Lignocellulosic materials modification and engineering in relation to viscoelastic perspectives

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Abstract. By 2010, only 52% or 94 million ha of the total land area in Indonesia was forested. Therefore, we should develop forest plantation with fast growing species, or better efficiency and quality improvement for the utilization of forest products. The conventional method of fabricating curved wooden products is to cut wooden beams into a curved wood, and to connect them for the expected curve shape. However, the disadvantages of this conventional woodworking process are wasting of wood raw materials, decreasing the wood strength due to fiber cutting, and reducing the beauty of wood fiber direction. These problems can be overcome by bending solid wood, as well as curving laminated veneer lumber (LVL). The bending method of solid wood requires special technique and equipment compared to curving method of LVL. The development of commercial products is required to understand basic knowledge of wood bending. This paper deals with lignocellulosic materials modification and engineering in relation to viscoelastic perspectives; describing viscoelastic property of wood and other lignocellulosic materials, and development of furnitures and bicycle frames from LVL curves. It is expected that more modified and engineered products can be made from lignocellulosic materials based on the knowledge of their viscoelastic properties.

1. Introduction
The Indonesian archipelago of about 17,000 islands is a home to some of the most biodiverse forests in the world. In 1900, the total forest represented 84% of the total land area [1]. The forest cover by 1950 is estimated to 145 million ha of primary forest and another 14 million ha of secondary and tidal forest [2]. In the early 1970s, Indonesia used this valuable resource to its economic benefit with the development of the country's wood-processing industries. From the late 1980s to 2000, production capacity has increased nearly 700% in the pulp and paper industries, making Indonesia the world's ninth largest pulp producer and eleventh largest paper producer [1]. Between 1990 and 2000, 20% of the forest area in Indonesia had been lost (24 million ha) and by 2010, only 52% or 94 million ha of the total land area was forested [3]. Even despite a moratorium on new logging contracts imposed in 2010, the rate of deforestation continued to increase to an estimated 840,000 ha in 2012, surpassing deforestation in Brazil [4].
After decreasing of natural forest area due to deforestation, following by collapse of plywood industries, Indonesia is now facing the era of forest plantation with fast growing species which mostly for pulp and paper industries, and oil palm plantation (Figure 1). Therefore, the more efficient and improved quality of forest products utilization is crucial.

2. Efficiency and quality improvement for the utilization of forest products

2.1. Conventional method of furniture manufacturing

Curved wooden products are generally applied to furniture products, as well as to residential building components (door frames/windows), musical and sport instruments, toys, and other necessities. The conventional method of obtaining curved wooden products is shaped by cutting the wooden beams into a curved wood, and connecting them to obtain the expected curve shape (Figure 2). Based on the process, this conventional method is easy to work, because it only uses simple wooden techniques and tools. However, the deficiency and disadvantages of this conventional woodworking process are wasting of wood raw materials, decreasing the wood strength due to fiber cutting, and reducing the beauty of wood fiber direction.

Figure 1. Decreasing of forest area in Indonesia.

Figure 2. Curved wooden products by conventional method.
2.2. Research and development on wood bending and LVL curves

These problems can be overcome by solid wood bending (Figure 3), as well as Laminated Veneer Lumber (LVL) curving (Figure 4) methods. Bending method of solid wood requires special technique and equipment compared to curving method of LVL, so that if it is to be developed commercially, it needs basic knowledge of wood bending.

Research Center for Biomaterials, Indonesian Institute of Sciences has been conducting research and development on wood bending technology since 2004 [5–8], and continuing with LVL [9–10]. The wood bending mechanism is almost the same as radial compression of wood [11], however the inner part of curve is compressed in longitudinal direction. Therefore, it can be done through its viscoelastic processes [12–14], i.e. (I) softening, it can be achieved by soaking the wood until water saturated and proceed with heating treatment; (II) forming, it may be possible when the wood is still soft when compared to dry wood; (III) setting, drying set is a drying process when wood is in a deformed condition; (IV) fixation, it is a particular effort/treatment for the wood that has been deformed does not return to its original shape or permanent.

![Figure 3. A hot press machine for the manufacturing of curved solid woods.](image)

![Figure 4. A simple clamp for the manufacture of curved LVL.](image)
3. Viscoelastic property of wood and other lignocellulosic materials

3.1. Theoretical definition of viscoelastic materials

To bend wood or other lignocellulosic materials, their viscoelastic property should be known. Viscoelasticity is a property of materials that exhibit both viscous and elastic characteristics when they are undergoing deformation. Viscous materials resist shear flow and strain linearly with time when a stress is applied. Elastic materials strain when stretched, and immediately return to their original state once the stress is removed. Viscoelastic materials have elements of both of these properties, and thus exhibit time-dependent strain. Whereas elasticity is usually the result of bond stretching along crystallographic planes in an ordered solid, viscosity is the result of the diffusion of atoms or molecules inside an amorphous material [15].

Depending on the change of strain rate versus stress inside a material, the viscosity can be categorized as having a linear, non-linear, or plastic response. When a material exhibits a linear response, it is categorized as a Newtonian fluid. In this case, the stress is linearly proportional to the strain rate. If the material exhibits a non-linear response to the strain rate, it is categorized as Non-Newtonian fluid. In addition, when the stress is independent of this strain rate, the material exhibits plastic deformation [15]. Many viscoelastic materials exhibit rubber like behavior that is explained by the thermodynamic theory of polymer elasticity. Some examples of viscoelastic materials include amorphous polymers, semicrystalline polymers, biopolymers, and metals at very high temperatures. Cracking occurs when the strain is applied quickly and outside of the elastic limit.

Unlike purely elastic substances, a viscoelastic substance has an elastic component and a viscous component. The viscosity of a viscoelastic substance gives the substance a strain rate dependence on time. Purely elastic materials do not dissipate energy (heat) when a load is applied, then removed. However, a viscoelastic substance loses energy when a load is applied, then removed. Hysteresis is observed in the stress–strain curve, with the area of the loop being equal to the energy lost during the loading cycle. Since viscosity is the resistance to thermally activated plastic deformation, a viscous material will lose energy through a loading cycle. Plastic deformation results in lost energy, which is uncharacteristic of a purely elastic material’s reaction to a loading cycle [15].

Specifically, viscoelasticity is a molecular rearrangement. When a stress is applied to a viscoelastic material such as a polymer, parts of the long polymer chain change positions. This movement or rearrangement is called creep under a constant stress. Polymers remain a solid material even when these parts of their chains are rearranging in order to accompany the stress, and as this occurs, it creates a back stress in the material. When the back stress has the same magnitude as the applied stress, the material no longer creeps. When the original stress is taken away, the accumulated back stresses will cause the polymer to return to its original form. The material creeps, which gives the prefix visco-, and the material fully recovers, which gives the suffix –elasticity [16].

The secondary bonds of a polymer constantly break and reform due to thermal motion. Application of a stress favors some conformations over others, so the molecules of the polymer will gradually "flow" into the favored conformations over time [17]. Because thermal motion is one factor contributing to the deformation of polymers, viscoelastic properties change with increasing or decreasing temperature. In most cases, the creep modulus, defined as the ratio of applied stress to the time-dependent strain, decreases with increasing temperature. Generally speaking, an increase in temperature correlates to a logarithmic decrease in the time required to impart equal strain under a constant stress. In other words, it takes less work to stretch a viscoelastic material an equal distance at a higher temperature than it does at a lower temperature. Both wood and cellulose materials are involved in such materials and the viscoelasticity which causes a variety of mechanical properties is closely related to thermal motions of a polymer chain of the material.
3.2. Bending strength of wood in softening condition
Viscoelastic property of wood can be simply measured by static bending test. Mechanical property changes due to the moisture content (MC) and/or temperature changes have been examined for 15 Indonesian wood species [18]. The densities ranged from 0.27 g/cm$^3$ for randu alas to 0.81 g/cm$^3$ for lamtoro wood.

![Figure 5. Static bending test in a water bath.](image)

The test was conducted at 20°C and 65% RH using a conventional testing instrument with a constant cross-head speed (5 mm/min), loading at a three-point bending system with a span of 80 mm. In the water-saturated state, the load was applied to the specimen standing in a water bath controlled at 20°C and 80°C (Figure 5).

![Figure 6. MOR of woods in softening conditions.](image)

Figure 6 shows modulus of rupture (MOR) plotted against air-dry density. The MOR increased linearly with air-dry density, regardless of wood species. The wet 20°C/air-dry ratios for the slopes in regression lines was 0.67, while wet 80°C/air-dry ratios was 0.27 [18]. Iida [19] has reported that the wet 20°C/air-dry ratios was 0.62, and wet 80°C/air-dry ratios was 0.29 for Japanese wood species. The influences of MC change on MOR were somewhat smaller for the 15 Indonesian wood species than for the Japanese wood species. On the other hand, the influences of combined MC and temperature changes to MOR did not differ between Indonesian and Japanese wood species.
3.3. Relaxation modulus of wood in softening conditions

Figures 7 shows the relative relaxation modulus values of kecapi wood at air-dry 20°C, wet 20°C and wet 80°C. Relative relaxation modulus (ratio of relaxation modulus after treatment divided by initial relaxation modulus) decreased with an increase in the MC and temperature. The value of relative relaxation modulus in wet 20°C/air-dry ratios was decreased to around 0.8. However, it was affected still more by both MC and temperature, the wet 80°C/air-dry ratios of relaxation modulus decreased to around 0.6. It was concluded that the hygro-thermal properties of lignin significantly affect the changes of elastic and strength properties of wood that occur due to hygro-thermal effects.

![Figure 7. Relative relaxation modulus of kecapi wood in softening conditions.](image)

3.4. Viscoelastic perspectives of other lignocellulosic materials

Furthermore, the softening behaviour and viscoelastic perspectives of other lignocellulosic materials, such as bamboo, rattan, and oil palm trunk was investigated. The specimens were tested at static bending in dry and wet conditions.

Figure 8 shows the decreasing of MOR of andong bamboo, sengon wood, air rattan and oil palm trunk from dry to wet condition. MOR of bamboo decreased drastically \((y = -675.5x + 1984)\), followed by wood \((y = -308.5x + 875.1)\), rattan \((y = -195.9x + 608.2)\) and oil palm trunk \((y = -65.19x + 386.7)\). It was suggested that besides the softening behaviour of their chemical components, bendability of lignocellulosic materials was depended on their anatomical structures. From the observations, the changes of anatomical structures occurred mostly in the inner parts of bamboo cell walls. This was due to the compression of the inner parts of bamboo in both longitudinal and radial directions, while in the middle parts and near the bark was remained unchanged. Therefore, it was concluded that bamboo could be more easily bended, because there were many parenchyma cells with thin-walled cells in the inner parts and more easily softened.
Figure 8. MOR of other lignocellulosic materials in softening conditions.

Wood and other lignocellulosic materials are composed of highly elongated cells whose walls have a complex multi-layered structure. In each layer, cellulose molecules are grouped together in long filaments called microfibrils embedded in a matrix composed of amorphous hemicelluloses and lignin. Both moisture and temperature act differently on the matrix and the microfibrils. Because of the crystalline nature of cellulose, water molecules cannot enter into the microfibrils. Bound water is held in the matrix and at the interface between the matrix and the microfibrils, and works as both a swelling agent and a plasticizer [20].

The elevation of temperature in wet condition softens the matrix, and its two main constituents, hemicelluloses and lignin, shift from the glassy state to something near the rubbery state. On the other hand, the microfibrils remain in the glassy state and are almost unaffected by moisture or heat. The softened wood and other lignocellulosic materials can be deformed easily. When a load is applied to the wood and other lignocellulosic materials, most of them are supported by the microfibrils at the local level. Softening of the matrix enables the relative displacement of the microfibrils so that their whole framework deforms elastically to adjust to the local loading. As lignin is a slightly cross-linked amorphous polymer, its deformation should be viewed as an increased viscoelastic strain rather than plastic flow [20].

When a deformed wet specimen is dried under restraint, the stress gradually decreases until it has disappeared. The departure of water molecules due to drying induces the reformation of hydrogen bonds between the molecules of the matrix. With the temperature decrease, this process leads to return the glassy state, where the elastic deformations of the microfibrils and the matrix are frozen. The deformation is apparently fixed in the deformed state (drying set). However, as soon as the matrix is softened again through re-moistening and heating, most of the deformation is recovered due to the liberation of energy-elastic strain stored in the microfibrils and entropy-elastic molecular movements in the matrix [20].

It is well known that many amorphous polymers change from glassy state to rubbery state and are capable of plastic deformation (flow) in the case of linear polymers when heated above their glass transition temperature (or softening temperature). It was reported that the softening temperature of hemicelluloses and lignin of wood in wet condition were about −40°C and 60°C~80°C, respectively [20]. Above the softening temperature of the matrix, wood can be easily deformed to more than a strain of 50%. In this process, the stress remarkably decreases with time due to the micro-Brownian motions of molecules of the matrix. It is well known that the decrease of stress is accelerated by the departure of moisture during drying. Plastic deformation (flow) does not occur, because the matrix has a cross-linked structure.
Actually, most of the deformation was recovered by boiling after measurement. It is considered that the recovery of deformation was occurred by the liberation of energy-elastic strains stored in the micro fibrils and the entropy-elastic molecular movements in the matrix by boiling. This fact proved that the permanent fixation did not result from flow of the matrix. Previous researches on the mechanism of permanent fixation of radial compressive deformation of wood by heat or steam treatment could be found in [21–25].

4. Latest findings and future research
4.1. Development of furnitures with curved LVL
Research activities that have been conducted for years need to be applied and promoted to the users. In 2015-2017 through funding of Science and Techno Park (STP) Program, Theoretical and Technical Training, as well as Technology Transfer for Wood and LVL Bending Technique has been carried out to Small Wooden Furniture Industries at Sumedang District, West Java.

The product that can be easily developed is LVL curve, because it does not require a hot press machine. Solid wood bending is more difficult to apply and require substantial investment, as it requires a press machine equipped with heating components, and can only be made with a certain curved radius (Figure 3), when compared to LVL curve. The LVL curve requires only clamp tools made with a radius as expected and can be glued without a hot press (Figure 9). A small company, CV. Putra Lingga Jaya at Sumedang West Java has already produced these furnitures through License Cooperative Agreement.

Figure 9. Furnitures from curved LVL.

4.2. Development of bicycles with lignocellulosic material frames
From the above experiences, the technology of LVL curving has a potential to be developed as a new innovation of bicycle frame components [26]. Bicycle frame of LVL curve was previously designed in accordance with the expectation. From the design, a clamp was then made as a tool to form the bicycle frame.

Bicycle frames of LVL curve have been made from 2 mm thickness of rubber and teak wood veneers to form 20 mm thickness (10 layers). The wood veneers were wrapped around a clamp and glued with 300 gr/m² weight natural rubber latex and cured under room temperature conditions for 24 hours before they are formed into a bicycle frame, more clamps were used especially on the extreme curves. The process of making bicycle frame is shown in Figure 10.
Since 2015, we have succeeded in making bicycle frames from LVL curve with various types and promoting them to some exhibitions. Two prototypes of fixie bikes were made with and without connections on their frames. Mountain bike types were made for both male and female, for which the upper part of female’s frame was lower than male’s frame.

It is expecting that the bicycles with LVL curve frame become an environmentally friendly vehicle for free-car areas, such as Botanical Gardens in the near future. By funding from STP Program, mass production process of the products will be developed through License Cooperative Agreement with a private company. In the near future, solid woods and other lignocellulosic materials, such as bamboo, rattan and oil palm trunk would also be developed for the bicycle frames.

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