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EXAMINATION OF THE DEGREE OF SHRINKAGE AND SWELLING OF THE DOUGLAS FIR CELL WALL

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ABSTRACT

Upon sorption of water, wood swells, and this swelling can be directly measured at the macroscopic level of the scale. The reverse process is called shrinkage. The two processes are reciprocal and can be studied at different levels of scale. More difficult to observe and measure is the sub-microscopic level, i.e. how the cell wall swells and shrinks. There is a difference in the structure of the radial and tangential cell walls due to the different dynamics of the cambium division.

The article studies the swelling and shrinkage of radial and tangential cell walls. An attempt has been made to model these processes geometrically at the cell level. Douglas fir early and late wood zones were studied.

Key words: wood, shrinkage, swelling, cell wall, Douglas fir.

INTRODUCTION

Swelling and shrinkage are processes that have been known and studied for a long time. These processes, as well as their difference in radial and tangential direction, can be explained at different scale levels (Peck 1957). It is known that the main factors influencing the shrinkage of wood are the angle of the microfibrils, the thickness of the cell wall and the walliness of the fibres. In the search for dependencies between these factors, eucalyptus wood was studied, with emphasis placed on the influence of its structure on the studied processes. The proportion of ray parenchyma was found to have a large effect on residual collapse (Wu Yi-Qiang *et al.* 2006). Within the hygroscopic range, water sorption occurs only at the cell wall level, where polar water molecules are hydrogen bonded to the hydroxyl sites of the polar molecules of the wood polymers. Except for the crystalline zones of cellulose, all wood polymers demonstrate, to varying degrees, an affinity for water (Rafsanjani *et al.* 2015).

Cell behaviour at the microscopic level is strongly influenced by cell arrangement, particularly by the alternation of earlywood and latewood (Murata and Masuda 2006). The general shrinkage and swelling of the wood results from the combination of these processes in the early and late wood taken together. Separated, however, they exhibit different behaviour. When observing one tree species (Pinus radiata), in some samples, a stronger shrinkage of the earlywood was observed, while in others, it was of the latewood. In the former, micro-cracks appear in the earlywood.

For rings with cracks, the tangential shrinkage of earlywood is higher than that of latewood, and for rings without cracks, the trend is reversed (Pang *et al.* 1999). When the relative humidity

of the surrounding air changes rapidly, the latewood deforms only in the radial direction. In addition, with rapid absorption of moisture, the diameter of the lumen decreases, while with slow absorption, it expands. When the specimen slowly absorbs moisture while it is in equilibrium with the water vapour pressure of the saturated solutions, the latewood swells in both radial and tangential directions, showing that a linear relationship can be established with increasing water content. Then, the radial expansion becomes slightly uneven. In a sample that has rapidly absorbed moisture from hot steam, the diameter of the tracheid lumen shrinks. However, it expands unevenly when the sample absorbs moisture slowly (Murata and Masuda 2006). In the measurements made on spruce wood, the high shrinkage anisotropy of the early and the low anisotropy of the latewood should be mentioned among the main results. Thus, it can be concluded that shrinkage at the cellular level results from two related effects: 1) properties of the cell wall (especially its local anisotropy) and 2) cell shape as a result of its formation from the cambium (Almeida *et al.*, 2014). Swelling and shrinkage anisotropy at the cell wall level cannot be explained solely by the structure of cellulose microfibrils, as their confinement effect is not strong enough to produce large transverse anisotropy (Rafsanjani *et al.*, 2014). It is also well known that the transverse shrinkage anisotropy of earlywood is more pronounced than that of latewood (Bonarski *et al.*, 2015). Experimental studies have shown that softwood shows different swelling patterns at the cellular scale depending on the position of the tracheid cells in the growth ring. Observations of thin-walled earlywood cells show anisotropic swelling behaviour, whereas swelling of thick-walled latewood cells is generally isotropic (Rafsanjani *et al.* 2015). Shrinkage and swelling at the cell level are realized differently in different cell wall layers. From the difference between the area of the tissues and the total dimensions of the cells, the value of shrinkage and swelling of the middle lamella can be determined. The variation of shrinkage or swelling at different hierarchical levels (tissue, cell, and cell wall) indicates that the hygroscopic middle lamella plays a role in tissue-level deformation (Fig. 1). It is likely that the complex system of wood can regulate hygroscopic deformations by fine-tuning its hierarchical structures (Zhan *et al.* 2021).

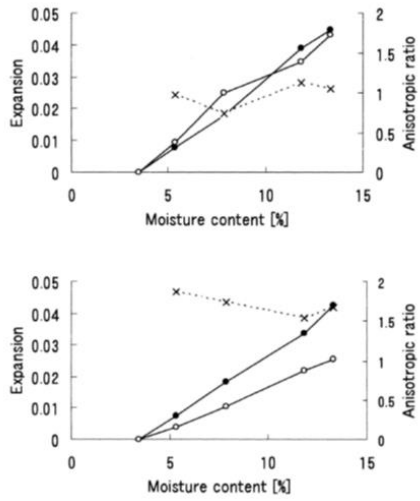

Figure 1: Tangential and radial swelling and swelling anisotropy. Above, for latewood only. Below are specimens from an entire annual ring. Filled circles indicate tangential swelling and empty circles indicate radial swelling. Crosses show an anisotropic relationship (2006 Murata K., M. Masuda).

Many attempts have been made to model these processes (Yamamoto 1999, Nakano, 2008, Schulgasser and Witztum, 2011). Mathematical models of the anisotropy of wood shrinkage were created. The formula describing the dynamic behaviour of shrinkage and swelling of wood fibres was obtained. Some of the models attempt to explain how the cell wall swells and dries. Wood cells are thought to swell both outward and inward toward the cell cavity, with no fundamental difference between coniferous, broadleaf, and tropical tree species (Fig. 2). The cross-section of the cell wall is analyzed theoretically, and the characteristic swelling parameters are obtained using a quadratic model (of the cross-section). An indicator is introduced, which is the ratio of cell wall thickness to external swelling (Nakano, 2008).

At the macroscopic level, the fact that the shrinkage in the tangential direction is significantly greater than the shrinkage in the radial direction and that the ratio of these shrinkages is inversely proportional to the ratio of the elastic moduli is also explained, and this relationship is used. The crystalline regions are an order of magnitude more complex than the matrix material. They are largely unaffected by water loss, i.e. they do not shrink. However, the matrix material is highly affected by water loss. It dries up to a large extent (Schulgasser and Witztum, 2015). The complexity of wood in terms of composition and molecular and hierarchical structure has challenged scientists for a long time. A comprehensive understanding of tissues requires a description of the materials at multiple length scales and needs an underlying molecular model. In certain works, a combination of experimental techniques and a physicochemical model is proposed to describe the changes at the nanometric/molecular level accompanying wood swelling (Barbetta *et al.*, 2018). It was found that as wood density increases, shrinkage increases in both the tangential and radial directions (Wu Yi-Qiang *et al.* 2006). For hardwoods, the relationship between specific gravity and bulk shrinkage is not completely linear (Herbert 1971).

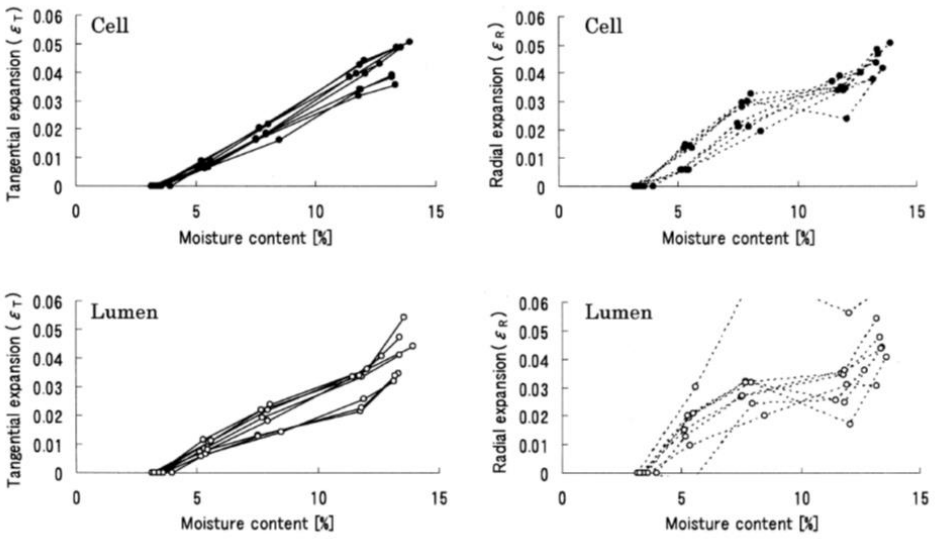

Figure 2: Relationships between water content and latewood tracheid swelling. The top (filled circles) shows the cell diameter. The lower part (with the empty circles) shows the diameter of the lumen. On the left (solid lines), the swelling is tangential, and on the right (dotted lines), it is radial (2006 Murata K., M. Masuda).

METHODOLOGY

CELL WALL STRUCTURE AND SWELLING

The secondary cell wall is located from the complex middle lamella towards the cell gap. It is composed of three layers – outer S1, middle S2 and inner S3. The secondary cell wall is about 10 times thicker than the primary cell wall. It is characterized by a spiral arrangement of the microfibrils at a different angle to the longitudinal axis of the cell. In the outer layer S1 of the secondary cell wall, which has a very small thickness (approximately equal to the thickness of the primary cell wall), two or more layers of lamellae with crossed microfibril spirals can be observed at an angle to the cell axis of 60° and about 45° respectively for the wood of coniferous and deciduous species (Fig. 3). In the widest middle layer S2 of the secondary cell wall, the microfibrils are arranged in a helix with an angle of inclination of 5–15° (rarely 30°) from the vertical. In the inner layer S3, the microfibrils are arranged along a flat spiral with an inclination angle of 50–80° in broad-leaved tree species and almost 90° in conifers (Wagenführ 1996).

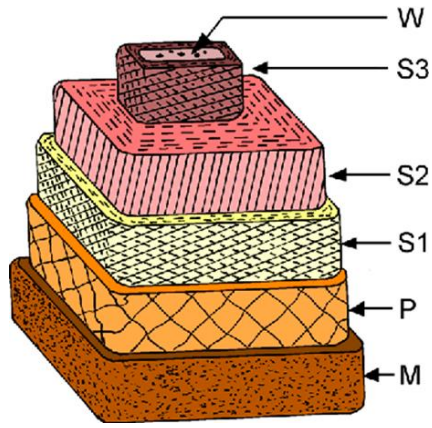

Figure 3: Cell wall layers of a wood cell (Bluskova 2009)

The first question to be asked in this experiment is: How many turns does the cellulose matrix make in the different layers of the secondary cell wall? In the S_1 layer, we have a 60° slope and tangled cellulose matrices, while in the S_2 layer, we have a 15° slope. The length of the side of this triangle with a height of 20 μm is sought. After simple geometric calculations, it can be seen that in layer S_1, the cellulosic matrices will make about ¼ of the turnover, and in layer S_2, about 1/3. This means that the microscopic slide to be measured will swell in a very different way than solid wood. It should not be forgotten that with a slide thickness of 20 μm, core rays will also not be present in the measured wood. The different shrinkage and swelling in the different growth directions are mainly explained by the structure of the cell walls (Bluskova 2009). When water is adsorbed, the middle layer (S_2) tends to swell proportionally to the number of microfibrils (i.e. proportional to its thickness), but the other two layers (S_1 and S_3) exert a retaining effect due to the different orientations of their microfibrils. The small longitudinal shrinkage is due to the orientation of the microfibrils in the S_2 layer. If these microfibrils were exactly parallel to the axis of the stem, longitudinal shrinkage would be zero. Its small value is due to the slight deviation from the axis of the cell. Large deviations (for example, in pressure

or juvenile wood) contribute to greater shrinkage and swelling. Longitudinal shrinkage of up to 10% has been measured in compressed wood.

DETERMINATION OF THE CROSS-SECTIONAL SHAPE OF TRACHEIDS

Similar to the classification of the shape of the cross-section of the tracheae (vessels) in broad-leaved wood, the plane of symmetry can be chosen as a leading factor here (Bardarov *et al.* 2019). According to the shape of their cross-section, tracheas are classified by being divided into:

- circle – all points of the cell wall are equidistant from the centre, and there are countless planes of symmetry;
- oval – all points of the cell wall are distributed along the radii of two circles, and there are two planes of symmetry;
- star – the points of the cell wall are distributed along a contour that does not form a single plane of symmetry.

In coniferous species, the most common cross-sectional shape of tracheids is a quadrilateral with rounded ends (Fig. 4). Here, the wounds of symmetry are also two, and this shape may be considered a particular case of the oval.

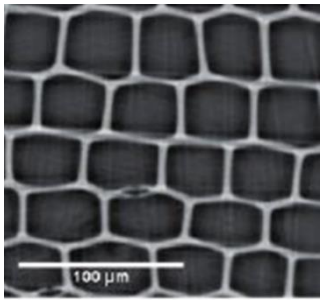

Figure 4: Cross-sectional shape of tracheids in conifers is defined as a "rounded quadrangle" (Derome et al., 2015)

The majority of cells shown in Fig. 5, however, are rather "rounded polygons" having one or even no planes of symmetry. A great variety of forms causes the so-called "reaction wood", which in conifers is called "compressive wood ". Here, the area of the washers is divided into compression, opposite (located on the other side of the stem axis) and normal wood (Enchev 1984). Normal wood has thin-walled tracheids with the characteristic shape described above. In compressed wood, the tracheids acquire a shape closer to a circle (Fig. 5).

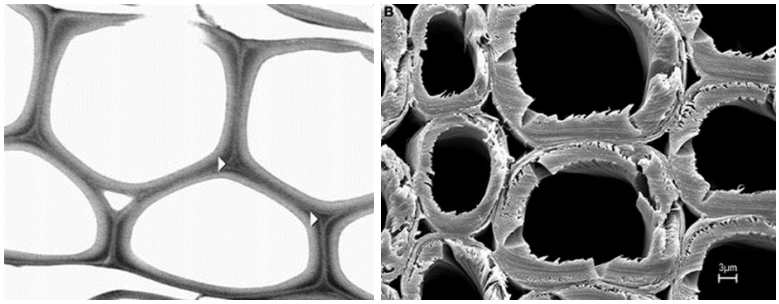

Figure 5: Cross-sectional shapes of normal (left) and compressed (right) wood. (Derome et al., 2015)

DEFINING THE CELL WALL ENVIRONMENT

To determine the dimensions of the tracheid, familiar designations of the cross section are used (Fig. 6):

- Lr – the size of the cell gap in the radial direction;
- Lt – the size of the cell gap in the tangential direction;
- 2Wr – the size of the cell wall in the radial direction;
- 2Wt – the size of the cell wall in the tangential direction.

Thus, the size of the cell in the radial direction will be L_r+2W_r, and in the tangential direction – Lt+2Wt. These dimensions may be called external to the cell. The internal dimensions of the cell are the dimensions of the cell gap – L_r and L_t.

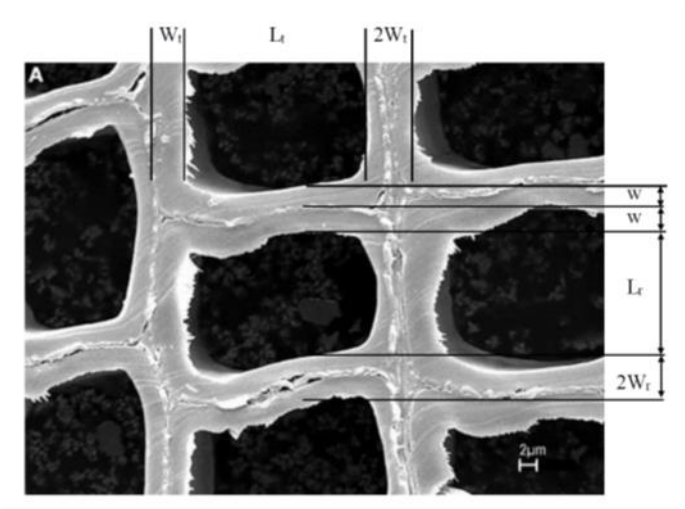

Figure 6: Average tracheid dimensions (Lr+2Wr=15 µm; Lt+2Wt=22 µm; Wr= 2 µm; Wt=3 µm)

It can be assumed that these dimensions are measured at w=0%, i.e. in an absolutely dry state. If the water content is increased to over 30%, the cell walls will reach their maximum swollen state. This means that each part of the cell wall will swell in width and thickness.

From studies at the macroscopic level, it is known that normal wood swells about 4% in the radial direction and about 8% in the tangential direction. Thus, the radial wall will probably swell by 4% in width and 8% in thickness. For the tangential wall, these values will be reversed. Verification of this statement is also the purpose of the work.

For cell wall media, the difference between the "outer "and "inner "size is taken:

$$\left(\frac{L+2W-L}{2}\right), \mu m \tag{1}$$

However, the thickness of the wall varies along its circumference. At the same time, determining the inner and outer diameter is easy (for all types of tracheid shapes). If the centre of mass (i.e. the intersection of the medians) is taken as the centre of the cell, the dimension to the midline will be L/2+W/2 (approximately).

Thus, half of the single wall is obtained for the middle. It is important to note that this is the original size. As it swells, the wall will swell to some degree outwards and some degree

inwards. After swelling, experimental measurements will determine how much the cell has swollen inwardly and how much outwardly.

The marking of the middle lines in the absolutely dry state (the black dashed line) and that in the swollen state (the green line) will be called "first" and "second" for brevity. The use of a rectangular shape for the tracheids is for simplicity (Fig. 6). The offset of the inner and outer walls relative to the middle line can be in three variants:

- the second line should be further out than the first;
- the first and second lines should match;
- the second line should be further inside than the first;

DETERMINATION OF THE SWELLING OF A TRACHEID TO DETERMINE THE DIMENSIONS OF THE TRACHEID

Those shown in Fig. 7 sizes are in an absolutely dry state. Colour lines are selected for the maximally swollen state. The variant in which the middle (green dashed) line is shifted outwards and the cell walls swell both outwards and inwards is chosen. The centre of the cell is assumed to remain constant after swelling. The distances indicated are as follows:

A_1 – the size of the cell in the final swollen state (taking into account the swelling along the width of the tangential wall);

A_2 – the size of the cell in the swollen state, but without taking into account the swelling along the width of the tangential wall;

A_3 – cell size in dry state;

A_4 – the size of the cell gap in the dry state;

A_5 – the size of the cell gap in the swollen state;

B_1 – the size from the midline to the outer border of the cell in the swollen state (corresponding to $(L_r+W)/2$), i.e. the cell wall distance and half the cell gap.

B_2 – the distance from the middle of the cell to the middle line in the swollen state, but without taking into account the swelling along the width of the radial wall;

B_3 – the size from the middle line to the outer border of the cell in a dry state;

B_4 – the distance from the middle of the cell to the middle line in the swollen state;

B_5 – half of the cell gap to the midline in the dry state;

B_6 – half of the cell gap in the radial direction in the dry state (corresponding to $L_r/2$);

B_7 – half of the cell cavity in a radial direction in a swollen state;

C_1 – the thickness of the double cell wall in the final swollen state (taking into account the swelling by the thickness of the tangential wall);

C_2 – the thickness of the double cell wall in the swollen state (without taking into account the swelling by the thickness of the tangential wall);

C_3 – the thickness of the double cell wall in the dry state;

C_4 – the thickness of the single internal cell wall in the dry state;

C_5 – the thickness of the single outer cell wall in the dry state;

C_6 – the thickness of the single inner cell wall in the swollen state;

C_7 – the thickness of the single outer cell wall in the swollen state;

C_8 – the size of the swelling of the cell wall outside;

C_9 – the size of the swelling of the cell wall inwards;

C_{10} – the size of the radial swelling of the radial cell wall;

C_{11} – displacement of the midline of the cell wall after swelling;

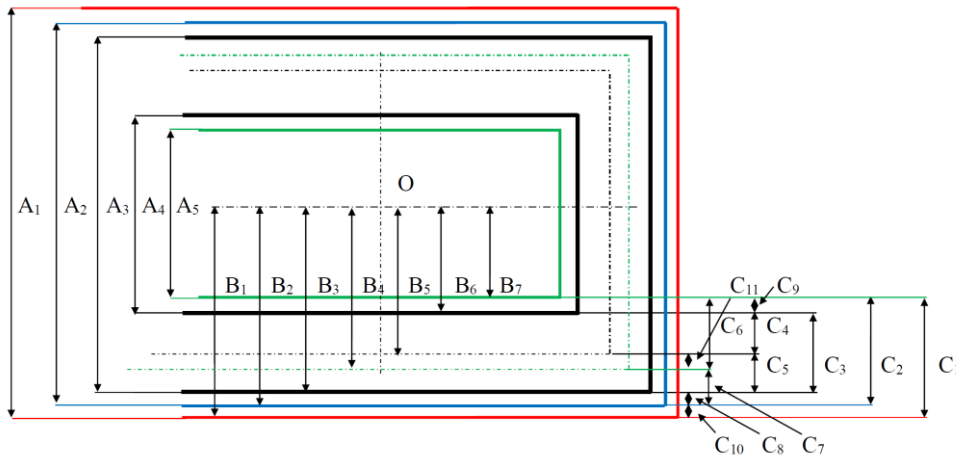

Figure 7: Estimated cell dimensions before and after swelling.

D1 – the sum of the gap and the two double walls in the dry state;
D2 – the sum of the gap and the two double walls in the swollen state;
D3 – cell swelling, %;
D4 – the sum of the cell cavity and the internal walls in the dry state;
D5 – the sum of the cell cavity and internal walls in a swollen state;
D6 – swelling of the single cell, without the involvement of the middle lamella;
D7 – swelling of the middle lamella;

RESULTS AND DISCUSSION

CALCULATION OF SWELLING

Determination of the unit (absolute) swelling of the tracheids depending on their location within the sample. With those shown in Fig. 3 dimensions (about $L_r+2W_r=12$ µm; $L_t+2W_t=20$ µm; $W_r= 5$ µm; $W_t=6$ µm) means that in the tangential direction we will have about 1000 tracheids ($20.10^3/20$). In the radial direction, the calculation of the number is very complicated because of the elongation of the size in the radial direction of the first (earliest) tracheids and their shortening (flattening) in the same direction at the end of the annual ring. That is why an average value is taken to begin with. According to her, there will be about 1700 tracheids in a radial direction ($20.10^3/12$). Each of these tracheids will need to swell (and consequently shrinkage) by a certain amount in the given direction to satisfy the above 4% and 8%. It will be important to calculate by what absolute value each wall changes in width and thickness.

Table 1: Average size change of individual tracheids in the respective direction (obtained on the basis of macroscopic measurements), µm

	Size at FSP is the cell walls µm	Size in absolute dry state µm	Swelling %	Shrinkage %	Absolute difference, µm	Swelling / shrinkage per individual tracheid, µm
Tangentially	21600	20000	8,0	7,4	1600	1,6
Radially	20850	20000	4,3	4,1	850	0,5

In the radial direction, there is an increase in the radial walls by 0.8 µm on both sides (B4-B5), due to the radial swelling of the radial walls. The tangential walls will swell about 0.25 µm on both sides (W2-W5) due to their radial thickness swelling.

$$\alpha_r = \sum \alpha_r^{'} = 850\ \mu m \tag{3.7}$$

$$\alpha_t = \sum \alpha_t^{'} = 1600\ \mu m \tag{3.7}$$

where:

α'r is the absolute radial swelling of each tracheid (about 0.5 µm);

α't – the absolute swelling in the tangential direction of each tracheid (about 1.6 µm).

At a thickness of the microscope slide of about 30 µm, the helices of the S_1 and S_2 layers cannot make a complete revolution around the cell gap. The heart rays (guilty, according to some authors, for the anisotropy of transverse drying) are also destroyed. Therefore, it is expected that the values of this property can be very different.

The rest of the cell parameters were calculated from them. A series of early and late tracheids were measured. From the values of the sought quantities, the following is striking:

- In early tracheids, the amount of cell wall shrinkage inward (C_8+C_{10}) changes as in the radial direction, the wall dries about 2.0 µm, while in the tangential direction, it is 3.5 µm (Fig. 8). In late tracheids, the wall dries out in the radial direction by 5.0 µm, and in the tangential direction by 6.0 µm (Fig. 9).

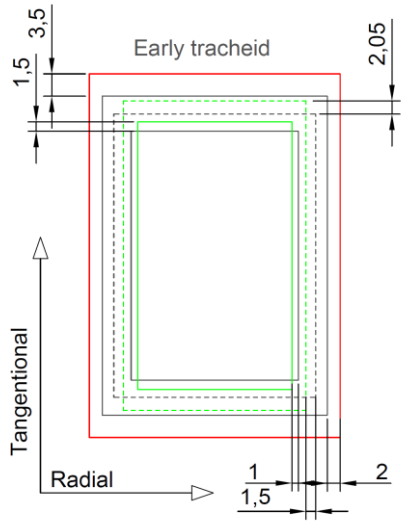

Figure 8: Desiccation of the cell wall and displacement of the midline in an early tracheid, µm (black lines are of the tracheid in the dry state, and dashed lines indicate the middle lamella)

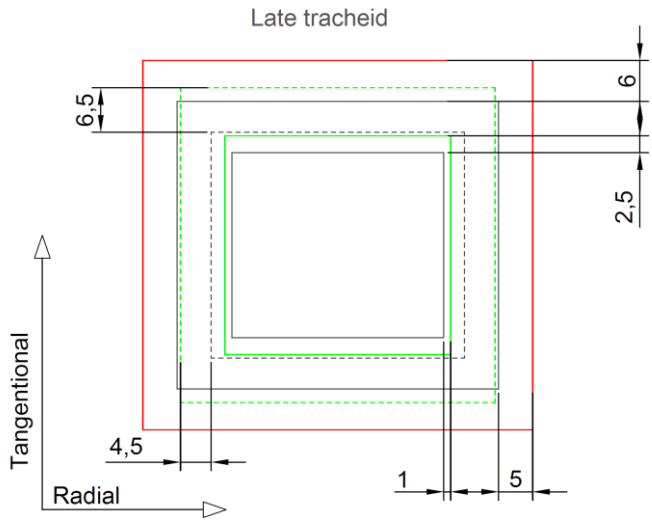

Figure 9: Desiccation of the cell wall and displacement of the midline in a late tracheid, µm

- The extent of inward drying of the cell wall (C_9), in early tracheids in a radial direction instead of shrinkage outward, the wall has retreated inward toward the cell gap (green line) by about 1.5 µm. In the tangential direction, the wall dries inward by about 1.0 µm. In late tracheids, the size has a negative sign in both directions, i.e., the wall retracts outwards by 1.0 µm in the radial direction and 2.5 µm in the tangential direction.

- The displacement of the midline of the cell wall, after desiccation (C_{11}) in the early tracheids in the radial direction, is moved outwards by about 1.5 µm, while in the tangential direction, it is moved inwards by 2.05 µm. In the case of late tracheids, the middle line is shifted inward at 4.5 µm in the radial direction and in the tangential direction – 6.5 µm.

- Analyzing the indicated dimensions, it was determined that the shrinkage of the cell, without the participation of the middle lamella (D_6), the early tracheids shrink in the radial direction 6.7%, and in the tangential direction – 8.9%. In late tracheids, shrinkage is 17.9% in the radial direction and 35.3% in the tangential direction, respectively.

- Thus, the shrinkage of the middle lamella (D_7) in the early tracheids in the radial direction is 7.6%, and in the tangential direction – 5.1%. In late tracheids, these values are respectively 2.9% in the radial direction and 2.2% in the tangential direction.

CONCLUSIONS

- Wood rays are not present in the measured microscopic preparation, which means that their role in the anisotropy of shrinkage and swelling can be determined;
- The thickness of the microscopic preparation is too small for the cellulose matrices in the layers of the secondary cell wall to make a complete turnover. This means that a microscope slide will swell in a very different way than solid wood;
- When experimentally measuring the swelling of a single tracheid, a thicker microscopic preparation should be made, and tree species with tracheids of correct cross-section shape should be selected (Picea abies or Abies alba);

- Regardless of whether the cell wall swells outward or inward, the absolute value of this swelling is too small to be measured by classical microscopic methods.

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