

Effects of natural and artificial ageing on the physical and acoustic properties of wood in musical instruments

著者別名	小幡谷 英一
journal or publication title	Journal of Cultural Heritage
page range	1-7
year	2016-06
URL	http://hdl.handle.net/2241/00144001

doi: 10.1016/j.culher.2016.02.011

Manuscript Number: CULHER-D-15-00230R1

Title: Effects of natural and artificial ageing on the physical and acoustic properties of wood in musical instruments

Article Type: Wooden musical instruments special issue

Keywords: Wood; ageing; musical instrument; hygroscopicity; acoustic property

Corresponding Author: Dr. Eiichi Obataya, Dr

Corresponding Author's Institution: Tsukuba University

First Author: Eiichi Obataya, Dr

Order of Authors: Eiichi Obataya, Dr

Manuscript Region of Origin: JAPAN

Abstract: The reversible and irreversible effects of natural and artificial hydrothermal ageing are reviewed with respect to the hygroscopicity and acoustic properties relevant to the practical quality of wooden musical instruments. Long-term natural ageing reduces the hygroscopicity of wood while improving its acoustic quality, but these changes are partly reversible by exposure to high humidity. Similar reversible changes are observed in hydrothermally treated wood, especially when the wood is heated at an intermediate relative humidity. These reversible changes are attributed to the annealing-like rearrangement of amorphous wood polymers or the temporary closure of micropores, but further investigation is necessary. Color change resulting from natural ageing is shown to be successfully reproducible by oven-heating.

1
2 **Title: Effects of natural and artificial ageing on the physical and acoustic properties of wood**
3
4 **in musical instruments**

5
6
7 **Author: Eiichi Obataya**

8 Graduate School of Life and Environmental Sciences

9 Tsukuba University

10 Ibaraki 305-8572, Japan

11 E-mail: obataya.eiichi.fu@u.tsukuba.ac.jp

12
13
14
15
16
17 **Abstract**

18 The reversible and irreversible effects of natural and artificial hydrothermal ageing are reviewed
19 with respect to the hygroscopicity and acoustic properties relevant to the practical quality of
20 wooden musical instruments. Long-term natural ageing reduces the hygroscopicity of wood while
21 improving its acoustic quality, but these changes are partly reversible by exposure to high humidity.
22 Similar reversible changes are observed in hydrothermally treated wood, especially when the
23 wood is heated at an intermediate relative humidity. These reversible changes are attributed to the
24 annealing-like rearrangement of amorphous wood polymers or the temporary closure of
25 micropores, but further investigation is necessary. Color change resulting from natural ageing is
26 shown to be successfully reproducible by oven-heating.

27
28
29
30
31
32
33
34 **Research aims**

35 This article describes the irreversible and reversible effects of both natural and hydrothermal
36 artificial ageing on the properties of wood relevant to the quality of wooden musical instruments.
37 The effects of ageing are usually attributed to irreversible chemical changes in wood polymers,
38 such as the recrystallization of cellulose and depolymerization of hemicelluloses. However, recent
39 investigations suggest that the effects of ageing are partly recoverable by rewetting or moistening
40 in humid conditions. This reversible phenomenon is likely caused by conformational changes
41 during the physical ageing of wood polymers, which should be considered for the appropriate
42 conservation of the properties of old wooden cultural artifacts.

43
44
45
46
47
48
49
50 **1. Introduction**

51 For musicians and artisans dealing with musical instruments made from wood, the ageing process
52 is not considered degradation but a treatment that improves the acoustic quality and stability of
53 wooden instruments. Old lumber is often priced higher than recently cut wood for making the
54 soundboard and bass bar of stringed instruments. According to Noguchi et al., aged pine wood
55
56
57
58
59
60
61
62
63
64
65

1
2 shows a higher sound velocity (V) and lower loss tangent ($\tan\delta$) than newly cut wood, while the
3 rigidity ratios (E'/G' in which E' is the dynamic Young's modulus and G' is the dynamic shear
4 modulus) of woods of different ages are nearly equal [1]. A higher V and lower $\tan\delta$ increase the
5 amplitude of sound radiation [2, 3], and the high E/G ratio of wood creates its characteristic
6 frequency response [4]. Therefore, wooden soundboards become more resonant while maintaining
7 their tone quality with long-term ageing. Moreover, long-term ageing reduces the hygroscopicity
8 of wood in general [5, 6]. The reduced hygroscopicity improves the dimensional stability of wood,
9 as well as stabilizing its mechanical and acoustic properties depending on the moisture content.
10 Thus, the empirical approach of musicians and artisans to the effects of ageing on the acoustic
11 quality and stability of wood is well-supported by material analysis.

12
13 The enhanced rigidity and reduced damping of aged wood have not yet been fully explained.
14
15 Noguchi et al. have speculated that cellulose crystallization contributes to the higher E value and
16 lower $\tan\delta$ value of aged wood [1], but this hypothesis was later negated: no significant difference
17 was recognized in the crystallinity of new and aged wood samples [7]. Yokoyama et al. also found
18 equivalent degrees of crystallinity between new and aged woods [6]. According to Inagaki et al.,
19 the degree of crystallinity increases by both ageing and hydrothermal treatments which cause the
20 decomposition of the amorphous regions in cellulose and hemicelluloses; however, the increase of
21 crystallite thickness was confirmed only for hydrothermally treated wood [8]. From these results,
22 the crystallization of cellulose can be concluded to be a minor mechanism for the enhancing
23 effects of ageing. Other possible mechanisms include hornification [9], the cross-linking reaction
24 in lignin, and the rearrangement of disordered polysaccharides, but those hypotheses require
25 further experimentation.

26
27 Meanwhile, the improved stability or reduced hygroscopicity of aged wood is fully explained by
28 the decomposition of hemicelluloses, which is the most significant chemical change in wood
29 during ageing [10, 11]. As hemicelluloses are the most hygroscopic components of wood, their
30 decomposition reduces the hygroscopicity. The decomposition of hemicelluloses also reduces the
31 ductility and toughness of wood; these amorphous polysaccharides are crucial to the tight
32 connections between reinforcing cellulose microfibrils and amorphous matrix polymers [12-14]. In
33 fact, the tensile strength [15], rupture energy in bending [6], impact bending strength [5, 16], and
34 cleavage resistance [5] are all significantly reduced by ageing. These adverse effects of ageing are
35 qualitatively similar to those induced by hydrothermal treatment, which involves the remarkable
36 hydrolysis of hemicelluloses [18]. The fragile nature of aged wood must be considered when
37 wooden parts of musical instruments are loaded with strong forces, such as the tension of strings.
38 In general, the effects of ageing are most frequently explained as results of irreversible changes,
39 such as the decomposition of hemicelluloses. However, recent investigations have suggested that
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

1
2 the effects of both natural and artificial hydrothermal ageing can be partly reversed by moistening
3 or rewetting the wood sample [18–20]. These reports imply that the wood ageing process involves
4 some recoverable, i.e., reversible changes in the wood cell wall. The reversible effect of ageing
5 may be related to the effects of seasoning and playing, which must be considered for the
6 appropriate conditioning of old wooden instruments. In this paper, the reversible and irreversible
7 effects of natural and artificial ageing on the physical properties of wood are described. In addition,
8 some recent studies on the color of aged wood are briefly discussed regarding the restoration and
9 imitation of the physical appearance of old wooden musical instruments.
10
11
12
13
14
15

16 **2. The effects of natural ageing**

17 **2.1. Change in hygroscopicity**

18 The hygroscopicity of wood sample can be evaluated by the equilibrium moisture content (EMC)
19 at a constant relative humidity (RH). In general, the EMC of wood at an intermediate RH
20 (60–65%) remains unchanged [15] or decreases during long-term ageing [5, 6]. Some aged wood
21 shows higher hygroscopicity than new wood [21], but in these cases, the aged wood is similar to
22 submersed (water-logged) wood in terms of moisture sorption characteristics [22]. As submersed
23 lumber is rarely used for musical instruments, these cases are irrelevant to this paper.

24 The reduced hygroscopicity of aged wood is usually attributed to the decomposition and loss of
25 hygroscopic hemicelluloses. This explanation holds if the effects of ageing are always irreversible.
26 However, the reduced hygroscopicity of aged wood was recently found to be recoverable by a
27 moistening treatment. Figure 1 shows the relative EMC of aged wood before and after the
28 moistening treatment [19]. In Fig.1, the EMC values of aged wood are normalized relative to those
29 of recently cut wood under the same conditions. The EMC values of aged wood are always lower
30 than those of recently cut wood, but with moistening at 100% RH for several days, its EMC values
31 are at least partially recovered, especially at low RH. Notably, the different trends in Fig.1 do not
32 reflect sorption hysteresis, because these EMC values are always determined in the adsorption
33 process. Thus, the effect of ageing is partly reversible with respect to hygroscopicity.

34 A possible interpretation for the cause of the temporary reduction in hygroscopicity by ageing is
35 the physical ageing or annealing of wood polymers. Figures 2 and 3 schematically illustrate the
36 fine structure of wood cell wall and the mechanism of internal stress during drying, respectively.
37 The wood cell wall is a composite in which crystalline cellulose fibers are embedded in
38 amorphous matrix substances. The crystalline cellulose is rigid and hydrophobic, whereas the
39 amorphous matrix polymers are swollen with moisture in green state and ready to shrink by drying.
40 As wood dries from its green state (Fig.3A), the shrinkage of amorphous wood polymers is
41 restricted by the adjacent crystalline cellulose. Therefore, the amorphous polymers are distorted
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

1
2 and dry under applied stress, as in the quenching of synthetic polymers (Fig.3B). Since the
3 amorphous wood polymers are glassy and immobile in dry conditions, the drying stress cannot
4 immediately be relaxed. During long-term ageing, however, it may gradually relax (Fig.3E) and
5 such an annealing-like rearrangement of wood polymers reduces the hygroscopicity of the wood.
6 On the other hand, the wood polymers recover their initial arrangements once they are re-swollen
7 and plasticized by moisture at high RH (Fig.3G). Therefore, the hygroscopicity is recovered by
8 moistening and subsequent drying. This hypothesis was originally proposed to explain the
9 temporary reduction in hygroscopicity resulting from oven heating [23], but can be applied to the
10 present case in which the natural ageing process can be regarded as a long-term hydrothermal
11 treatment at ambient temperature. The annealing effect may also account for the reduction in
12 hygroscopicity caused by repeated humidity cycling [24]. Although it is technically difficult to
13 observe the slight conformational changes in amorphous wood polymers, X-ray diffractometry
14 may give useful information about the distortion of amorphous wood polymers, as it can detect the
15 deformation of cellulose crystals accompanied by the shrinkage and swelling of amorphous wood
16 polymers [25].

17
18 Another potential mechanism underlying the temporary reduction in hygroscopicity by ageing is
19 the closure of micropores. Kojiro et al. [18] have determined the presence of micropores smaller
20 than 0.6 nm in new and aged cypress wood (0.1 – 1000 y) by CO₂ adsorption measurements. The
21 micropores decreased in number with ageing, but increased after rewetting and drying. This
22 coincides with the temporary reduction in EMC from ageing and its recovery by moistening,
23 particularly at a low RH (Fig.1). The temporary closure of micropores is possibly responsible for
24 the enhanced rigidity of aged wood, as well as the reduction in hygroscopicity by repeated
25 humidity cycling [24].

26
27 As shown in Fig.1, the EMC values of aged wood remain lower than those of new wood at a
28 higher RH, whereas at a low RH they become closer to those of new wood after the moisture
29 recovery treatment. This trend is similar to that observed in oven-heated wood, whereas steamed
30 wood shows a reverse trend [26]. The greater reduction in EMC at a higher RH is also observed in
31 formaldehyde-treated wood [27] in which the swelling of wood is restricted by the tight covalent
32 bonds formed between the amorphous wood polymers. These facts imply that the irreversible
33 effects of ageing result from cross-linking or similar structural changes that restrict the swelling of
34 wood, rather than a decreasing number of active adsorption sites.

35 36 37 38 39 40 41 42 43 44 45 46 47 48 49 50 51 52 53 **2.2. Changes in vibrational properties**

54 In general, a higher specific dynamic Young's modulus (E'/ρ), higher sound velocity (V), and
55 lower mechanical loss tangent ($\tan\delta$) are desired for the soundboards of musical instruments [2-4,
56
57
58
59
60
61
62
63
64
65

1
2
3 25]. These vibrational properties depend not on the density and wood species, but on the
4 microfibril angles in the cell wall [26, 27]. In some species, $\tan \delta$ is considerably reduced [28] or
5 enhanced [29] in the presence of water-soluble extractives, but the particular effects of these
6 extractives are not detailed here.

7
8
9 Noguchi et al. have found that long-term ageing improves the acoustic quality of red pine wood
10 [1]. This coincides with the empirical knowledge of musicians and artisans. However, recently,
11 Maejima et al. have found that the vibrational properties of aged wood are influenced by
12 moistening [19]. Figure 4 shows the relative E'/ρ and $\tan \delta$ values of aged wood at 25°C and 60%
13 RH as a function of EMC. The EMC in the desorption process is higher than that in the adsorption
14 process because of the moisture sorption hysteresis. In addition, moistening affects the EMC of
15 aged wood, as described above. Notably, E'/ρ decreases and $\tan \delta$ increases after moistening at
16 100%RH. This indicates the temporary effects of enhanced E'/ρ and reduced $\tan \delta$ by ageing;
17 however, these temporary effects are reset by the moistening process. The temporary effects must
18 be closely related to the annealing of wood polymers and closure of micropores, but a convincing
19 mechanism for this relationship has not yet been reported.

20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65
The temporary and recoverable effects of ageing must be considered when we wish to maintain the
acoustic quality of old wooden instruments. Once an old instrument is exposed to highly humid
conditions, the temporary effects of ageing will disappear and the ageing-improved quality of the
instrument may degrade.

3. Artificial ageing of wood

Hydrothermal treatment is an ecologically friendly method to improve the practical performance
of wood. The dimensional stability, durability, and mechanical properties of wood are generally
improved when the wood is heated in air, oil, or water vapor at temperatures higher than 100°C.
Recent investigations on the hydrothermal treatment of wood and its potential applications were
reviewed by Sandberg et al. [17].

The hydrothermal treatment is sometimes referred to as artificial or accelerated ageing, because
the chemical reactions involved in natural ageing are accelerated by the elevated temperature.
Strictly speaking, however, the claim of accelerated ageing has little basis in reality, because the
ageing of wood involves many different chemical reactions following different reaction kinetics.
Chédeville and co-researchers have confirmed that ultraviolet resonance Raman spectra of
artificially aged wood were dissimilar to those of naturally aged wood when the wood was heated
to 160°C or above [33]. This suggests that the properties of naturally aged wood are not easily
reproduced by the hydrothermal treatment of new wood. However, particular properties of aged
wood, such as enhanced rigidity, reduced hygroscopicity, and darkened color, can be successfully

1
2 reproduced by appropriate hydrothermal treatment. In this section, the effects of hydrothermal
3 treatment on the hygroscopicity, vibrational properties, and color of wood are described,
4 summarizing its potential for the imitation of naturally aged wood.
5
6
7

8 **3.1 Reduction in hygroscopicity**

9
10 In general, hydrothermal treatment is conducted in dry air (0% RH) or in saturated water vapor
11 (100% RH); these extreme conditions are easily produced by using an oven and autoclave,
12 respectively. Oven-heating in dry air reduces the EMC of wood, and the reduction in the EMC is
13 greater at a higher RH. Steam-heating the wood, meanwhile, reduces the EMC to a greater degree
14 at a lower RH [26]. These inverse trends suggest that the effects of oven-heating and steaming are
15 qualitatively different, and that the heating relative humidity (HRH) is important in determining
16 the moisture sorption characteristics of hydrothermally treated wood. Recently Borrega and
17 Kärenlampi [34] found that the largest reduction in EMC (at 19°C and 65%RH) was achieved by
18 heating at an intermediate HRH (50%). They speculated that irreversible hydrogen bonding,
19 so-called hornification, was responsible for the particular effect of hydrothermal treatment at an
20 intermediate HRH. However, Endo et al. found more recently that the reduced hygroscopicity of
21 hydrothermally treated wood was considerably recovered by moistening, especially when the
22 wood was heated at an intermediate HRH [20]. Figure 5 shows the effects of HRH on the EMC
23 values of hydrothermally treated wood at 25°C and 60% RH. The EMC is minimized by heating at
24 an intermediate HRH (40–80%), but once the hydrothermally treated wood is moistened under
25 highly humid conditions, its EMC recovers significantly, showing a monotonic decrease with an
26 increasing HRH. The recovery in EMC cannot be attributed to moisture sorption hysteresis
27 because the EMC values are always determined in the adsorption process: the moistened samples
28 were completely dried in vacuo and then conditioned at 25°C and 60% RH for a few months
29 before determining their EMC values.
30
31
32
33
34
35
36
37
38
39
40
41

42 Figure 6 shows the effects of WL on the EMC of hydrothermally treated wood both before and
43 after the moistening treatment. As suggested by Borrega and Kärenlampi, the EMC is effectively
44 reduced by heating at an intermediate (29–64%) HRH (Fig.6a), but this particular effect disappears
45 after the moistening treatment (Fig.6b). These results indicate the temporary reduction of the EMC
46 of wood by hydrothermal treatment, particularly when the wood is heated at an intermediate HRH.
47 The temporary effect of hydrothermal treatment presumably results from the annealing or physical
48 ageing of wood polymers as observed during the natural ageing of wood. When wood is heated at
49 a low HRH, the thermal activation of the wood polymers allows their rearrangement accompanied
50 by the relaxation of the drying stress, as mentioned above. Such an annealing effect is maximized
51 at an intermediate HRH because a certain amount of moisture can act as a plasticizer, easing the
52
53
54
55
56
57
58
59
60
61
62
63
64
65

1
2 rearrangement of wood polymers. At a higher HRH, the annealing effect (or temporary reduction
3 in EMC) decreases because less initial drying stress exists to be relaxed. Meanwhile, the
4 irreversible effects monotonically increase with an increasing HRH, because the decomposition of
5 wood polymers such as the hydrolysis of hemicelluloses is accelerated in the presence of moisture.
6 The temporary and reversible effects of hydrothermal treatment have not been considered in many
7 cases. When we evaluate the hygroscopicity of hydrothermally treated wood, the irreversible effect
8 should be clearly distinguished from the reversible effect; otherwise, we may overestimate the
9 practical performance of the treatment regarding metrics such as dimensional stability.
10 Additionally, the reversible effect should be considered when hydrothermally treated woods are
11 used to imitate and restore the properties of old cultural artifacts, because the dimensional and
12 moisture-dependent mechanical properties may irreversibly change after exposure to high
13 humidity, as would occur in the rainy season.
14
15
16
17
18
19
20
21
22

23 **3.2 Changes in vibrational properties**

24 Many investigations have been made on the mechanical properties of hydrothermally treated or
25 kiln-dried wood for reasons of practical importance [17]. In general, the mechanical properties of
26 hydrothermally treated wood are well correlated with the WL, irrespective of heating temperature.
27 With increasing WL, the bending Young's modulus (E) of wood increases slightly and then
28 decreases whereas its bending strength and toughness decrease monotonically [35-38]. The E'/ρ
29 values are also enhanced by both dry heating [39] and steaming [40]. These enhancing effects have
30 usually been attributed to the recrystallization of cellulose, while the reduced strength and ductility
31 are attributed to the decomposition of hemicelluloses [39, 41].
32
33
34
35
36

37 Meanwhile, little information is available regarding the $\tan\delta$ of hydrothermally treated wood.
38 According to Kubojima et al., the $\tan\delta$ of spruce wood in the direction parallel to its fibers
39 increases by heating the wood at 160°C for several hours in dry nitrogen gas, whereas that of green
40 spruce wood slightly decreases when heated in an autoclave at high HRH [40]. These changes in
41 $\tan\delta$ may reflect different structural changes in the wood polymers, such as the cross-linking
42 reaction in lignin [42, 43] and the recrystallization of cellulose [39, 41]. By heating at higher
43 temperatures or for longer durations, the $\tan\delta$ increases to a remarkable degree, probably because
44 of the depolymerization of hemicelluloses. These results imply that relatively mild heating may
45 enhance the acoustic quality of wooden soundboards.
46
47
48
49
50

51 Few attempts have been made on investigating the effects of heating at an intermediate HRH, and
52 little attention has been paid to the reversible effects of the hydrothermal treatment. Endo et al.
53 recently reported a precise measurement of the E'/ρ and $\tan\delta$ of spruce wood, before and after
54 hydrothermal treatment at 120°C and various HRH (0–95%) for 1–7 days [20]. Figure 7 shows the
55
56
57
58
59
60
61
62
63
64
65

1
2 changes in E'/ρ and $\tan\delta$ values of Sitka spruce wood with hydrothermal treatment at 57–64%
3 HRH. With heating at an intermediate HRH, the E'/ρ increased and the $\tan\delta$ decreased. These
4 results indicate that the acoustic qualities of wood can be improved by heating at an intermediate
5 HRH. However, the effects of hydrothermal treatment are, importantly, recoverable and not
6 permanent. In fact, the vibrational properties of hydrothermally treated wood approach those of
7 unmodified wood after a moistening treatment, as shown in Fig.7. That is, the effects of
8 hydrothermal treatment could be easily overestimated unless the wood is tested after sufficient
9 moistening.

10 Figure 8 shows the *irreversible* effects of hydrothermal treatment on the E'/ρ and $\tan\delta$ values of
11 spruce wood. When wood is heated at a high HRH (89–95%), the E'/ρ decreases remarkably and
12 $\tan\delta$ increases steeply with increasing WL. At 0% HRH, the E'/ρ and $\tan\delta$ remain almost
13 unchanged, because the thermal degradation proceeds very slowly in the absence of moisture. In
14 contrast, a slight but significant increase in E'/ρ and decrease in $\tan\delta$ are observed when the wood
15 is heated at an intermediate (29–82%) HRH. This suggests that hydrothermal treatment may
16 improve the acoustic quality of wooden soundboards, and that appropriate humidity control is
17 necessary for effective modification with minimal degradation. Further investigations are
18 necessary to clarify the effects of HRH at different heating temperatures, which will allow
19 time–temperature–humidity conversions in the hydrothermal treatment of wood.
20
21
22
23
24
25
26
27
28
29
30

31 32 **3.3 Changes in color**

33 The color of wood darkens gradually with long-term ageing, even when the wood is carefully
34 conserved indoors, kept dry, protected from sunlight, and prevented from biological attacks. The
35 color of wood is especially important for the imitation and restoration of old wooden instruments.
36 According to Matsuo et al. [44-46], the darkened color of aged wood can be precisely reproduced
37 by oven-heating, because the time–temperature superposition principle is applicable to the color
38 changes. Thus, the oven-heating of wood is a useful method for the restoration and imitation of the
39 visual properties of old wooden artifacts.
40
41
42
43
44
45

46 47 **4. Effects of forced vibration (playing effect)**

48 Many musicians believe that the quality of instrument is improved by continuous playing; however,
49 sensory evaluation tests indicate that playing has no significant effect on the tone quality of guitars
50 and violins [47, 48]. Continuous vibration, on the other hand, has been reported several times to
51 affect the vibrational properties of wood. Sobue and Okayasu found that the $\tan\delta$ of wood
52 decreased exponentially with time under forced vibration, whereas the E' remained unchanged,
53 irrespective of wood species [49]. A similar phenomenon was reported by Hunt and Balsan [50].
54
55
56
57
58
59
60
61
62
63
64
65

1
2
3 Sobue and Okayasu hypothesized that the $\tan\delta$ of wood polymers was increased by the drying
4 stress remaining in the wood cell wall, and that continuous vibrations may have stimulated the
5 relaxation of the drying stress, thus reducing the $\tan\delta$ of wood. This speculation seems reasonable,
6 because continuous vibration only slightly affected the $\tan\delta$ of spruce wood at 100% RH, in which
7 only a small amount of drying stress remained in the wood cell wall [51]. If the relaxation of
8 drying stress is a major mechanism underlying the so-called playing effect, the changes in
9 properties induced by the playing effect may be recovered by moistening and subsequent drying.
10 Therefore, it is advisable to conserve wooden musical instruments in dry conditions, because the
11 improved acoustic performance may degrade when the wood is exposed to humid conditions.
12
13
14
15
16
17

18 **5. Conclusions**

19
20 The reversible and irreversible effects of natural and artificial ageing are discussed, with respect to
21 the hygroscopicity and acoustic properties relevant to the practical quality of wooden musical
22 instruments. Long-term natural ageing affects the hygroscopicity and acoustic properties of wood,
23 but the effects of ageing are at least partially recovered by moistening the wood under highly
24 humid conditions. Similar reversibility is observed in hydrothermally treated wood, especially
25 when the wood is heated at an intermediate relative humidity. The physical or annealing-like
26 rearrangement of amorphous wood polymers and/or the temporary closure of micropores are
27 hypothesized as mechanisms underlying the reversible changes in wood properties; further
28 investigations are necessary to test these hypotheses.
29
30
31
32
33
34
35

36 **Acknowledgements**

37 The author appreciates Ms. Kaoru Endo and Mr. Shuoye Chen for their permission to reproduce
38 their presentation. The author is also grateful to Dr. Miyuki Matsuo and Dr. Marc Borrega for
39 providing useful information about the hydrothermal treatment of wood.
40
41
42
43

44 **References**

- 45 [1] T. Noguchi, E. Obataya, K. Ando, Effects of aging on the vibrational properties of wood, *J.*
46 *Cultural Heritage* 13S (2012) S21–S25.
47
48 [2] H. Yano, H. Matsuhisa, Study on the timber of wood II, analysis of the sound spectrum of
49 wood using viscoelastic Timoshenko equation, *Sci. Rep. Kyoto Prefectural Univ.* 43 (1991) 24–31.
50
51 [3] T. Ono, Frequency responses of wood for musical instruments in relation to the vibrational
52 properties, *J. Acoust. Soc. Jpn. (E)* 17 (1996) 183–193.
53
54 [4] T. Ono, Transient response of wood for musical instruments and its mechanism in vibrational
55 property, *J. Acoust. Soc. Jpn. (E)* 20 (1999) 117–124.
56
57
58
59
60
61
62
63
64
65

- 1
2 [5] J. Kohara, Studies on the permanence of wood VI, the changes of mechanical properties of old
3 timbers (in Japanese), Bull. Kyoto Prefectural Univ. 6 (1954) 164–174.
4
5 [6] M. Yokoyama, J. Gril, M. Matsuo, H. Yano, J. Sugiyama, B. Clair, S. Kubodera, T. Mitsutani,
6 M. Sakamoto, H. Ozaki, M. Imamura, S. Kawai, Mechanical characteristics of aged Hinoki wood
7 from Japanese historical buildings, Comp. Rend. Phys. 10 (2009) 601–611.
8
9 [7] T. Noguchi, E. Obataya, K. Ando, H. Yamamoto, Y. Ogawa, M. Wada, Vibrational properties of
10 aged pine wood (*Pinus densiflora*), Abstracts of the 62th Annual Meeting of the Japanese Wood
11 Research Society (2012), C16-04-1515.
12
13 [8] T. Inagaki, H. W. Siesler, K. Mitsui, S. Tsuchikawa, Difference of the crystal structure of
14 cellulose in wood after hydrothermal and aging degradation: A NIR spectroscopy and XRD study,
15 Biomacromolecules 11 (2010), 2300–2305.
16
17 [9] K. L. Kato, R. E. Cameron, Structure-property relationships in thermally aged cellulose fibers
18 and paper, J. Appl. Polym. Sci. 74 (1999), 1465-1477.
19
20 [10] J. Kohara, Studies on the permanence of wood VII, the influence of age on the components of
21 wood (*Chamaecyparis obtusa* Endl.) (in Japanese), Bull. Kyoto Prefectural Univ. 6 (1954)
22 175–182.
23
24 [11] R. Widyorini, M. Yokoyama, J. Sugiyama, S. Kawai, T. Mitsutani, S. Kubodera, M. Ozaki, M.
25 Sakamoto, M. Imamura, Evaluation of aged wood from historical Japanese buildings II – Changes
26 in chemical component, Abstracts of the 57th Annual Meeting of the Japanese Wood Research
27 Society (2007) C09-1015.
28
29 [12] M. Åkerholm, L. Salmén, Interactions between wood polymers studied by dynamic FT-IR
30 spectroscopy, Polymer 42 (2001), 963–969.
31
32 [13] K. Abe, H. Yamamoto, Change in mechanical interaction between cellulose microfibril and
33 matrix substance in wood cell wall induced by hygrothermal treatment. J Wood Sci 52
34 (2006), 107–110.
35
36 [14] R. Kaida, E. Obataya, M. Yoshida, F. Ishiguri, J. Tanabe, T. Taniguchi, M. Kurita, K. Baba, T.
37 Hayashi, Occurrence of xyloglucan in the xylem of poplar stems for wind and earthquake,
38 Abstracts of the International Symposium on Wood Science and Technology, Tokyo, Japan (2015),
39 5FS-O04.
40
41 [15] Y. Hirashima, M. Sugihara, Y. Sasaki, K. Ando, M. Yamasaki, Strength properties of aged
42 wood I, tensile strength properties of aged Keyaki and Akamatsu woods (in Japanese), Mokuzai
43 Gakkaishi 50 (2004) 301–309.
44
45 [16] Y. Hirashima, M. Sugihara, Y. Sasaki, K. Ando, M. Yamasaki, Strength properties of aged
46 wood III, static and impact bending strength properties of aged Keyaki and Akamatsu woods (in
47 Japanese), Mokuzai Gakkaishi 51 (2005) 146–152.
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

- 1
2
3 [17] D. Sandberg, P. Haller, P. Navi, Thermo-hydro and thermo-hydro-mechanical wood
4 processing: An opportunity for future environmentally friendly wood products, *Wood Mat.Sci.Eng.*
5 8 (2013) 64–88.
6
7 [18] K. Kojiro, Y. Furuta, M. Ohkoshi, Y. Ishimaru, M. Yokoyama, J. Sugiyama, S. Kawai, T.
8 Mitsutani, H. Ozaki, M. Sakamoto, M. Imamura, Changes in micropores in dry wood with elapsed
9 time in the environment. *J. Wood Sci.* 54 (2008), 515–519.
10
11 [19] H. Maejima, K. Endo, E. Obataya, Effects of moistening treatment on the hygroscopicity and
12 vibrational properties of aged wood, Abstracts of the International Symposium on Wood Science
13 and Technology, Tokyo, Japan (2015), 1WP-P01.
14
15 [20] K. Endo, H. Maejima, E. Obataya, Hygroscopicity and vibrational properties of
16 hydrothermally-treated wood, Abstracts of the International Symposium on Wood Science and
17 Technology, Tokyo, Japan (2015), 1WP-P05.
18
19 [21] L. G. Esteban, F. G. Fernández, A. G. Casasús, P. Palacios, J. Gril, Comparison of the
20 hygroscopic behaviour of 205-year-old and recently cut juvenile wood from *Pinus sylvestris* L.,
21 *Ann. For. Sci.* 63 (2006) 309– 317
22
23 [22] L. G. Esteban, P. Palacios, F. G. Fernández, A. Guindeo, M. Conde, V. Baonza, Sorption and
24 thermodynamic properties of juvenile *Pinus sylvestris* L. wood after 103 years of submersion,
25 *Holzforschung* 62 (2008), 745–751.
26
27 [23] E. Obataya, B. Tomita, Hygroscopicity of heat-treated wood II. Reversible and irreversible
28 reductions in the hygroscopicity of wood due to heating (in Japanese), *Mokuzai Gakkaishi* 48
29 (2002) 288–295.
30
31 [24] L. G. Esteban, J. Gril, P. D. Palacios, A. G. Casasús, Reduction of wood hygroscopicity and
32 associated dimensional response by repeated humidity cycles. *Ann. For. Sci.* 62 (2005), 275–284.
33
34 [25] K. Abe, H. Yamamoto, Mechanical interaction between cellulose microfibril and matrix
35 substance in wood cell wall determined by X-ray diffraction. *J Wood Sci* 51 (2005), 334–338.
36
37 [26] E. Obataya, T. Higashihara, B. Tomita, Hygroscopicity of heat-treated wood III, Effects of
38 steaming on the hygroscopicity of wood (in Japanese), *Mokuzai Gakkaishi* 48 (2002) 348–355.
39
40 [27] R. Yasuda, K. Minato, M. Norimoto, Moisture adsorption thermodynamics of chemically
41 modified wood, *Holzforschung* 49 (1995), 548–554.
42
43 [28] M. Norimoto, Structure and properties of wood used for musical instruments I, on the
44 selection of wood used for piano soundboards (in Japanese), *Mokuzai Gakkaishi* 28 (1982), 407–
45 413.
46
47 [29] E. Obataya, T. Ono, M. Norimoto, Vibrational properties of wood along the grain, *J. Materials*
48 *Sci.* 35 (2000) 2993–3001, 6317.
49
50 [30] R. Hori, M. Müller, U. Watanabe, H. C. Lichtenegger, P. Fratzl, J. Sugiyama, The importance
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

- 1
2 of seasonal differences in the cellulose microfibril angle in softwoods in determining acoustic
3 properties, *J. Materials Sci.* 37 (2002) 4279 – 4284.
- 4
5 [31] M. Matsunaga, M. Sugiyama, K. Minato, M. Norimoto, Physical and mechanical properties
6 required for violin bow materials, *Holzforschung* 50 (1996), 511–517.
- 7
8 [32] E. Obataya, M. Norimoto, Acoustic properties of a reed (*Arundo donax* L.) used for the
9 vibrating plate of a clarinet, *J. Acoust. Soc. Am.* 106 (1999), 1106–1110.
- 10
11 [33] C. G. Chédeville, A. S. Jääskeläinen, J. Froidevaux, M. Hughes, P. Navi, Natural and artificial
12 ageing of spruce wood as observed by FTIR-ATR and UVRR spectroscopy. *Holzforschung* 66
13 (2012), 163–170.
- 14
15 [34] M. Borrega, P. Kärenlampi, Hygroscopicity of heat-treated spruce wood, *Proceedings of the*
16 *Nordic Workshop in Wood Engineering*, Skellefteå, Sweden (2007).
- 17
18 [35] M. A. Millett, C. C. Gerhards, Accelerated aging: Residual weight and flexural properties of
19 wood heated in air at 115° to 175°C, *Wood Science* 4 (1972), 193-201.
- 20
21 [36] E.Obataya, S.Shibutani, K.Hanata, S.Do, Effects of high temperature kiln drying on the
22 practical performances of Japanese cedar wood (*Cryptomeria japonica* D. Don) II : changes in
23 mechanical properties due to heating, *J.Wood Sci.* 52 (2006), 111-114.
- 24
25 [37] M. Borrega, P. Kärenlampi, Mechanical behavior of heat-treated spruce (*Picea abies*) wood at
26 constant moisture content and ambient humidity, *Holz Roh Werkst* 66 (2008), 63–69.
- 27
28 [38] Y. Kubojima, T. Okano, M. Ohta, Bending strength and toughness of heat-treated wood, *J.*
29 *Wood Sci.* 46 (2000), 8–15.
- 30
31 [39] Y. Kubojima, T. Okano, M. Ohta, Vibrational properties of sitka spruce heat-treated in
32 nitrogen gas, *J. Wood Sci.* 44 (1998), 73–77.
- 33
34 [40] Y. Kubojima, T. Okano, M. Ohta, Vibrational properties of heat-treated green wood, *J. Wood*
35 *Sci.* 46 (2000), 63–67.
- 36
37 [41] M. T. R. Bhuiyan, N. Hirai, N. Sobue, Changes of crystallinity in wood cellulose by heat
38 treatment under dried and moist conditions, *J. Wood Sci.* 46 (2000) 431–436.
- 39
40 [42] B. F. Tjeerdsma, H. Militz, Chemical changes in hydrothermal treated wood: FTIR analysis of
41 combined hydrothermal and dry heat-treated wood, *Holz als Roh- und Werkstoff* 63 (2005),
42 102–111.
- 43
44 [43] H. Wikberg, S. L. Maunu, Characterisation of thermally modified hard- and softwoods by ¹³C
45 CPMAS NMR, *Carbohydrate Polymers* 58 (2004), 461–466.
- 46
47 [44] M. Matsuo, M. Yokoyama, K. Umemura, J. Gril, K. Yano, S. Kawai, Color changes in wood
48 during heating: Kinetic analysis by applying a time-temperature superposition method, *Appl. Phys.*
49 *A* 99 (2010), 47–52.
- 50
51 [45] M. Matsuo, M. Yokoyama, K. Umemura, J. Sugiyama, S. Kawai, J. Gril, S. Kubodera, T.
- 52
53
54
55
56
57
58
59
60
61
62
63
64
65

- 1
2 Mitsutani, H. Ozaki, M. Sakamoto, M. Imamura, Ageing of wood: Analysis of color changes
3 during natural aging and heat treatment, *Holzforschung* 65 (2011), 361–368.
4
5 [46] M. Matsuo, E. Karami, S. Bardet, J. Gril, J. Froidevaux, P. Navi, Modelling of the changes in
6 wood properties during hygro-thermal treatment, Abstracts of the 63th Annual Meeting of the
7 Japanese Wood Research Society (2013), X27-06-1645.
8
9 [47] B. M. Clemens, J. Kadis, D. M. Clemens, E. Pollak, P. Clark, J. R. Groves, Effect of vibration
10 treatment on guitar tone: a comparative study, *Savart J.* (September 2014), 1–9.
11
12 [48] Ra Inta, J. Smith, J. Wolfe, Measurement of the effect on violins of ageing and playing,
13 *Acoustics Australia* 33 (2005), 25–29.
14
15 [49] N. Sobue, S. Okayasu, Effects of continuous vibration on dynamic viscoelasticity of wood (in
16 Japanese), *J. Soc. Mat. Sci. Jpn.* 41 (1992), 164–169.
17
18 [50] D. G. Hunt, E. Balsan, Why old fiddles sound sweeter, *Nature* 379 (1996), 681.
19
20 [51] H. Akahoshi, S. Chen, E. Obataya, Effects of continuous vibration on the dynamic
21 viscoelastic properties of wood, Annual Conference of COST FP1302 WoodMusICK, London, UK
22 (2015).
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

1
2
3 *Captions for figures*
4

5 **Figure 1** Relative equilibrium moisture content (EMC) values of aged red pine wood (279 y
6 after cutting) as a function of relative humidity (RH) [19].
7

8 *Open plots:* aged wood as received; *filled plots:* aged wood previously moistened at 25°C and
9 100% RH for several weeks. All EMC values were determined in the adsorption process.
10
11

12 **Figure 2** Multi-layered structure of the wood cell wall (A) and fiber-matrix structure of each
13 layer (B).
14
15
16

17 **Figure 3** A viscoelastic model to explain the drying stress and its relaxation due to ageing and
18 hydrothermal treatment.
19

20 a, Viscoelastic matrix; b, adsorbed water; c, elastic crystalline part.
21
22
23

24 **Figure 4** E'/ρ and $\tan\delta$ of aged red pine wood (279 y after cutting) at 25°C and 60% RH plotted
25 against the EMC [19].
26

27 *Filled circles:* aged wood as received; *open triangles:* desorption process after moistening
28 treatment at 25°C and 100% RH; *open squares:* adsorption process after the moistening treatment
29 and subsequent drying in vacuo. The E'/ρ and $\tan\delta$ values are normalized against those determined
30 before the moistening treatment. Each plot indicates the average value of 23 wood specimens.
31 Broken lines indicate the estimated values based on the moisture dependence of unmodified wood.
32
33
34
35
36

37 **Figure 5** Effects of heating relative humidity (HRH) on the EMC of hydrothermally treated
38 Sitka spruce wood at 25°C and 60% RH [20].
39

40 *Open plots:* hydrothermally treated at 120°C for 1 day; *filled plots:* hydrothermally treated at
41 120°C for 1 day and then moistened at 25°C and 100% RH. All EMC values were determined in
42 the adsorption process.
43
44
45
46

47 **Figure 6** EMC values of hydrothermally treated Sitka spruce wood at 25°C and 60% RH plotted
48 against WL before (a) and after (b) the moistening treatment at 25°C and 100% RH [20].
49

50 *Crosses:* unmodified; *open circles:* heated at 0% HRH (heated dry); *filled circles:* heated at
51 29–39% HRH; *filled triangles:* heated at 57–64% HRH; *filled squares:* heated at 77–82% HRH;
52 *open squares:* heated at 89–95% HRH (steamed). Wood specimens were hydrothermally treated in
53 an autoclave at 120°C for 1–7 days. All EMC values were determined in the adsorption process.
54
55
56
57
58
59
60
61
62
63
64
65

1
2
3 **Figure 7** Changes in E'/ρ and $\tan\delta$ of spruce wood by hydrothermal treatments at 120°C and
4 57–64% HRH as a function of heating duration [20].

5 *Open symbols*: hydrothermally treated; *filled symbols*: hydrothermally treated and moistened;
6 *circles*: changes in E'/ρ ; *squares*: changes in $\tan\delta$.
7
8
9

10 **Figure 8** Changes in E'/ρ and $\tan\delta$ of spruce wood at 25°C and 60% RH due to hydrothermal
11 treatment at 120°C for 1–7 days [20].

12 *Open circles*: heated at 0% HRH (heated dry); *filled circles*: heated at 29–39% HRH; *filled*
13 *triangles*: heated at 57–64% HRH; *filled squares*: heated at 77–82% HRH; *open squares*: heated at
14 89–95% HRH (steamed). The E'/ρ and $\tan\delta$ values of hydrothermally treated wood are normalized
15 by those determined in an unmodified state.
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

Figure1
[Click here to download high resolution image](#)

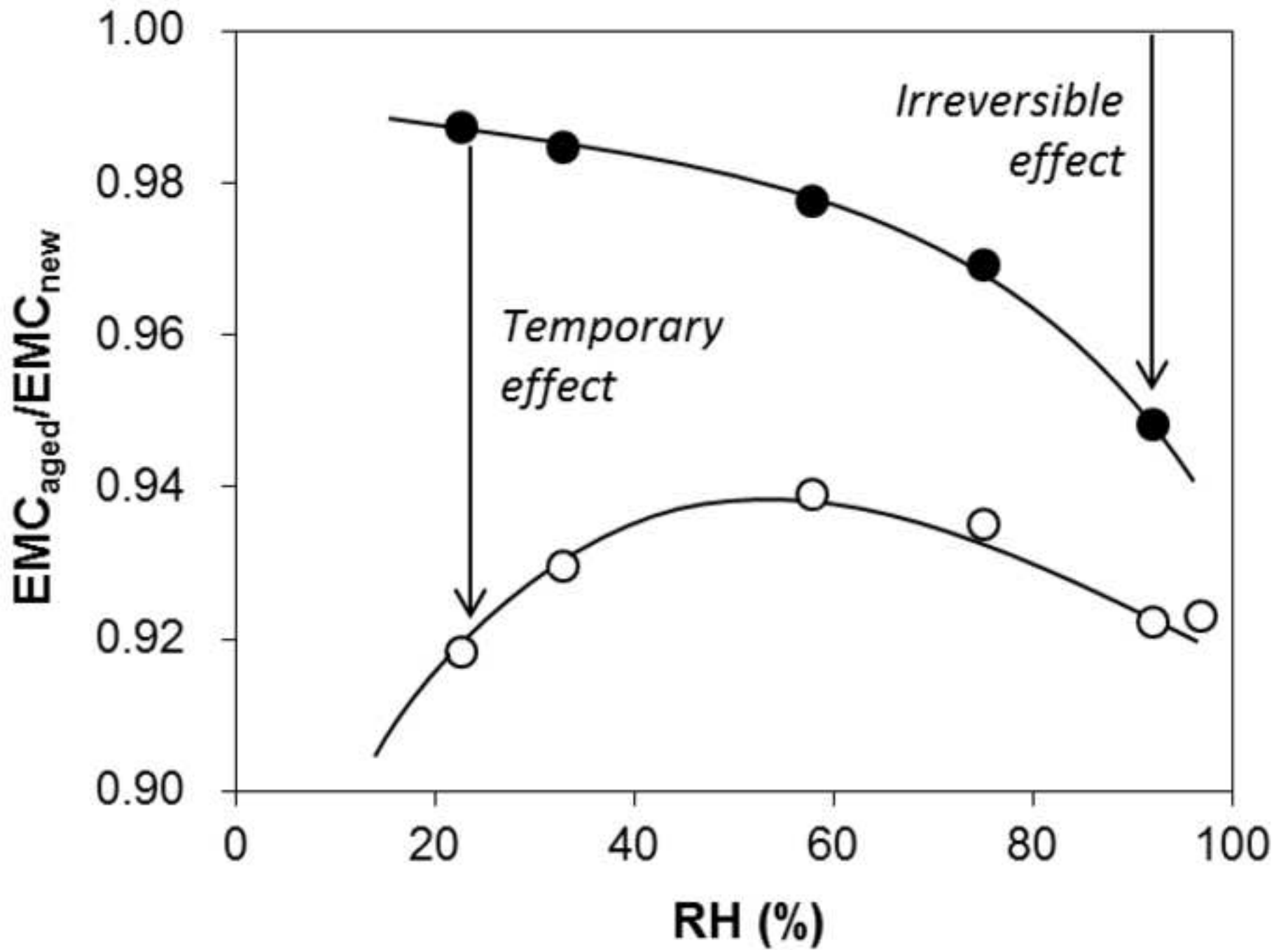


Figure2
[Click here to download high resolution image](#)

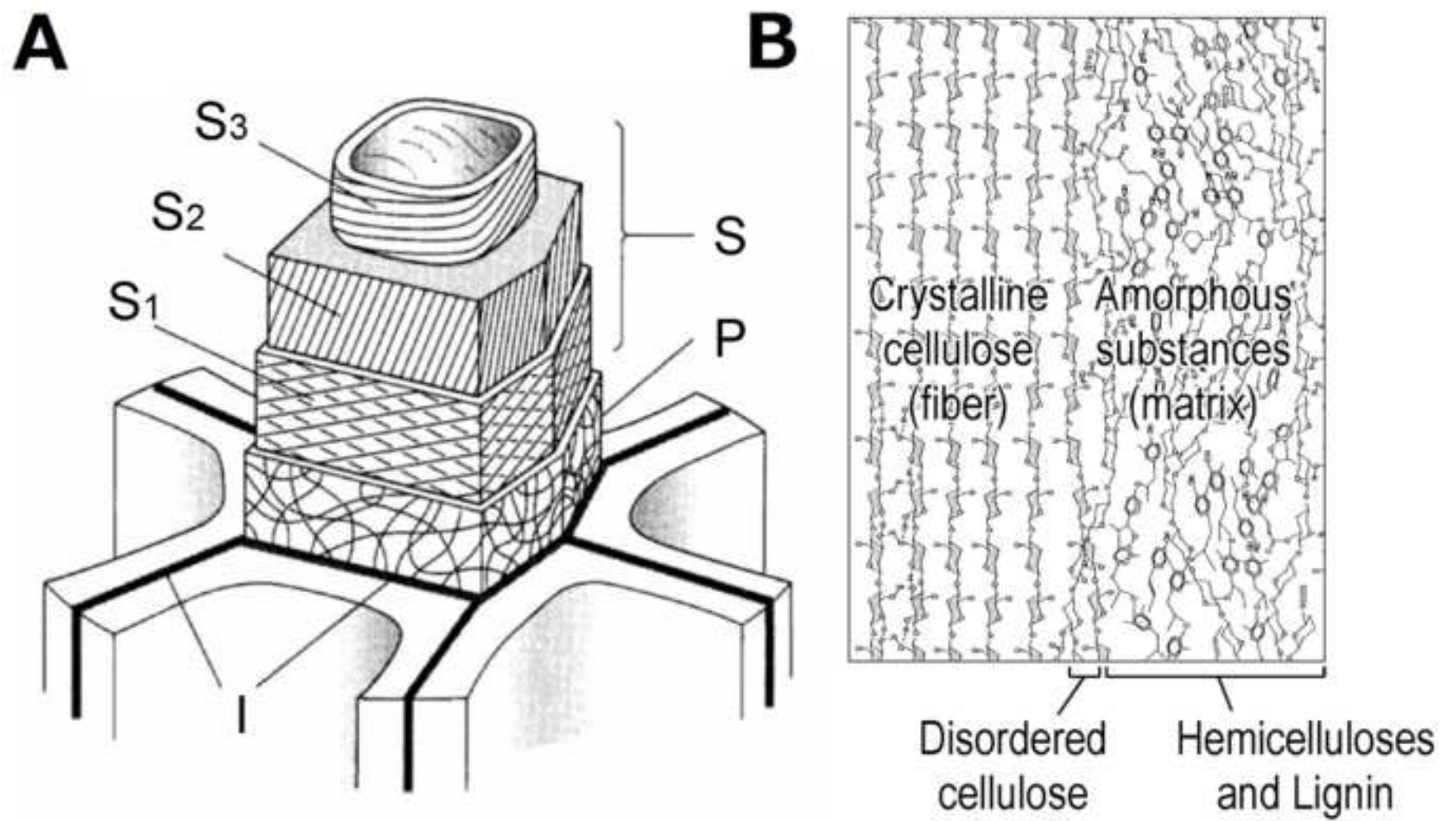


Figure3

[Click here to download high resolution image](#)

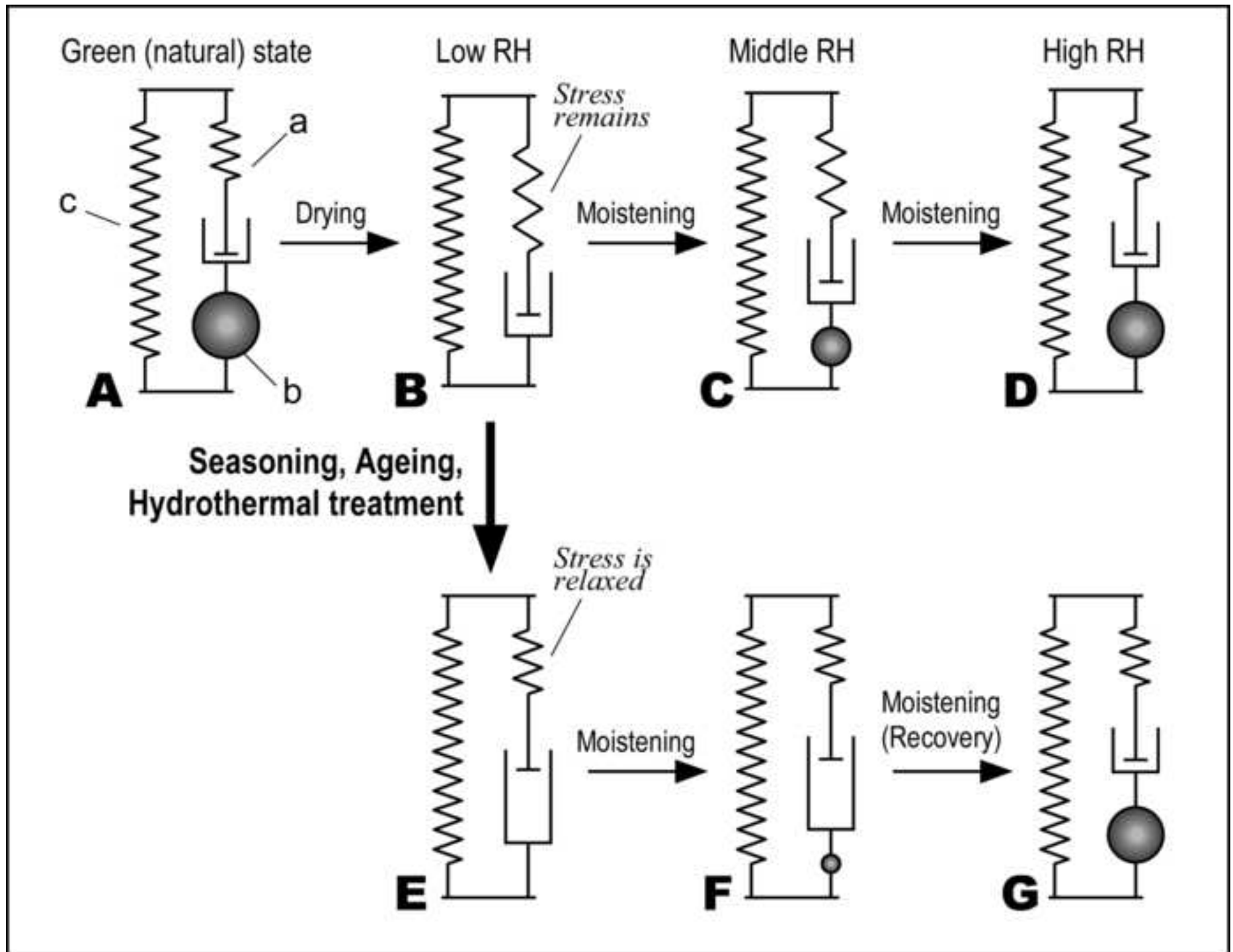


Figure4
[Click here to download high resolution image](#)

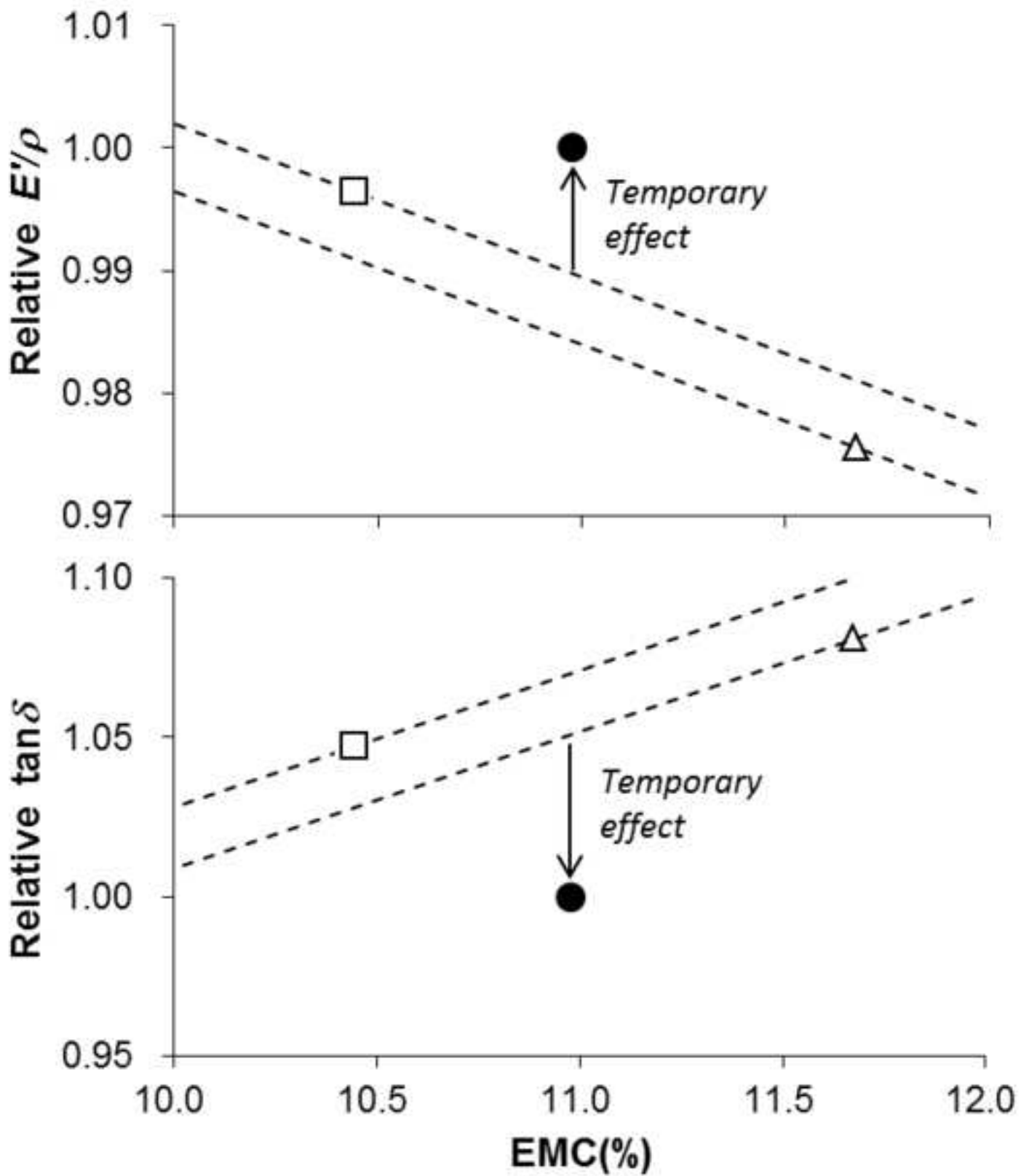


Figure5
[Click here to download high resolution image](#)

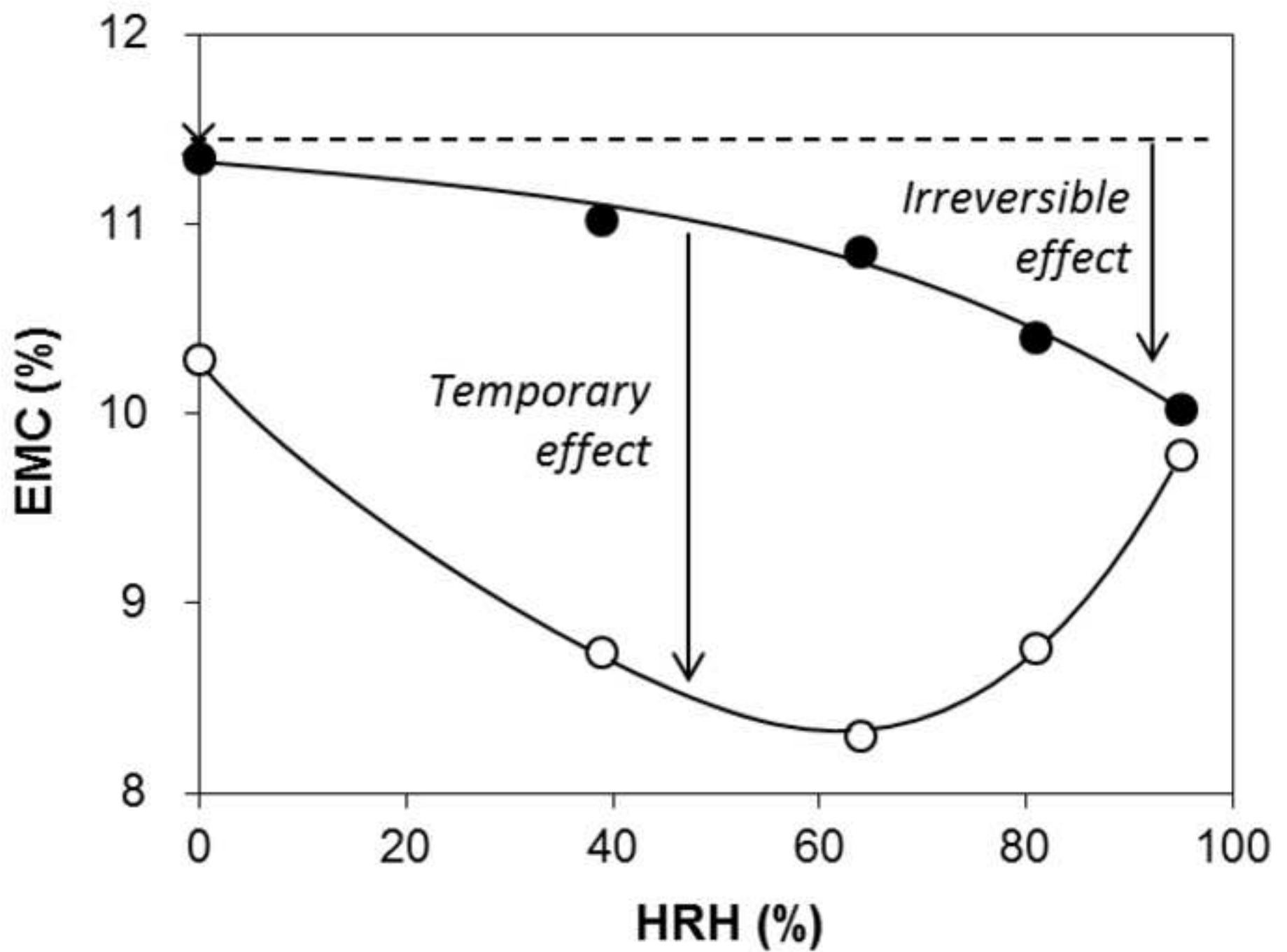


Figure6
[Click here to download high resolution image](#)

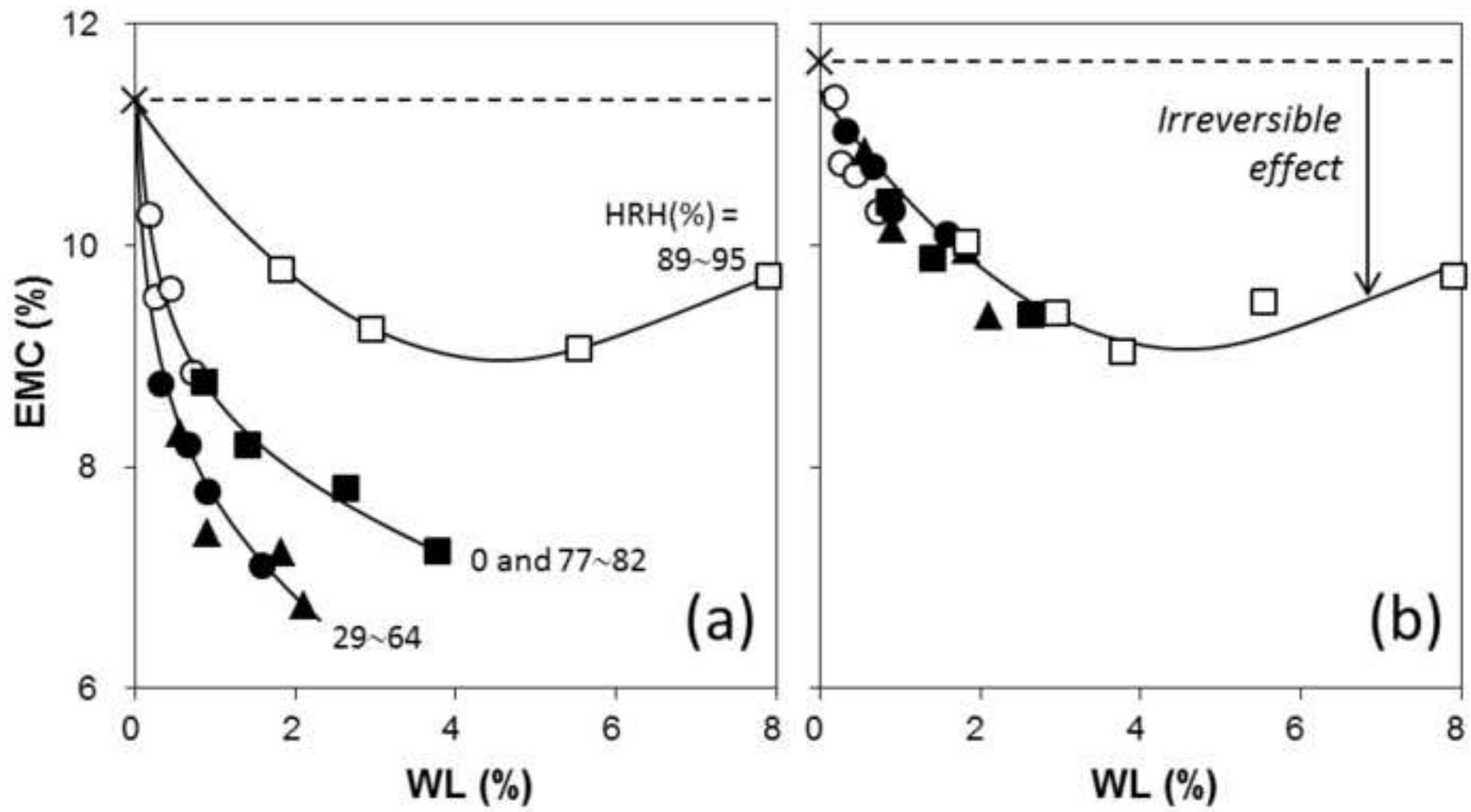


Figure7
[Click here to download high resolution image](#)

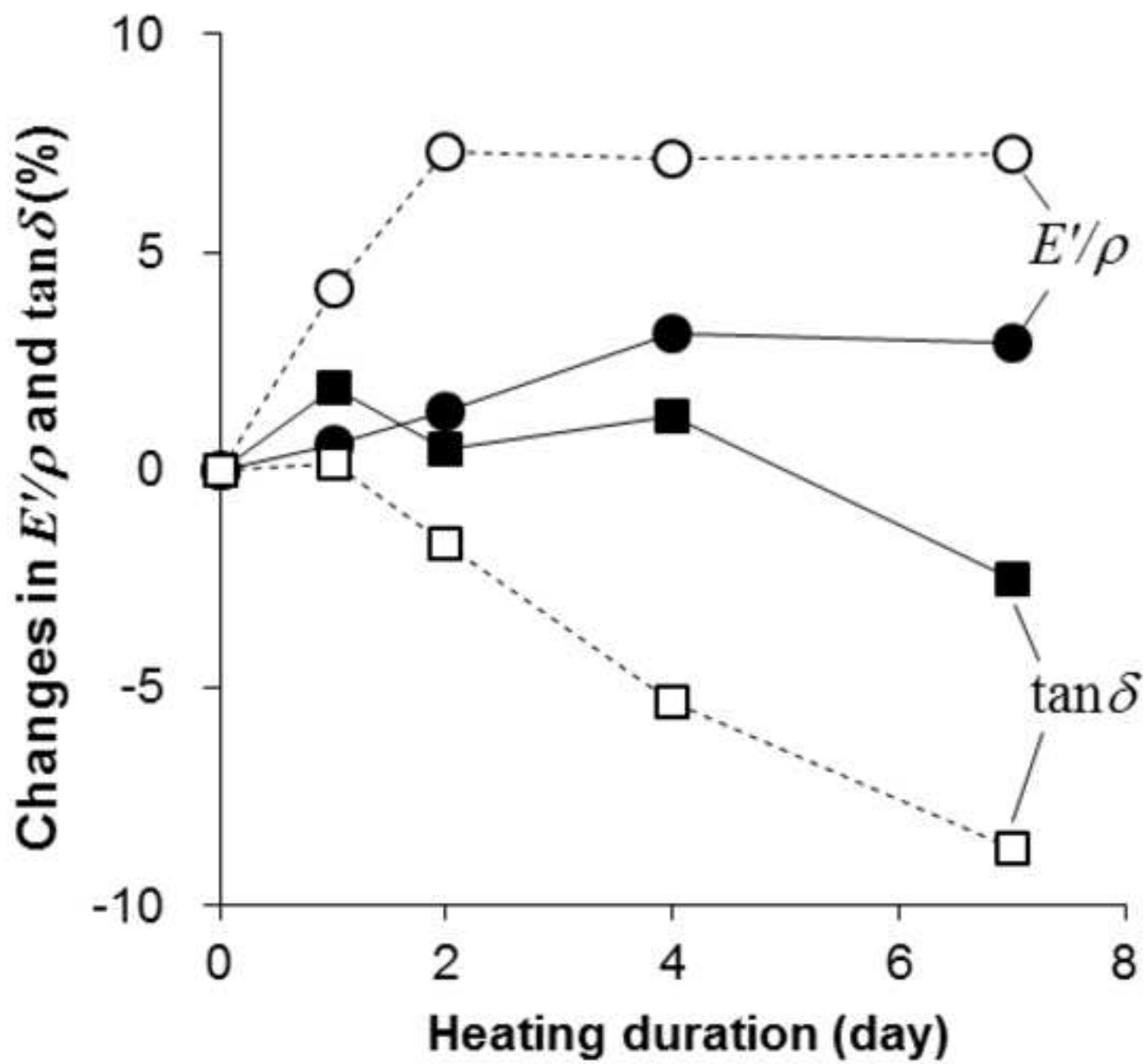
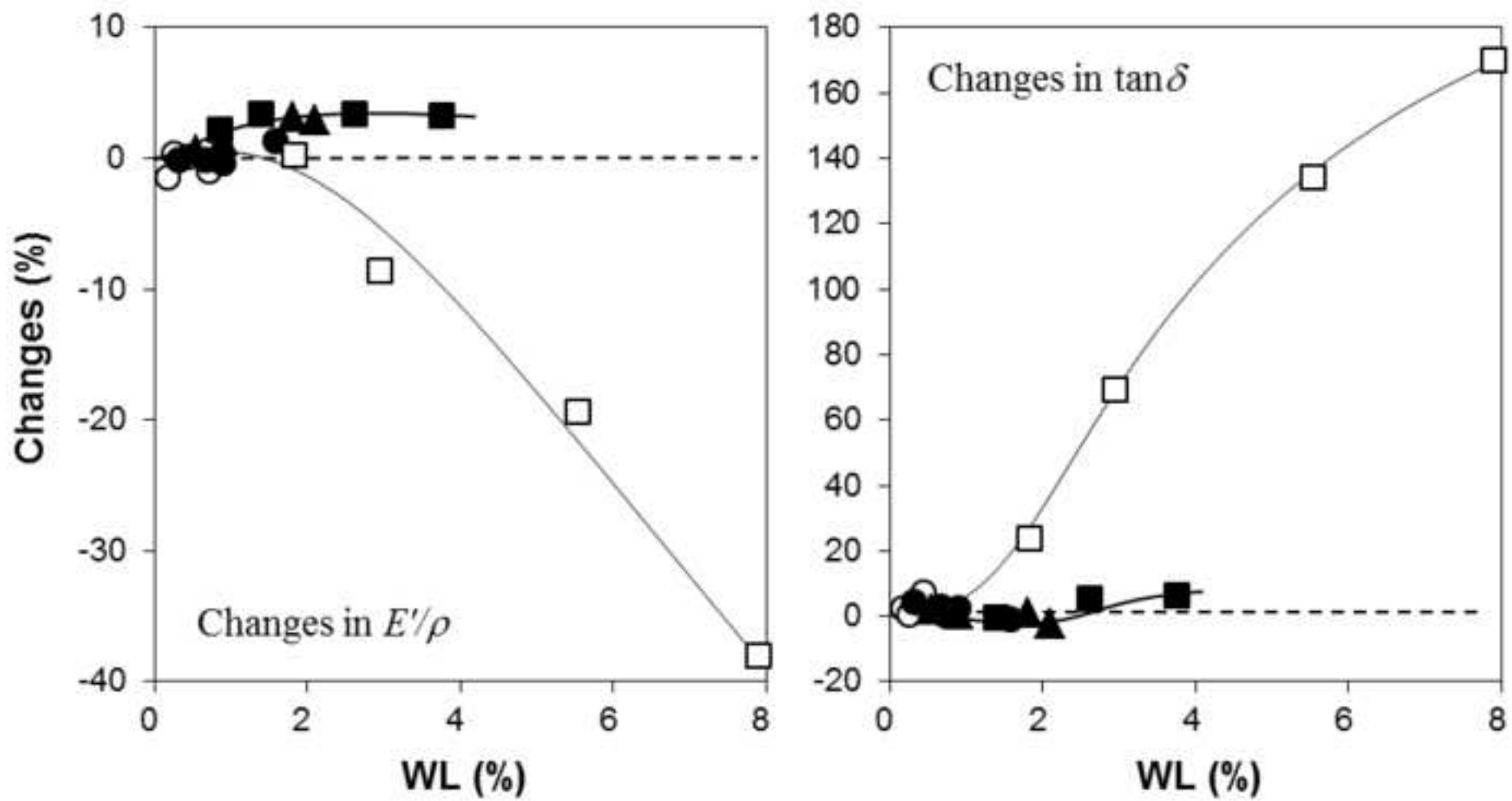


Figure8
[Click here to download high resolution image](#)



For Reviewer #1

Thank you for your suggestions. Unfortunately I could not find articles you have suggested. I understand that wood properties vary widely even in a species, but I believe that the results presented in this manuscript are sufficiently reliable respect to their reproducibility. In this manuscript, I just want to suggest that the reversible changes should be considered when we discuss the effects of ageing and hydrothermal treatment.

For Reviewer #2

Thank you for your comments. I have added some illustrations (Figures 2 and 3) for clearer understanding of wood structure and drying stress.