device consists of a heat source, a copper slab (40 mm thick), a copper cylinder (l = 50 mm, d = 25 mm), a heat sink, and insulation. The heat source is a temperature-adjustable hotplate. A thermocouple wire is placed in a hole inside the copper cylinder 4 mm below its upper surface. It is supposed that this temperature is similar to the surface temperature. The heat sink is a cylindrical copper rod (l = 15 mm, d = 12 mm). Its temperature is measured through a hole that is drilled in the middle of the rod. The copper cylinder and the heat sink are insulated with Kaowool insulation material.

When a sample is placed between the copper cylinder and the heat sink heat transfer through the sample can be described with the following equation (Gupta et al., 2003).

$$\frac{\partial T}{\partial t} = \alpha * \frac{\partial^2 T}{\partial x^2} \qquad \text{where } \alpha = \lambda_p \Big/ (c_{p,cu} * \rho_{cu}) \qquad (13)$$

λ_p is the thermal conductivity of the sample, and $c_{p,cu}$ and ρ_{cu} are the specific heat capacity (315 J/kgK) and density of copper, respectively. If assumptions of a quasi-stationary state and of the heat transferred through the sample being totally absorbed by the heat sink are made, the equation above can be described as follows (initial condition $T = T_o$ at $t = 0$).

$$\ln\left(\frac{T_0 - T_\infty}{T - T_\infty}\right) = \frac{A_m * \lambda_p * t}{L * m_{cu} * c_{p,cu}} \quad (14)$$

Here, T is the time dependent temperature of the heat sink, T_∞ is the constant temperature of the heat source, A_m the surface area of the heat sink that is in contact with the sample (1.131 cm^2), L the sample thickness, and m_{cu} the mass of the heat sink (15.067 g). Other terms apart from the thermal conductivity λ_p are known or can be measured. Hence the logarithmic temperature difference can be plotted as a function of time, resulting in a linear curve with slope K. Hence thermal conductivity can be solved from Equation 14, as follows.

$$\lambda_p = \frac{L * m_{cu} * c_{p,cu} * K}{A_m} \quad (15)$$

Thermal conductivity is measured in the mean temperature of the heat source and the heat sink. Due to the increasing temperature of the heat sink during an experiment the average value of the sink is used to represent its temperature. Hence, ranging the temperature of the heat source, the thermal conductivity can be determined at different temperatures, varying from 309 to 370 K.

Results of the thermal conductivity measurements

Thermal conductivity measurements were taken for Finnish pine and birch, and for chars derived from these wood species. The chars were charred in an oven with a low heating rate. The char samples were 3 – 6 mm thick and the wood samples were 4 – 15 mm thick; however, thickness of the samples did not have an effect on the obtained thermal conductivity results. Each sample used in the measurements was at least 20 mm wide.

The temperature range of the heat source used for the measurements was 323 – 433 K. The temperature varied less than 1 K during each experiment. The heat sink was at room temperature in the beginning of each experiment. The measurements were well reproducible, with the greatest variation of measured thermal conductivity values of ±5 % observed for wood samples when thermal conductivity was measured along the wood grains at the highest temperature. The variation was approximately ±2 % for char thermal conductivities. Calibration of the device could not be conducted because appropriate material that would have a similar steady thermal conductivity as the measured samples could not been found. Hence, the values obtained from the measurements should be considered with care.

The dependence of pine wood thermal conductivity on grain direction was measured in 3 directions. Figure 4 illustrates the thermal conductivities in tangential and in radial directions being almost equal. The thermal conductivity parallel to the grain is more than 2 times greater than the conductivities in tangential and in radial directions. This agrees with the literature, although the absolute value of the conductivity in the grain

direction is higher than that reported in the literature. Nevertheless Grønli's (1996) pyrolysis model agreed best with the measurements when he used a constant value of 0.35 W/mK for pine thermal conductivity.

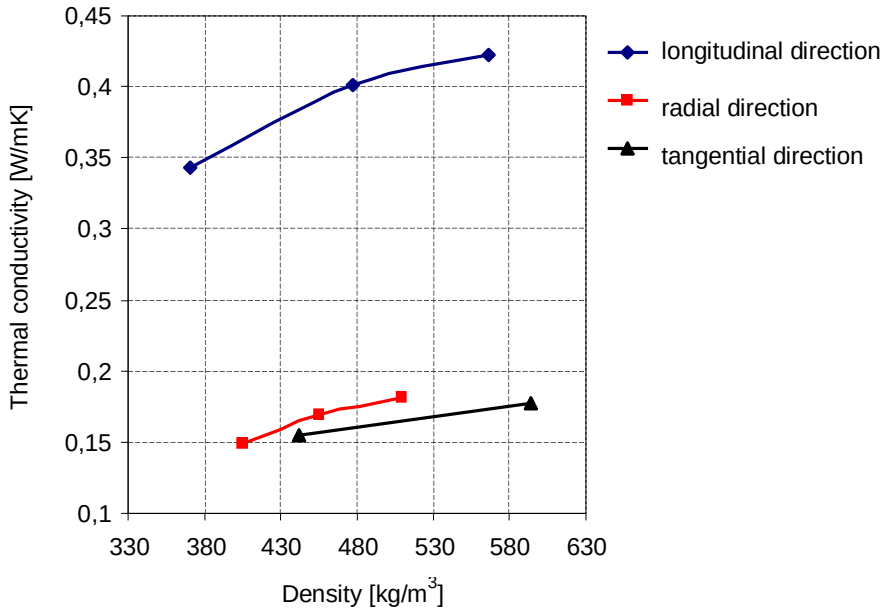

Figure 4. Pine wood thermal conductivity as measured in longitudinal, radial and tangential directions at a temperature of 309 K.

Figure 5 illustrates the measured thermal conductivities for pine char and birch char. The measured char thermal conductivities agree with previous measurements reported in the literature. Grønli (1996) and Gupta et al. (2003) measured values of 0.071 – 0.1159 W/mK for softwood char at temperature range of 310 – 363 K and density range of 200 – 299 kg/m_n^3. According to the measurements also char thermal conductivity is a function of grain direction, although the dependence is not as great as that with wood.

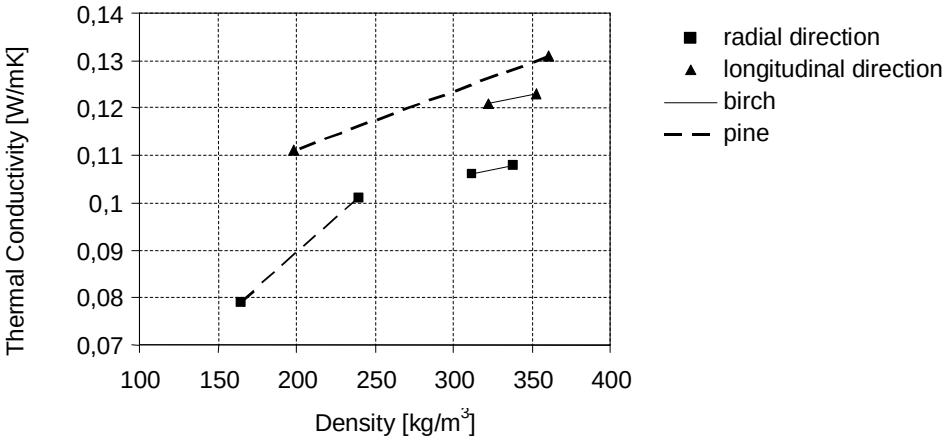

Figure 5. Measured pine char and birch char thermal conductivities as a function of density at a temperature of 336 K.

Pine wood thermal conductivity was measured as a function of temperature in a radial direction. The density of the studied pine sample was 455 kg/m^3. The conductivity increased 5 % from 0.168 W/mK at 309 K to 0.177 W/mK at 370 K. Gupta et al. (2003) observed an increase of 13 % in the conductivity at temperature range of 310 – 341 K, whereas Suleiman et al. (1999) measured an increase of 22 % in the conductivity perpendicular to the grains at temperatures of 293 – 373 K.

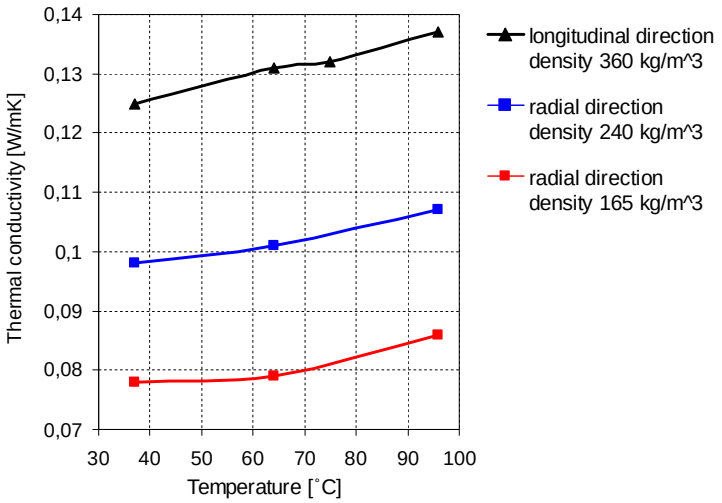

Figure 6. Measured pine char thermal conductivity as a function of temperature.

The measured pine char thermal conductivity dependence on temperature is illustrated in Figure 6. The thermal conductivities of these samples increased almost linearly by 9 – 10 % at the temperature range of 309 – 370 K. Gupta et al. (2003) measured an increase of 22 % at a temperature range of 310 – 341 K for the bark-derived char.

Thermal conductivity of a pyrolysing wood particle

The thermal conductivity during pyrolysis is reported to change due to the density change of wood to char. In addition, the decrease in conductivity is a consequence of the lower thermal conductivity of the char material compared to the conductivity of the wood material. This is illustrated in Figure 7, where thermal conductivity is presented as a function of density. The figure shows that samples with similar densities have different thermal conductivities, depending on their conversion levels. This indicates that the thermal conductivity of wood char material is lower than that of wood material. Hence, the thermal conductivity of a pyrolysing wood particle can not be presented as a function of density only, but also the conversion level has to be taken into account.

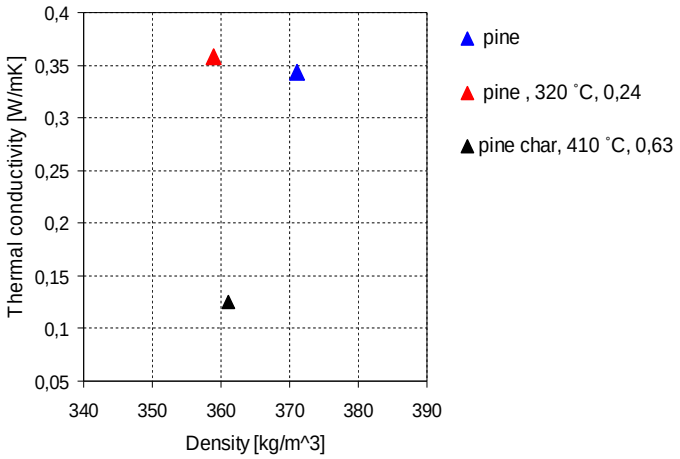

Figure 7. Thermal conductivity of wood and wood char of similar densities at different conversion degrees. Degrees centigrade indicates particle pyrolysis temperatures followed by conversion levels (0.24 and 0.63).

Figure 8 presents the thermal conductivity for a pyrolysing pine wood as a function of density at different conversion levels. The conductivity decreases most at a temperature range of 613 – 653 K. The mass loss is also greatest at that temperature range. After 653 K wood is completely pyrolysed and the conductivity is affected only by density and not by conversion level anymore.

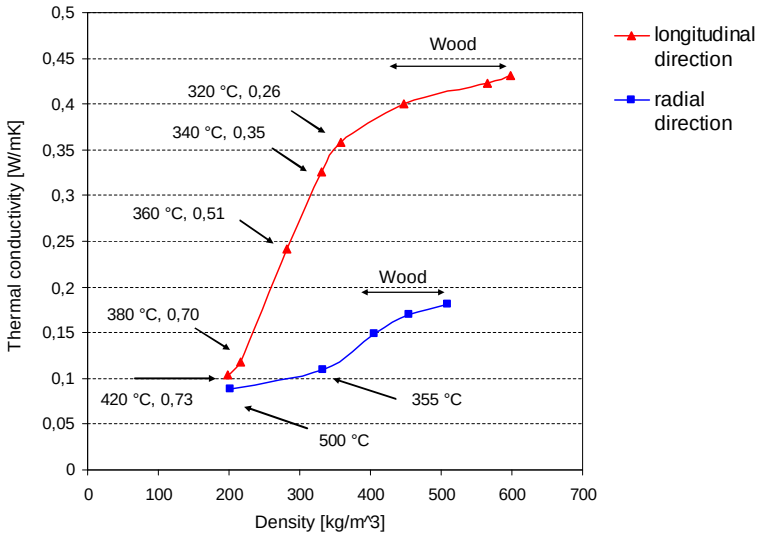

Figure 8. Thermal conductivity at different conversion levels. The conductivities are measured at 309 K.

Figure 9 presents a draft of the thermal conductivity of a pyrolysing pine-wood particle based on the measurements conducted in TUT. The blue curve is an estimation of the conductivity if the temperature-dependence of wood char thermal conductivity is ignored. The black curve in Figure 9 presents the thermal conductivity of the pyrolysing wood when temperature dependence is taken into account. The curve is drawn according to Equations (16)-(18).

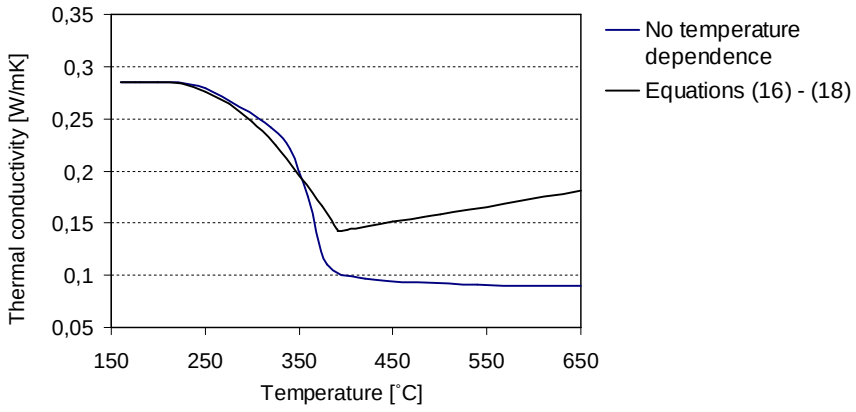

Figure 9. A draft of the thermal conductivity of a pyrolysing wood particle as a function of temperature.

$$\lambda_w = 0.285 \quad \text{(W/mK)} \quad T \le 473K \quad (16)$$

$$\lambda_{w-c} = -0.617 + 0.0038T - 4 * 10^{-6} T^2 \quad \text{(W/mK)} \quad 473K \le T \le 663K \quad (17)$$

$$\lambda_c = 4.429 * 10^{-2} + 1.477 * 10^{-4} T \quad \text{(W/mK)} \quad 663K \le T \le 923K \quad (18)$$

The thermal conductivity of fresh wood is an average of parallel and perpendicular grain directions with density of 450 kg/m_n^3. Temperature dependence is ignored due to density variation of wood. The wood pyrolyses between 473 – 663 K and the thermal conductivity decreases to that of char. Char thermal conductivity is assumed to follow linear dependence on temperature also at higher temperatures, as it was measured at the temperature range of 309 – 370 K. Char thermal conductivity is assumed to be 0.09 W/mK at 309 K, increasing approximately to 0.145 W/mK at 673 K and to 0.175 W/mK at 923 K.

The effect of radiation is not taken into account in Figure 9 due to the limited temperature range of the Fitch-type device. Also the correlations for radiative thermal conductivity give arbitrary values. The effect of a radiation correlation by Chan (1985) on thermal conductivity is 0.04 W/mK at 673 K, increasing to 0.11 W/mK by 923 K. Di Blasi's (1996) radiation correlation affects the thermal conductivity by 0.03 W/mK at 673 K and by 0.07 W/mK at 923 K. A correlation for radiative heat transfer inside a fuel particle by Atkinson and Merrick (1983) affects the least, only by 0.02 W/mK at 923 K. Other radiation correlations are listed in Ahonen (2008).

It is questionable how accurately the measurements that were conducted at low temperature can predict char behavior at elevated temperatures. Also a large amount of assumptions are made. Hence, one can easily criticize the results and assumptions made. However, the authors are convinced that the thermal conductivity that is presented in Figure 9 is closer to the real values of a pyrolysing wood particle than if constant values were used.

CONCLUSIONS

Wood pyrolysis is controlled by chemical kinetics, heat transfer or mass transfer. Excluding very small particles, the process is usually controlled by the inner heat transfer of wood. In addition to thermal conductivity and the specific heat of wood and its char, the heat transfer is affected by specific heats of volatiles and reaction enthalpy of the pyrolysis reactions.

The literature survey clarifies that the specific heat capacities of wood and wood char are known fairly accurately and the variation between values presented in the literature is modest. Studies on wood thermal conductivity suggest differing values for the conductivity and only a few correlations for wood thermal conductivity could be found in the literature. Only a modest number of studies on wood char thermal conductivity were found. However, these studies propose significantly similar values for wood char thermal conductivity at low temperatures. Data on wood char thermal conductivity at high temperatures hardly exists in the literature.

The experiments were conducted on pine and pine char thermal conductivities using a Fitch-type device. The results show that the thermal conductivity of pine char is significantly less than that of pine wood, and the thermal conductivity of a pyrolysing particle changes due to density and material property changes. The values measured for the thermal conductivities close to room temperature are slightly higher than those found in the literature. However, the temperature-dependency obtained from the measurements is smaller than other reported measurements indicate. The experiments also showed that wood anisotropy has an effect on thermal conductivity. However, it is sufficient to examine the conductivity in two directions, namely parallel to the grain direction and perpendicular to the grain direction.

A draft correlation for the thermal conductivity of a pyrolysing wood particle is presented based on the measurements conducted in TUT. The correlation will be used in a model for a pyrolysing particle that will be written utilizing the results reported in this paper and Ahonen (2008). The model will be validated with drop-tube pyrolysis experiments that have been planned for this year, 2009.

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