Effect of wood chips properties on mineralogical composition and microstructure of silicate matrix based composites

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Abstract. The research presented in this article was focused on proving the effect of wood chips parameters on the hardening process of silicate matrix in cement-bonded particleboard. Namely content of humidity in the chips (in range 30 to 90%) was significant. This amount of humidity was chosen with respect to real conditions of cement-bonded particleboards production. Influence of humidity could be reflected in the content of sugar (cellulose etc.) in wooden chips. The sugars affect the hardening of matrix negatively. Thus, phase composition and microstructure of matrix of cement-bonded particle boards were analysed during 2 to 28 days. Investigation of physico-chemical parameters and microstructure was also supplemented by verification of the basic material characteristics of cement-bonded particleboards (strength and modulus of elasticity in bending, transverse tensile strength perpendicular to the plane of the board, etc.). Slightly negative effect of increasing chip humidity content on the mineralogical composition and microstructure including physical parameters was shown in scope of the experiments. Therefore attention during cement-bonded particleboards production has to be paid even to moisture of the chips.

1. Introduction
The production process of cement-bonded particleboards consists of several phases. First, considering the given humidity, the basic raw materials are weighed – chips, cement, water, additives. The additives are mainly used for closing of board surface part structures. Another reason is to prevent release of sugars which are contained in the chips. During the boards production a mixture with humidity up to 45% is put in layers on steel plates by using machine equipment. The layering is done in three steps – first the bottom visual layer, then core and finally the visual one again. The visual layer contains more cement bonding material and smaller chips resulting in more compact structure. Thickness of visual layer moves within several mm. Metal plates with mixture are stacked one over another. Then the stack is pressed in a hydraulic press. By pressing the original height is reduced to 1/3. The stack in this pressed condition is placed in a steaming chamber for several hours. The temperature here does not exceed 55°C. Further approximate 7 days the boards mature freely in the production plant with subsequent drying in tunnel drying plant where the temperature does not exceed 90°C. In the last production phase the boards are processed to required formats, surface quality with eventual application of surface treatment. The mixture for production of cement-bonded particleboards is characterized with a certain optimum humidity as it was stated above. The chip humidity is closely related to this. Considering the wood processing, transport and storage condition (also the weather) the chips are delivered to the production plant with variable humidity. In order to preserve the production process continuity it is not possible to dry the chips to a constant weight or humidity respectively. The chips are usually delivered with absolute humidity content within 30 to
90%. In case of lower humidity content just adding of water will do. However with humidity content close to the upper limit, not only the amount of mixed water batch must be reduced but also the amount of water for dissolving of mineralization mixtures should be reduced. Theoretically the hydration process could be negatively influenced due to insufficient mineralization of chips. It can also be assumed that during pressing water out of the chips over its surface parts the already mineralized layer could be damaged - „drifted out“ of the mineralizing component from these surface parts. Presence of sugars in cement matrix changes reaction of tricalciumaluminate (C₃A), which is the fastest reacting component of cement. Extraction components from wood may be also absorbed on the core of calcium hydroxide, therefore they slow down hydration of tricalciumsilicate (C₃S) [3]. According to [4-5] inhibiting components (hemicellulose etc.) act so that an impermeable film is created around non-hydrated grains of cement and for this reason, formation of calciumhydrasilicates (CSH) and portlandite (CH) are subsequently slowed down. Also the pressing the water out of the chips could result in drifting out (“pushing away” respectively) of the cement from the chip surface. This effect could participate on imperfect coverage of chips with cement paste which is connected namely with radical decrease of strengths. Objective of the research presented in the article is proving the effect of wood chips parameters on the hardening process of silicate matrix in cement-bonded particleboard.

2. Methodology of testing
Three formulas were tested in total. Differences were only in water distribution in the given mixture. Particularly the chips with humidity 30%, 60% and 90% were used (formulas - mixtures – W30, W60 and W90). For particular formulas the water amount was set in a way its content was the same in the resulting mixture for all the formulas. Therefore the mixing water amount was gradually reduced in dependence on growing chip humidity. In case of the last formula containing chips with 90% humidity the mixing water was completely eliminated and in addition also a part of the water necessary for dissolution of mineralizing components. The prepared mixture was layered in forms and subsequently pressed with thrust approximately 25 N.mm⁻². The mixture pressed in this way was moved into the steaming chamber. The specimens were removed from this ambient and taken out of forms after approximately 10 hours. In respect of the production of boards the very phase of mixture pressing can be considered as problematic one. With the mixtures containing chips with higher humidity content a water displacement may occur. At water leak also a certain amount of mineralizing additives or cement eventually could be drifted out. With the humidity contained in chips compared to the mixture where the “free” mixing water is included this risk is much higher. The reason is the fact that the water travels from chips (therefore over their surface) which contains water solution including mineralizing components and it is wrapped with cement paste. This would then negatively result in the hydration process course and therefore on final properties of cement-bonded particleboards. The hydration process was monitored at age of 2, 3, 8, 14 and 28 days. The mineralogical composition of the matrix was analyzed – identification and quantification of typical hydration phases. XRD and DTA were used for evaluation of crystalline phases. Microstructure was examined using SEM. Namely the board matrix subject of interest during microstructure investigation. At age of 8, 14 and 28 days also basic physical and mechanical parameters – bulk density, strength and modulus of elasticity in bending, transversal tensile strength vertically to board plane were analyzed according to CSN EN 310, 319 and 323.

3. Results and discussion
3.1. Physical and mechanical characteristics of tested boards
The physical and mechanical parameters were monitored at age of 8 days (after second heating cycle), 14 days (expedition of the boards in real production conditions) and 28 days (normative requirements). As it is clear from test results all the formulas are balanced with respect of the monitored properties (see table 1). The boards made from chips with 30% humidity are evaluated as the best ones. Further it
is evident that with growing age the differences between particular formulas decrease. Also the comparison of strength parameters with modulus of elasticity is interesting. Here we can see differences in course of particular quantities. Bending strengths are characterized with growing tendency of dependence on growing input humidity of chips. On the contrary the highest tensile strengths vertically to board plane had the formula with chips of 60% humidity. Upon the course of physical and mechanical properties a slight influence of chip humidity can be assumed.

Table 1. Parameters of mixtures containing chips with different amount of humidity, age up to 28 days.

| Formula – age (days) | Bulk density (kg.m⁻³) | Bending strength (N.mm⁻²) | Modulus of elasticity in bending (N.mm⁻²) | Transverse tensile strength (N.mm⁻²) |
|---------------------|-----------------------|---------------------------|------------------------------------------|-------------------------------------|
| W30-08              | 1340                  | 11.2                      | 5326                                     | 0.54                                |
| W60-08              | 1360                  | 10.6                      | 5384                                     | 0.52                                |
| W90-08              | 1330                  | 9.9                       | 4992                                     | 0.51                                |
| W30-14              | 1310                  | 12.2                      | 5531                                     | 0.57                                |
| W60-14              | 1320                  | 12.0                      | 5491                                     | 0.55                                |
| W90-14              | 1310                  | 11.3                      | 5283                                     | 0.53                                |
| W30-28              | 1270                  | 13.1                      | 6359                                     | 0.63                                |
| W60-28              | 1260                  | 12.9                      | 6385                                     | 0.65                                |
| W90-28              | 1240                  | 12.4                      | 6302                                     | 0.59                                |

3.2. Mineralogical composition and microstructure

Base phase components were identified in the silicate matrix by XRD. The emphasis was namely placed on clinker minerals and hydration products. Basically the identical mineralogical components with only imperceptible differences were identified in all analyzed samples. However the courses of particular diffraction lines are far more interesting. Two facts can be partially assessed from these courses. In the first line the peak intensity of identified components give information on their amount in the analyzed material. Further the intensity value of particular minerals characterizes their morphology - i.e. how is the given material developed. In the figures 1-4 there are selected diffraction schemes. Complete development of diffraction lines is shown in figure 1 (chips saturated by 30% water) and figure 2 (chips saturated by 90% water). For illustration representative samples analyzed at age of 2 and 28 days were selected (see figure 3 and 4). It can be seen very well how portlandite and calcite develop what is accompanied by decrease of diffraction maximums of clinker materials. The differences are also notable between the formulas. In figure 3 a quite significant difference can be seen at $\theta = 18^\circ$, which is a peak characteristic for portlandite (decreasing peak intensity with growing input chip humidity and lower amount of mixing water).

Table 2. DTA results – content of CaO from portlandite and calcite, age from 2 up to 28 days.

| Age (days) | CaO – W30 (%) | CaO – W60 (%) | CaO – W90 (%) |
|------------|---------------|---------------|---------------|
| 2          | 9.31          | 8.54          | 8.59          |
| 3          | 14.64         | 13.85         | 12.71         |
| 8          | 15.94         | 16.18         | 14.79         |
| 14         | 17.76         | 17.12         | 16.85         |
| 28         | 19.21         | 18.63         | 18.05         |

For hydration grade quantification the content of CaO falling on Ca(OH)₂ (portlandite) and CaCO₃ (calcite) was selected. This value can be considered as relatively objective indicator of hydration reaction course. However the weight loss related to exothermic delay at temperature from 200°C to
500°C had to be excluded as first. Within this interval a thermal decomposition of mineralized chips occurred. Regarding objectivity the values were related only to the matrix during calculations. Results of setting of CaO content falling on portlandite and calcite are stated in the following table (see table 2). The resulting values of CaO content circa correspond to bending strength (see table 1).

Amount of CaO (falling on monitored components – Ca(OH)₂ and CaCO₃) clearly points to a slightly negative influence of chip humidity as filler for production of cement-bonded particleboards. Only with samples at age of 8 days the CaO content was increased with formula W60. Further it is clear that differences are stronger between the formulas W60 and W90. The influence of water amount reduction on account of necessary dose for mixing of mineralizing additives can be considered as well.

![Figure 1. XRD patterns of W30 from 2 (top brown curve) to 28 days (bottom black curve).](image1)

![Figure 2. XRD patterns of W90 from 2 (top brown curve) to 28 days (bottom black curve).](image2)

![Figure 3. XRD patterns of mixtures W30-02 (bottom curve), W60-02 (middle curve) and W90-02 (top curve) (P-portlandite, K-calcite, A-alite, B-belite, C-celite).](image3)

![Figure 4. XRD patterns of mixtures W30-28 (bottom curve), W60-28 (middle curve) and W90-28 (top curve) (P-portlandite, K-calcite, A-alite, B-belite, C-celite).](image4)
The figures 5-8 show the differences in microstructure of particular formulas at age of 2, 8, 14 and 28 days. Developed crystals are perceptible mainly from 8-day age. Figure 6 represent clear difference in progress of portlandite crystals development whereas significant change of this mineralogical
Component proportion is evident in case of W30. General, in the case of the mixture W30 (i.e. 30% water saturated chips) it can be stated that portlandite crystals were identified slightly often within microstructure. Finding of clinker minerals was very difficult. Developed CSH phases in the gel form could be identified in all mixtures, however in boards W30 could be evaluated slightly better.

4. Conclusions
Moderately negative influence of chip humidity on hydration course and final properties of cement-bonded particleboards was proved by the performed experiments and their detailed assessment. This fact was confirmed either by physical, mechanical tests and physico-chemical analyses as well. The differences are more noticeable in case of application of chips with 90% humidity (W90), when the mixing water had to be completely eliminated and also partially the water necessary for mineralization of chips. It is probable that mineralization layer or coating of chips was upset and thus sugars contained in chips probably released (extraction of sugars from wood by water washing is one of the possible ways). This resulted in slowing down of the hydration process. This fact can be seen well with particular diffraction lines and further with quantified CaO amount falling on portlandite and calcite. In spite of the above mentioned they are only minor scale changes. In addition it can be stated that requirements in regard of board properties are met (the strengths and modulus of elasticity meet the minimum standard criteria) even with the formula W90. Considering also the microstructural analysis outputs it can be stated that probably no significant drifting out of cement paste wrapping chips occurred. With respect of the found knowledge it is necessary to monitor the water content in chips with aim to use chips with the lowest possible humidity. In case of chips with higher humidity it is necessary to suitably regulate the mixing water amount or amount of the water necessary for dissolving of mineralizing additives. The reason is to preserve the constant humidity of the fresh mixture of the cement-bonded particleboards and therefore the provision of required parameters (strengths, modulus of elasticity, bulk density etc.).

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