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STIFFNESS OF A COMPOSITE MATERIAL MANUFACTURED WITH TANNIN'S BINDER OF AUTRANELLA CONGOLENSIS

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ABSTRACT: In last decades, the need for implementation of biodegradable materials is a showy concern for the planet earth because they contribute to the fight against environmental pollution and the valorization of plant resources. For this purpose, a composite developed with the tannin's binder and reinforcement of autranelle congolensis has been established. Throughout this work, two types of materials were manufactured under the same experimental. The implementation of biodegradable materials made with tannin's binder of autranelle congolensis (type1 materials) and synthetic materials based on Urea Formaldehyde (type2 materials). At the end of testing, it appears that type 1 materials are stiffer than type 2 materials: Indeed, the three-point bending tests performed on sample of type 1 materials when sizing rate varies between 25 and 78%, gave a Young's modulus varies between 43.832 and 466.652 MPa against 24.200 to 266.001 MPa for type 2 materials. A volume effect on the stiffness has established on the two types of materials: stiffness is increasing with the length between supports. For both materials, Young's modulus increases with the length between supports. The stiffness increases with temperature and the sizing rate. Materials having a high sizing rate are less resistant to moisture. The type 2 materials are resistant to moisture than type 1 materials. The binder developed with the tannin of autranelle congolensis has a gel time between 50 and 152 seconds.

KEYWORDS: biodegradable; urea formaldehyde; sizing rate; volume effect; absorption rate.

1 INTRODUCTION AND OBJECTIVES

Since the beginning, nature has always been generous to humans. It made available to them, fossil resources, oil resources, plant and animal very helpful. These elements constitute for them a wealth because they are used in the timber industries, aeronautics, bituminizing, decoration, tanning...**[5].** At the time when humanity suffers from environmental threats, minds are mobilizing to deal with this scourge, the biodegradables materials manufactured using natural resources are an alternative.

From eighteenth century to the twentieth century, a long period during which petroleum was king, materials were manufactured by using a synthetic matrix as UF, PF, MF… **[1].** to fully exploit these naturals wealth, it is important for us to manufacture materials based with the tannin's binder from tropical plants and more particularly based of wood used in Cameroon: Autranelle congolensis, species most used in plating. It is in this perspective that we want to: Develop the tannin's binder; manufacture the materials based tannin's binder; manufacture materials based Urea Formaldehyde; dry the materials by varying the temperature; make bending tests "three points"; calculate stiffness; compare the two materials.

The main objective of this work is to consider the replacement of urea formaldehyde resins by natural binder developed with tannin while comparing the two types of materials (materials made with tannin's binder and those based on urea formaldehyde); consider the manufacture of biodegradable particle board based tannin's binder of autranelle congolensis and finally contribute to the reduction of environmental pollution.

To achieve this, we are going to review materials and methods used, give results and interpreted them.

2 MATERIALS AND METHODS

The bark of autranelle congolensis (plant known in Cameroon on the name mukulungu) used for the development of binder and reinforcements of materials come from the company ECAM- PLATING, a company located at Mbalmayo - Cameroon. Tests have taken place in the Natural Substance Laboratory $N°3$ of the University of Yaoundé I and in Laboratory of Mechanics, Materials and Structures of the National Advanced School of Engineering, University of Yaoundé I, and Cameroon.

2.1 MATERIALS

2.1.1 BINDER'S DEVELOPMENT

Materials that have allowed us to develop the binder after extraction of tannin **[2]** are:

- Tannin: bark's extract of autranelle congolensis;
- Cassava flour: load in the development of the adhesive;
- KOH: an additive making the middle basic while changing the PH;
- Hexamine: additive acting as hardener;
- A boiler water bath;
- A kite balloon, containing glue;
- The beakers containing tannin;
- A spatula to stir the glue;
- Test tube and beaker for determination of gel time;
- An electronic balance for measuring the constituents used in the development...

2.1.2 MANUFACTURING OF MATERIALS

The materials used for the manufacture of composite materials and bending tests three points are:

- A mold $150 \times 15 \times 15\ mm$ to produce samples of materials;
- Binder's tannin used in the manufacture of type 1 materials;
- Urea formaldehyde: resin use to produce type 2 materials;
- An oven for drying the material depending temperature ;
- Lead compactness materials;
- Three-point bending tests machines to calculate the Young's modulus of different samples produced:
- The weights;
- A dial gauge for reading travel.

2.2 METHODS

The extraction of tannin **[3]** leads us to the development of binder's tannin, manufacturing type 1 and 2 materials, and "three-point" bending tests. After that test, Young's modulus and other values such as mechanical tensile strength, slenderness's effect, heat resistance were determined and these mechanical characteristics of type 1 and type 2 materials were compared. The specimens are reinforced by particles of autranelle congolensis (wood).

2.2.1 TANNIN'S BINDER' DEVELOPMENT

Prepare tannin in water solution $N°1$ at 50% concentration and cassava flour in water solution $N°2$ at 30% on the tannin solids base then mix solution $N°1$ and solution $N°2$.The pH was adjusted to 10 with a potassium hydroxide (30%)**.** Finally add 5% of hexamine solution (30%) on the tannin solids based **[2].**

2.2.2 MANUFACTURING OF COMPOSITE MATERIALS

Two types of materials were manufactured: the biodegradable materials manufactured with tannin's binder (type 1 materials) and materials manufactured with Urea Formaldehyde's resin (type 2 materials).
The manufacturing of materials is summarized **[4]** as below:

- Crushing: This is an operation which consists to reduce wood in bark particles;
- sifting: Removal large and dust particles because they would give defectives composites.
- sizing: quantity of binder used is between 25 and 78 % of the total composite weight. These resins are shuffled directly with the wood particles before molding;
- mixing : mixing wood particles and the binder to form "cake" or "dough";
- casting : dough was poured into a mold of $150 \times 15 \times 15$ mm;
- pressing: it takes place under significant pressure cold ($20\ KN/_{mm^2}$);
- Heating : it is made by variation of the temperature ($60\ to\ 120°\ C$);
- Removal from the mould: it is carried out after evaporation of certain water in the materials.

2.2.3 DRYING MATERIALS

The heating of the materials at a T temperature ($80\ to\ 90°C$) allows the evaporation of water contained in the materials.

2.2.4 BENDING TESTS

The principle of the "3-point" bending test **[5]** is to determine the maximum tensile strength of a material on two supports with an application of the force at the midway between the supports at a constant speed over a test tube. By using the device below, put a weight on a pan suspended at the specimen and wait until the comparator stabilizes to read the deflection on its dial. The operation is repeated until reaching a value m to the rupture or the appearance of cracks on the specimen.

Fig.1: testing machine "3 point" bending

2.2.5 RIGIDITY

Young's modulus were determined and other values such as the mechanical tensile strength, the absorption rate and the resistance to humidity.

2.2.5.1. YOUNG'S MODULUS

There are links between the two variables (y and F), the geometry of specimen (L, h and b) and the characteristics of the material (σ, ε and E).

The specimen of rectangular shape is placed on two supports and loaded single in the middle by a load until failure.

The Young's modules E_i are determined by:

$$E_i = \frac{1}{4b}\left(\frac{L}{h}\right)^3 \frac{\Delta F}{\Delta f} \qquad (1)$$

And the stiffness $\boldsymbol{K}$ is deducted by:

$$K = \frac{EA}{L} \qquad (2)$$

L: Distance between two supports;

B: width of the sample ($15\ mm$)

H: sample thickness (15 mm).

The average Young's modulus $\overline{\boldsymbol{E}}_{\boldsymbol{m}}$ of each sample is given by:

$$\overline{\boldsymbol{E}}_{\boldsymbol{m}} = \frac{\sum_i^n E_i}{n} \qquad (3)$$

$i = 1\ to\ n;\ n = 35$ samples per test

The plot of a histogram makes it possible to study a distributive modules based the sizing the rate and temperature.

2.2.5.2. SQUARE ERROR

The values of square error are derived by the formula:

$$\boldsymbol{\sigma_m} = \sqrt{\frac{\sum_{i=1}^n (\boldsymbol{E_i} - \overline{\boldsymbol{E}}_{\boldsymbol{m}})^{\,2}}{\boldsymbol{n}}} \qquad (4)$$

2.2.5.3. Tensile strength

The bending stress is expressed by the following classical equation [2]:

$$\boldsymbol{\sigma} = \frac{\boldsymbol{3}}{\boldsymbol{2}} \frac{\boldsymbol{FL}}{\boldsymbol{BH^2}} \qquad (5)$$

L: distance between supports; F: applied load;

B: width of the specimen; H: thickness of the specimen.

At the break: $$\sigma_{\max} = \frac{3}{2}\frac{F_{\max}L}{BH^2} \qquad (6)$$

2.2.5.4. EFFECT OF SLENDERNESS

To know the effect of volume on the tensile strength, the three points bending tests were performed with different lengths between the supports ($120, 90, 60$ and $30\ mm$).

2.2.5.5. MOISTURE RESISTANCE

It is determined by the $NF\ B\ 51\ 262$ **[6]**.It describes " $V100$ " method which consists of immersing the specimens of $50x50\ mm$ in boiling water for at least one hour. The samples are left to cool in cold water for 1 to 2 hours, removed from the water, free of excess water by sponging wet and then tested.

2.2.5.6. WATER ABSORPTION

The rate water absorption (TA) in each specimen ($50\ X\ 50\ mm$) is determined by N$\boldsymbol{FEN}$ $\mathbf{317}$ [$\mathbf{7}$] :

$$TA = \frac{m_f - m_i}{m_i} \times 100 \qquad (7)$$

m_f: Final mass material after immersion in water;

m_i: Initial mass of the material

A relationship between water absorption and the razing rate in materials was determined.

2.2.5.7. AVERAGE'S COMPARISONS

Population means (E_1, E_2, E_3, E_4, E_5) were compared by applying the "test of Z " or " reduced gap " is to compare the parameters by testing their difference [$\mathbf{8}$] (in this case compares the average of Young's moduli of the samples) . These comparisons have allowed us to retain the different populations (E_1,E_2,E_3, E_4,E_5).

To do so it:

- Take two samples from two different populations i and j;
- Consider the averages of two populations i and j , and calculate their variance : $S^2{}_i\ et\ \ S^2{}_j$
- calculate $Z = \frac{|\bar{E}_i - \bar{E}_j|}{\sqrt{\frac{S^2{}_i}{n_i}+\frac{S^2{}_j}{n_j}}}$ (8);
- If $Z\ <\ 1.96$, the difference between the parameters is not significant and it is concluded that the average population denoted $E_i$is not significantly different from the average population denoted E_j;
- If $Z\ >\ 1.96$, the difference between the parameters is significant, $E_i$is different from E_j.

3 RESULTS AND DISCUSSION

3.1 RESULTS

3.1.1 TANNIN'S BINDER DEVELOPMENT

The adhesive obtained is black, viscous and gel time is less than 200 seconds; it binds all wood materials and plastics; for good adhesion, apply high pressure for a few seconds **[5].**

3.1.2 MANUFACTURING OF COMPOSITE MATERIALS

330 Specimens of size $150 \times 150 \times 15\,mm$ were manufactured: 165 of type 1 materials and 165 of type 2 materials . The sizing rate in each case allows us to combine these samples: 5 peoples at 35 specimens per people [$2\ \times\ (5\ \times\ 35)\ =\ 330$].

Sizing rate is defined as (table 1):

Table 1:proportion of composite material's constituents

Sizing rate	Renforcement rate	Populations
25	75	E1
33	67	E2
50	50	E3
65	35	E4
78	22	E5

3.1.3 STIFFNESS

Young's modulus, Tensile strength, volume effect

Table 2: Summary of mechanical properties

Binder	Caractéristiques	E5	E4	E3	E2	E1
Tannin's binder of autrenelle congolensis	σ_{max} (MPa)	0,777	0,977	1,222	1,355	1,533
	F_{max} (N)	350	440	550	610	690
	$\overline{E}_m$ (Mpa)	466,652	266,63	162,922	82,901	43,832
	$Lmin(mm)$	30	30	30	30	30
	$A\ (mm2)$	450				
	$K\ (N/mm)$	6999	3999	2443	1243	657
Formaldehyde urea	σ_{max} (MPa)	0,666	0,777	0,977	1,222	1,355
	F_{max} (N)	300	350	440	550	610
	$\bar{E}_m$ (MPa)	266,001	194,604	77,100	45,134	24,200
	L_{min} (mm)	30	30	30	30	30
	A	450				
	K	3990	2919	1156	667	363

The study of stiffness depending the sizing rate gives:

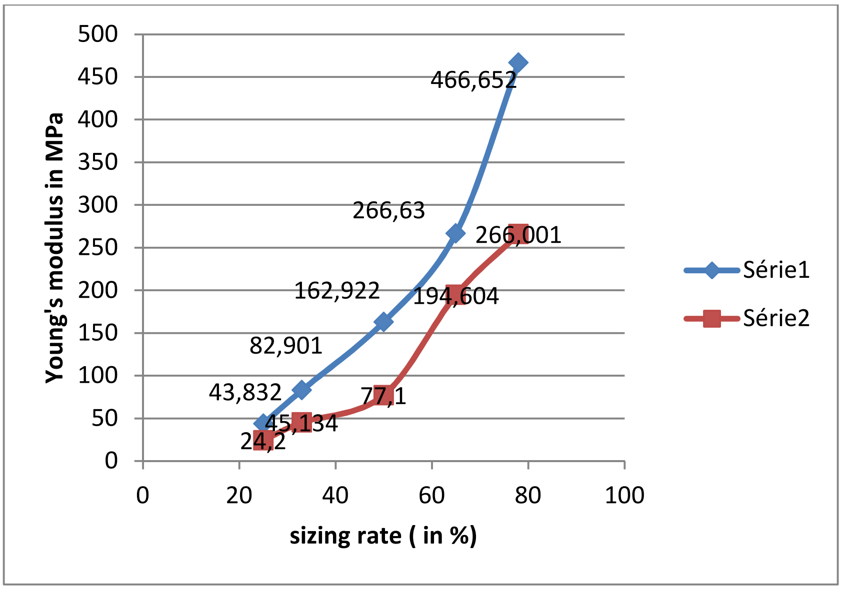

Fig. 2: Young's modulus depending of the sizing rate

On the figure above, the Serie 1 give the evolution of the rigidity of autranelle congolensis depending on the sizing rate and series 2 give that of type 2 materials.

The Young's modulus increases with the sizing rate. The different populations are manufactured at different temperatures and different sizing rate.

By varying the length L between the supports at a given temperature, we have:

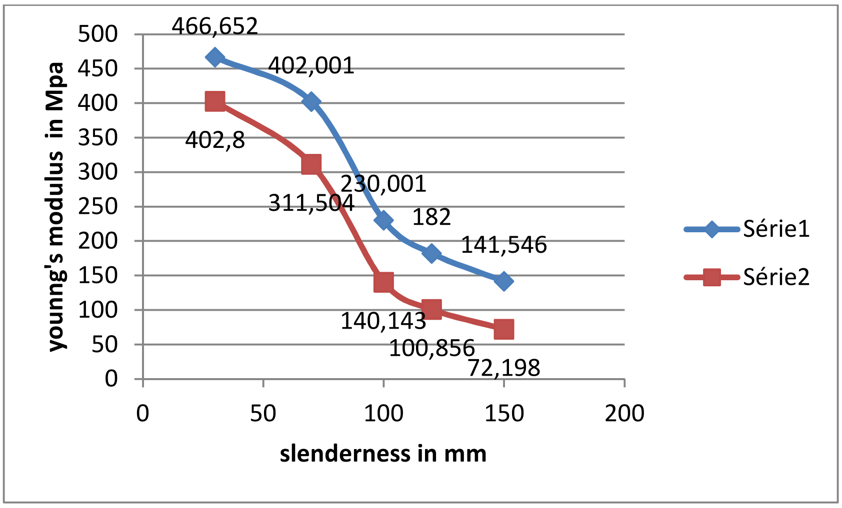

Fig. 3: Young's modulus depending the effect of slenderness

The graph above shows the evolution of the Young's modulus depending the effect of slenderness. Series 1 shows the plot of Young's modulus of type 1 materials depending the slenderness and the series 2 that type 2 materials .In each case, the Young's modulus decreases progressively as

the length between the supports is high. For a minimal length ($L = 30\ mm$), the rigidity is the highest.

The histogram below shows the evolution of the modulus depending the temperature in each case of studied materials, for length $L = 30\ mm$:

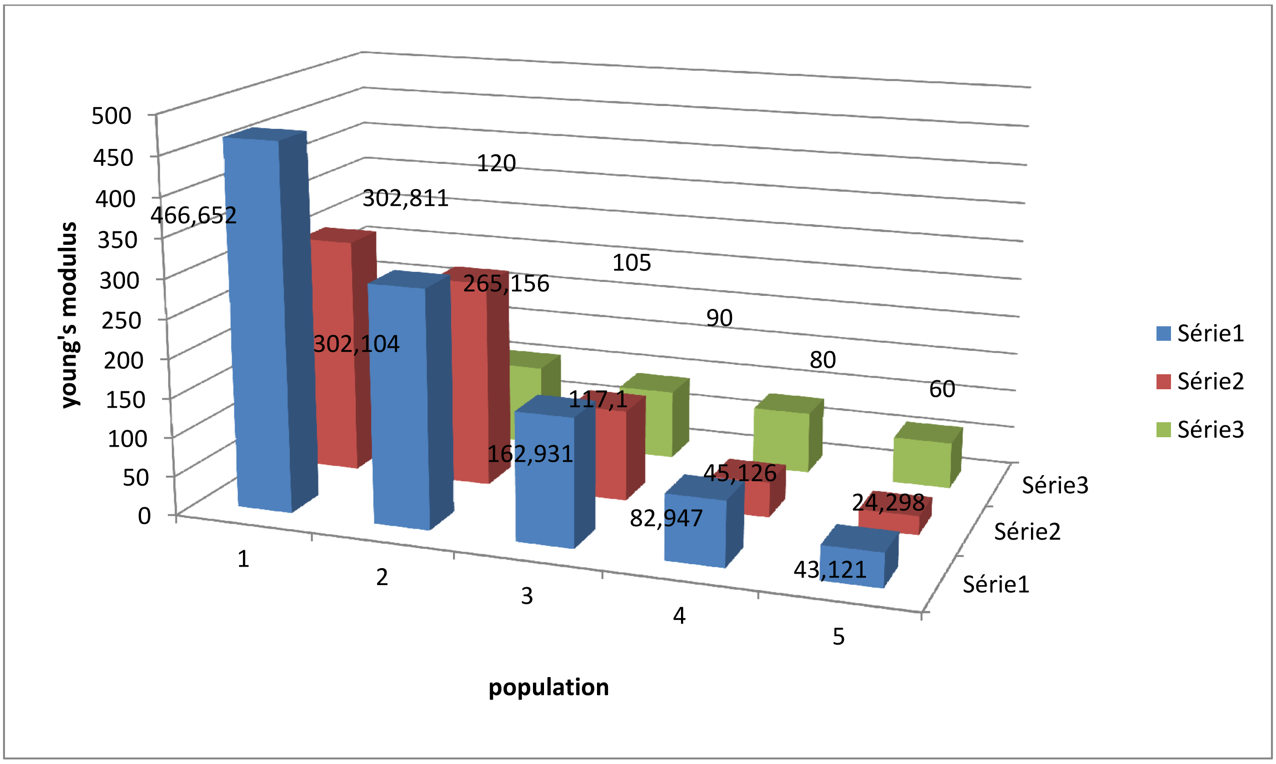

Fig.4: Young's modulus depending the sizing rate and temperature

Series 1 show the variation of Young's modulus of material manufactured with Tannin's binder depending temperature, the series 2 those type 2 materials and Series 3 the variation of temperature depending population.

On the abscissa axis are the five different populations of each type of materials. The Young's modulus increase simultaneously with the temperature and the sizing rate.

Square error

Square errors were calculated with a risk of 5% by varying the temperature and sizing rate.

3.1.4 Moisture Resistance

Study of moisture resistance has led us to conclude that type 2 materials with high temperature sintering are more resistant than the type 1; both caught in the same experimental conditions.

3.1.5 Water absorption rate

The calculation of the absorption rate in each sample depending temperature allows us to obtain:

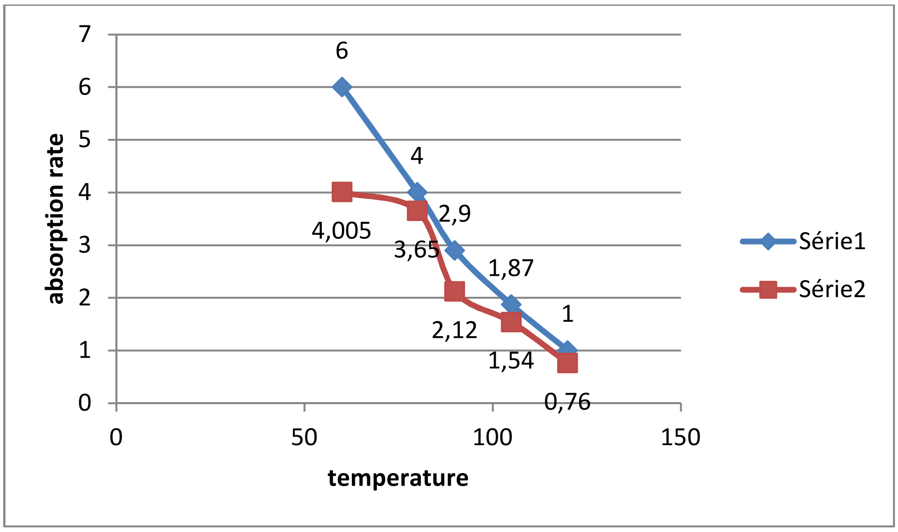

Fig. 5: Absorption rate depending the temperature

The absorption rate increased simultaneously with the drying temperature and the sizing rate.

3.1.6 Comparison

Given the different results above, type 1 materials are stiffer than type 2 materials.

3.2 Discussion

Mechanicals properties are increased with the sizing rate and drying temperature, it means that, when the quantity of water contained in the material is evaporated with the temperature and time, materials become stiffer. The high sizing rate is one of adverse effects in a composite material because materials are not resistant to moisture. The ratio: $(young's\ modulus\ of\ type\ 1\ \ materials\ )\ /\ (\ Young's\ modulus\ of\ type\ 2\ material)\ \geq 1.15$ means that Mechanicals characteristics of type 1 materials are at least 1.15 times those of type 2 materials. In fact, tannins' binder are more important than Urea Formaldehyde's resin and mechanical properties of materials manufactured with tannin's binder of autranelle congolensis are greater than those manufactured with Urea Formaldehyde's resin. Materials manufactured with tannin's binder can be resistant to moisture contrary to materials manufactured with urea formaldehyde resin. Materials manufactured with tannin's binder of autranelle congolensis with 65% rate would give the same results as materials containing 78% of Urea Formaldehyde's rate (Young's modulus of type 1 materials is 402.001at 65 % rate against 402.8 for type 2 materials at 78 % ....), these values may be justified by good mechanical performance of tannin's binder of autranelle congolensis. The Sizing rate content in the biodegradable material must be as low as possible to increase the resistance to moisture and not only for economic reasons, as it is the case in the synthetic materials **[7].**

4 Conclusion

Materials made from tannin's binder of autranelle congolensis have better characteristics than those based on Urea Formaldehyde: so we can replace urea formaldehyde resin by tannin's binder of autranelle congolensis to manufacture biodegradable materials (chipboard) and contribute to the fight against environmental pollution. It is question of encouraging the development of green adhesives (especially in Africa) based on data derived from plants (tannins, lignin, and cellulose) to produce biodegradable material and suggests methods of conservation of these adhesives.

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