

# Incorporation of Clay into Natural Rubber (Hevea) for the Production of Tile Adhesive

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## Abstract

Natural rubber latex is the white liquid in the form of the colloidal dispersion of rubber globules suspended in the aqueous liquid. Produced in large quantities in Ivory Coast, the local transformation of natural latex has so far remained insignificant, although some attempts have been made to use it in the manufacture of flexible facade briquettes for rounded walls. Thus, this study aims to incorporate clay as a filler in natural latex for use as an adhesive for tile installation. To do this, different proportions of clay paste were added to the natural latex and the resulting mixtures were used to make the sample and tile adhesive. From the analysis of the results obtained, it appears that the samples with a clay paste density of 0.8 and 1 absorb less water and shows good pull-out strength. The mixtures of 30% and 35% latex and 0.8 and 1 clay paste density respectively have pullout stresses greater than 1 N/mm<sup>2</sup>. According to NF EN 1348, these adhesives can therefore be used as tile adhesive.

## Keywords

Adhesive Mortar, Rubber Latex, Clay, Tile, Adhesion

## 1. Introduction

Cote d'Ivoire produces 60% of Africa's rubber, making it the largest rubber producer in Africa and the seventh largest (7th) producer in the world. It reached 800,000 tonnes in 2018 compared to 603,000 tonnes in 2017 and 468,000 in 2016 [1]. Forecasts are for a 20% increase over the next three years. But despite this steady rise, individual incomes are falling as global rubber production increased in three years from 9 million tons to 13 million tons in 2017, but demand has not kept up and world prices have fallen from \$5,000 per ton to just \$1,000, leading to a collapse in income for rubber farmers. As more rubber latex is exported than processed locally, this has led to a decline in the economy of the sector. The build-

ing field uses a wide range of products: bricks, tiles and briquettes all bonded with mortar. The mortar is a mixture of aggregates (particles less than 5 mm) and hydraulic binders (cement or lime) or organic, added water in a previously determined proportion to obtain a plastic paste facilitating the installation of masonry elements. The main function of a mortar is to bind the masonry elements together so that they form a block [2]. In addition, there are different types of mortars depending on the job.

These are cement mortars, lime mortars, cement-lime mortars, earth mortars and adhesive mortars. The last group, adhesive mortars are in powder form (bagged) composed of one or more hydraulic binders (cement), organic (resin, latex), mineral fillers (sand, etc.), admixtures and they are used for tiling walls and floors (interior and or exterior). Latex is now widely used in industrial formulations where it is valued for its adhesion properties in various fields such as paint, paper, adhesive and the cement industry [3]. The addition of latex indeed gives these mortars mechanical properties, adhesion or water permeability properties that only the hydraulic binder cannot achieve [4]. However, the most commonly used latexes in mortars are synthetic types (derived from petrochemicals) [5] and mention may be made, *inter alia*, of poly (vinyl acetate) or vinyl acetate copolymers, vinylidene chloride and polyacrylates and their copolymers. In addition, companies producing synthetic latex are risky for workers because of diseases such as: leukemia, lymphoma and myeloma because the products used are toxic and the mortars obtained are expensive which results in the abandonment of these adhesive mortars in favor of cement mortar for laying tiles. As for latex from rubberwood, it finds its application in tires after vulcanization, in aeronautics, mechanics, textiles and the medical field. In addition, its industrial production results in vulcanization fumes which also contribute to atmospheric pollution. Natural Rubber (NR) is produced in large quantities in Côte d'Ivoire and its local processing has so far remained insignificant. Research within materials science laboratory (Abidjan-Côte d'Ivoire) on the use of latex as a binder has made it possible to make materials based on natural latex:

- Sablatex (sand-latex) for facades of rounded walls [6];
- Briquettes made from natural latex and laterite [7].

Thus, this study is carried out with the aim of making an adhesive without cement and without synthetic latex, in other words by substituting it with natural rubber latex. In fact, natural rubber latex in its liquid form is little used because in the space of a day it coagulates so its storage is problematic. In contrast, this work aims to propose a method of using this liquid latex for the production of fluid adhesive that can be stored over a long period (at least one year) before this coagulation occurs.

## 2. Materials and Method

### 2.1. Raw Material

The rubber latex used for making the adhesive tiles comes from a private rubber

plantation located in the municipality of Dabou (Côte d'Ivoire). It is used in its liquid form for making samples. After sampling, an ammonia solution is added to it in order to keep it in the liquid state and send it to the laboratory for being looked into.

The clay used is extracted on the site of Dabou. The samples collected are dried for two weeks and then reduced into pieces of about two centimeters with a jaw crusher before being crushed in a ball mill. The powder obtained is passed through a 1 mm sieve. It is the passerby thus collected that serves as the basic raw material for this study. This clay is clayey silt composed mainly of kaolinite.

## 2.2. Preparation of Samples

Liquid latex contains particles of monomers dispersed in an aqueous phase (serum).

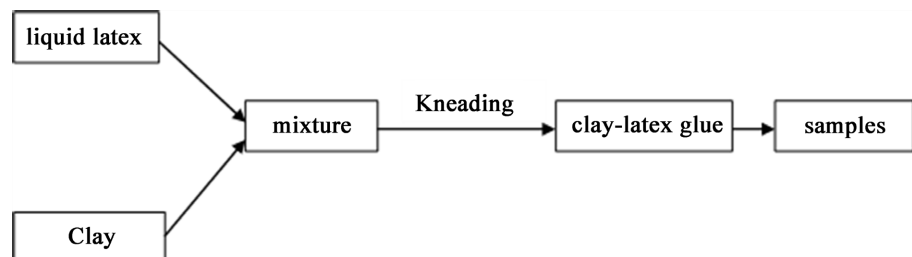
As it coagulates, the monomer particles clump together forming a glue that binds the elements together. These processes rely on this ability of natural latex to bind together for use as a tile adhesive. Liquid latex has very low viscosity. It flows easily. Also to give it smoothness, a mineral filler has been added. The mineral filler used is clay. Clay was used because it has a low specific surface and its surface is negatively charged (kaolinite), what allows them to bond with resins and polymers. Two processes were used to develop tile adhesive based on natural rubber latex.

- First process: liquid latex plus mineral filler as a tile adhesive.

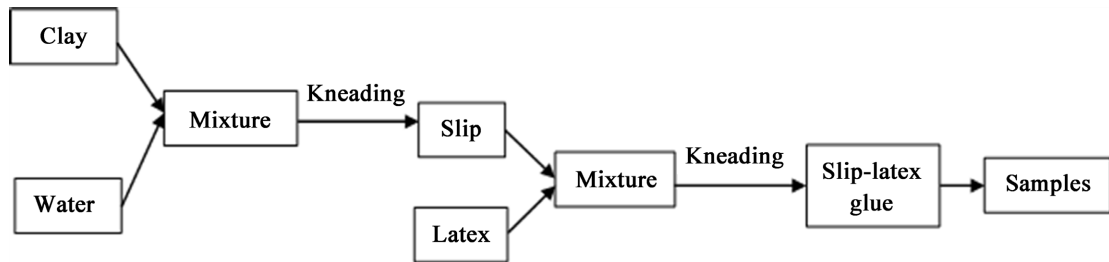
This first process is summarized in **Figure 1** and it consists of two phases: mixing and kneading. Clay is added to an amount of latex (mixture). Then, the clay-latex assembly is kneaded with hand for about 25 seconds to obtain a smooth paste visibly. The product obtained is an adhesive which is carried in the  $4 * 4 * 16 \text{ cm}^3$  molds.

- Second process: liquid latex plus slip (clay + water) as tile adhesive.

Clay has a water absorption capacity that should be taken into account. To account for this fact, a clay slip was used as a filler in this process. A water/clay ratio was defined (0.2; 0.4; 0.6; 0.8; 1). For each ratio, the added latex content varies from 20%, 23% and 26%. **Figure 2** summarizes the different phases that make up the latter: two mixing phases and two mixing phases. The clay is first added to the water and then the whole is kneaded for 30 seconds to



**Figure 1.** Diagram of the clay-latex adhesive tile production process.



**Figure 2.** Diagram of the glue-tile production process (clay + water) + latex.



**Figure 3.** A sample of adhesive mortar.

give the slip. Latex is added to the slip obtained. Then, the whole is kneaded again for 25 seconds so as to obtain a visibly homogeneous paste. It is the slip-latex glue that is worn in the molds.

**Figure 3** indicates a sample of adhesive mortar in a mould of type  $4 * 4 * 16$  cm.

### 2.3. Formulation of the Adhesive Mortar

Naturally, liquid latex behaves like a glue that can be used to bind things together. However, this glue is too fluid to be used as a mortar. Also, different proportions of clay were added as fillers. This adhesive must have sufficient fluidity to perform this function of binder. This is why the first latex content was taken at 16% with 84% clay which corresponds to the least fluid dough possible. In addition, to limit the amount of latex and increase the fluidity of the different mixtures, different amounts of water were added to the mixture. These different formulations used for the design of the different adhesives are presented in **Table 1**.

### 2.4. Characterization of the Adhesive Mortar

#### 2.4.1. Absorption Tests

Water absorption (Abs) is the capacity of certain materials to absorb water in the liquid state [8]. After the preparation and drying of the samples, they are immersed in water for 24 hours. After 24 hours, the samples are removed from water and their surface wiped with a cloth to remove surface water from them. The water absorption by immersion (Abs) expressed as a percentage of the dry mass is calculated by the following relationship.

**Table 1.** Proportion of the different constituents.

Username	Latex (%)	Clay (%)	Water/clay
	LA	Ar	E/Ar
ArLA <sub>16</sub>	16	84	-
ArLA <sub>23</sub>	23	77	-
ArLA <sub>28</sub>	28	72	-
ArLA <sub>33</sub>	33	67	-
ArLA <sub>37</sub>	37	63	-
ArLA <sub>44</sub>	44	56	-
ArLA <sub>50</sub>	50	50	-
ArLAE <sub>20-0,2</sub>	20	80	
ArLAE <sub>23-0,2</sub>	23	77	0.2
ArLAE <sub>26-0,2</sub>	26	74	
ArLAE <sub>20-0,4</sub>	20	80	
ArLAE <sub>23-0,4</sub>	23	77	0.4
ArLAE <sub>26-0,4</sub>	26	74	
ArLAE <sub>20-0,6</sub>	20	80	
ArLAE <sub>23-0,6</sub>	23	77	0.6
ArLAE <sub>26-0,6</sub>	26	74	
ArLA <sub>20-0,8</sub>	20	80	
ArLAE <sub>23-0,8</sub>	23	77	0.8
ArLAE <sub>26-0,8</sub>	26	74	
ArLAE <sub>20-1</sub>	20	80	
ArLAE <sub>23-1</sub>	23	77	1
ArLAE <sub>26-1</sub>	26	74	

ArLA = clay + latex, ArLAE = clay + latex (NR) + water.

$$Abs(\%) = \frac{m_H - m_S}{m_S} \times 100 \quad (1)$$

with: *Abs*: water absorption;

$m_H$ : wet mass (g);

$m_S$ : dry mass (g).

#### 2.4.2. Initial Adhesion Test

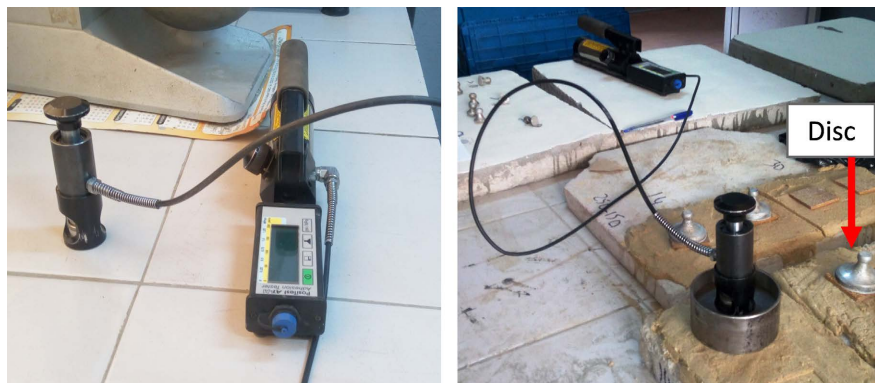
The adhesion test measured according to EN 1348 is a test carried out to determine the ability of the mortar to adhere correctly to the substrates to which it is applied. Adhesion is measured by evaluating the minimum tensile stress required to loosen or break the coating perpendicular to the substrate. The adhesion strength corresponds to the value read on the traction device (**Figure 4**).

### 3. Results and Discussion

#### 3.1. Choice of the Design Process



For masonry work, the glue (binder) used to build the masonry units must be neither too fluid so that it does not flow outside the partitions nor too rigid for its handling to be easy. Therefore, it is necessary to find the right design process. To find the right compromise between fluidity and consistency of the fresh glue from natural latex, on the one hand their workability and homogeneity were analyzed and on the other hand their appreciated hardness.

**Table 2** shows the appearance of different glues from the different production processes. The mixture obtained after kneading in method 1 (clay + latex) gives a non-homogeneous paste with the formation of lumps. As for method 2 (clay + latex + water), the mixture obtained after kneading is visibly a homogeneous paste. Thus, when clay powder is added to the latex, the latter adsorbs the water contained in the latex. This speeds up the coagulation reactions which lead to



**Figure 4.** Pull-of adhesion tester.

**Table 2.** Appearance of the different mortars.

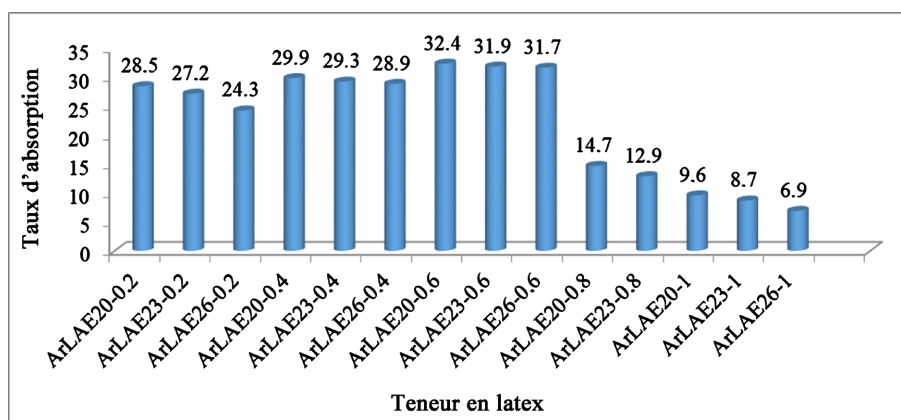
	Mixed	Appearance of mixtures	Image
<b>Process 1</b>	clay + Latex	Training lumps	
<b>Process 2</b>	(clay + water) + Latex	Homogeneous paste visibly	

the formation of lumps. The presence of lumps in the glue is a defect. This method 1 cannot therefore be used. Similar results were obtained by [9]. He showed in his works that the ammonia-containing latex clay mixture for making briquettes gave the formation of lumps. So when he increased the amount of latex in his composition, he got a rubbery sample. Thus, it is essential to obtain a homogeneous mixture in order to ensure good adhesion of the mortar to the surfaces. It is the glue obtained by method 2, which results in a good quality mixture, free of lumps. Liquid natural rubber latex alone was not used as a glue in this work because of its excessively high fluidity, too long open time and poor ability to resist sunlight.

### 3.2. Water Absorption

This absorption of materials leads to many damages. The results of the immersion absorption test performed are shown in **Figure 5**.

**Figure 5** indicates that for the same water/clay ratio, the absorption rate decreases when the amount of latex increases from 20% to 26%. For the water/clay ratio of 0.2, for example, the absorption is 28.5% to 20% latex and increases to 24.3% to 26% latex. The increase in the amount of latex also results in an increase in the amount of rubber monomer. However, during drying, a polycondensation of these monomers occurs. The film formed following this polycondensation sees its volume increased with the latex content. This rubbery film is generally impermeable to water. Thus, when the amount of latex increases in the glue, the volume of rubbery film also increases consequently the glue becomes less and less impermeable therefore absorbs less water. This figure further indicates that for the same latex content, absorption increases with the water/clay ratio up to a maximum value before decreasing. At 26% latex for example, the absorption goes from 27.2% to 31.9% when the water/clay ratio increases from 0.2 to 0.6 then the absorption drops from 31.9% to 8.7% when the water/clay ratio increases from 0.6 to 1. The latex content being constant, the quantity of monomer present in each mixture does not vary. In addition, latex particles have the property of film-forming (latex film formation, the process by which an aqueous



**Figure 5.** Absorption rate as a function of the latex content.

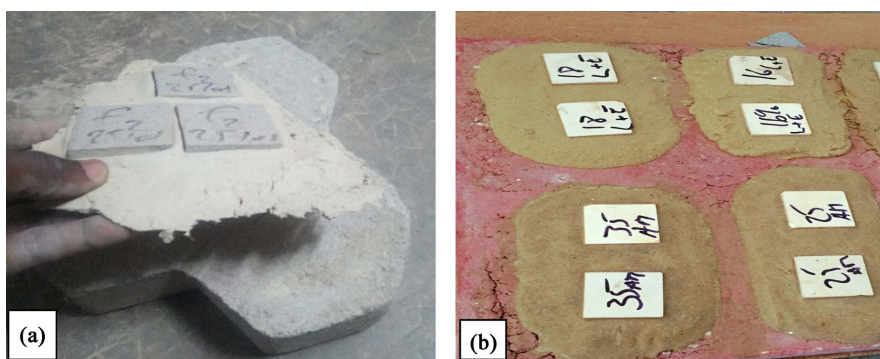


dispersion of polymer particles is transformed into continuous material) [10]. Thus, the evaporation of water influences the film formation of the latex. For the water/clay ratios varying from 0.2 to 0.6, the mixture being firm, the migration of the monomers for their film formation is less and less easy. Thus, filmification occurs in places despite the extent of shrinkage and the dry glue has a discontinuous structure with permeable areas in places. This explains why for these water/clay ratios, the water absorption of the glue increases. When the water/clay ratio changes from 0.6 to 1, the workability of the glue becomes normal. The rubbery monomers can easily migrate into the fresh material to form, along with the importance of drying shrinkage, a continuous film formation. This is why the absorption decreases. On latex cement mixtures, [11] finds a decrease in water absorption. He explains this by bringing together the monomers contained in the latex until they reach their maximum compactness and their coalescence (*i.e.* fusion) with the hydrated cement particles. A film then forms which reduces the pores in the mortars. Similarly, [12] show that the addition of styrene-butadiene (SB) in cement mortar greatly decreases the water-accessible porosity as well as the water absorption. On the other hand, [4] states that in latex-adjuvanted mortars, latex particles fill the pores of the matrix or seal them with a continuous polymer film.

### 3.3. Mechanical Properties of Adhesive Tiles: Adhesion of Adhesives to Tiles and Substrate

Mechanical properties are the most sought-after parameters of mortars. To assess them, the various mortars were subjected to pull-out tests.

The tile adhesive used in laying traditional tiles is applied between the tiles and the substrate to which the tiles are to be bonded. It thus ensures adhesion between the two elements. So adhesion seems to be the essential criterion for the use of an adhesive in the laying of tiles. Also, all the glues produced were subjected to the adhesion test. According to the results, after (28 days) of drying (Figure 6), the adhesives ArLAE0.2, ArLAE0.4 and ArLAE0.6 have a bad adhesion with the tiles and the support while there is a good adhesion between the tiles and the substrate with ArLA0.8 and ArLAE1 adhesives.



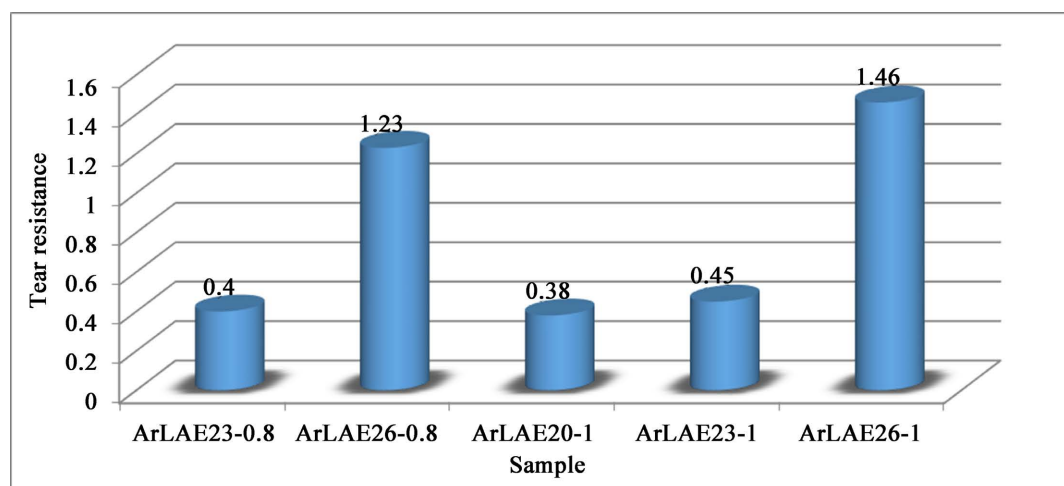
**Figure 6.** Adhesion of glues: (a) No adhesion of glues; (b) perfect adhesion of glues.



This poor adhesion of ArLAE0.2, ArLAE0.4 and ArLAE0.6 glues is due to the fact that there is no reaction between the latex and the various supports. The adhesion of the various adhesives to the substrate and to the tiles is certainly ensured by the anchoring of the latex in the roughness and voids on the surface of the substrate and the tile. In order for the glue to penetrate these different surface roughness, it must first be wet. Thus, for water/clay ratios of 0.2, 0.4 and 0.6, the lack of adhesion is linked to the fact that the glues are too dry: their workability is very firm to firm; the presence of clay makes it difficult to flow. The liquid fraction contained in the glue cannot sufficiently soak the surface of the support. This is therefore not moistened enough to allow adhesion between the two materials. With all other glues prepared, the bond strength was determined because these contain sufficient water and have workability which is either normal or fluid. The variations in pullout stresses (adhesion) observed are explained by the increase in the amount of latex and the existence of microcracks in the dry glue due to drying shrinkage. Indeed, when the amount of latex increases in the glue, the anchoring points of the rubber monomers in the various supports multiply, hence the increase in the adhesion force.

[13] [14] have shown that the cracking of mortars causes local detachment and causes a loss of adhesion between the two materials. The presence of microcracks at the interface induced by the restricted shrinkage is, according to these authors, at the origin of the adhesion losses. However, to be considered as a tile adhesive, the EN NF 1348 standard would require that during the tensile adhesion test, the tear strength obtained is  $\geq 1 \text{ N/mm}^2$ . Below this value, this standard considers adhesive mortars as having poor adhesion. The real pullout stress of the mortars was determined according to the EN NF 1348 standard and the results are presented in **Figure 7**.

According to standard EN NF 1348, an adhesive mortar which has good adhesion and which must be used for tiling work must have a pull-out stress (traction adhesion) greater than or equal to  $1 \text{ N/mm}^2$ . Below this value, this standard



**Figure 7.** Tear resistance of ArLAE mortars (0.8 and 1).

considers adhesive mortars as having poor adhesion. The adhesives ArLAE26-1 and ArLAE26-0.8 have good adhesion because they have pull-out stresses greater than  $1 \text{ N/mm}^2$  from 18% latex content. ArLAE1 adhesives with 30 % latex content have good adhesion because the pull-out stress obtained is  $2.6 \text{ N/mm}^2$ . Similarly, ArLAE26-1 and ArLAE26-0.8 adhesives have peel strength of over  $1 \text{ N/mm}^2$ .

#### 4. Conclusion

The homogeneity of the mixtures made it possible to choose a process for developing glues based on natural rubber latex. The different mixtures produced exhibited different homogeneities. The clay-latex mixture gives a non-homogeneous paste with the formation of lumps and the clay-latex-water mixture is a homogeneous mixture. The clay-latex mixture was therefore given up because of its poor homogeneity. ArLA0.8 and ArLAE1 adhesives absorb less water than other manufactured adhesives. The pull-out test showed that the adhesives ArLAE0.2, ArLAE0.4, ArLAE0.6 have poor adhesion with the tiles, the support and the adhesives ArLAE23-0.8, ArLAE20-1 and ArLAE23-1 have pull-out strength of less than  $1 \text{ N/mm}^2$ . ArLAE26-0.8 and ArLAE26-1 give better results and can be used as tile adhesive. However, when the thickness of the bonding mortar is large, this mortar has practically no rigidity. Therefore, it should be considered to increase its stiffness by adding granular material.

#### Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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