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**Abstract:** The remarkable push of technologies increasingly recommends materials for their constructions. Hence the extraction of alumina is for different uses. The exploitation of bauxite pollutes the surrounding areas in this case the discharge of mud wheel. Furthermore, the excessive use of dangerous conventional synthetic insecticides continues to be in circulation in Cameroon despite the known adverse effects. These problems lead us to formulate bio insecticides from rejection of the extraction of alumina and extracts of *S. aromaticum* and *Citrus sinensis* which respectively offer us essential oils eugenol and limonene having insecticidal properties against the main insects, pests of corn stocks, *Sitophilus zeamais*. From the kinetic point of view of the adsorption of the extracts of eugenol, limonene and their association by the red mud, the adsorption curve shows that the Eu/Li mixture (30/70) is better by 0.36 mg/g during the 24 h. The results showed a high toxicity of the Eu-Li binary mixture of ratio 30/70 with respective mortality rates of 95.88% after 8 days of exposure to the concentration of 5 mL/L on the adults of *Sitophilus zeamais*. The other composites induce an average rate of 80%. The results from the physicochemical analysis of corn before and after treatment with formulated insecticides showed a slight loss in nutritional quality of 14.90% in ash, 12.5% in protein, 16.14% in lipids and 13.63% in total sugars for 180 days. The powdery formulation from red mud and Eu-Li (30/70) mixture can therefore ensure the protection of corn stocks against *Sitophilus zeamais* in a farming environment.

Key words: Insecticidal activity, red mud, Syzygium aromaticum, Citrus sinensis, Sitophilus zeamais, physicochemical properties.

# **1. Introduction**

Global grain production is of critical importance to a country's economy. In Africa, and particularly in the northern part of Cameroon, the cultivation of cereals occupies a prominent place because of their strong demand, whether by local consumers or by industries (Nguemtchouin et al., 2010). To increase cereal yields, compensate for post-harvest losses and better preserve cereals, agricultural producers increasingly resort to fertilizers and insecticides (Sékou et al., 2000). Insecticides thus remain an essential means of controlling infestation of foodstuffs during storage (Nguemtchouin et al., 2014). Unfortunately, some synthetic insecticides such as chlorine derivatives have a negative impact on the environment and can induce chronic intoxication of consumers, resistance in pests (Attia and Frecker, 1984). In addition, these are not easily accessible to all farmers. In the search for alternative methods of controlling these pests, the plant kingdom offers many possibilities. Tapondjou et al. (2003). Biopesticides of plant origin have the advantage of being biodegradable and less toxic than

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synthetic insecticides (Pegalepo et al., 2019, Papa et al., 2015, Diouf et al., 2014, Tapondjou et al., 2003). The plants from which biopesticides are prepared are widely found in nature and are found in human food as condiments or spices. These plants have important medicinal properties. Thus, plant species such as Azadirachta indica, Ocimum gratissimum, Monodora myristica, Momordica charantia have been commonly used for the preparation of biopesticides (Nguemtchouin et al., 2013, Adesina et al., 2012, Tapondjou et al., 2003). Fixing these two extracts by adsorption on a solid support such as a composite constitutes a particularly interesting approach. Indeed, the use of natural resources as a raw material for the production of composite materials is of great interest, from an economic and ecological point of view (Zakariae, 2016). The use of composites in recent decades has increasingly opened up prospects for improving the properties of materials (Mbey et al., 2012). Among composites, polymer-clay materials arouse great interest (Okamoto et al., 2005). In this field, the use of clays as nanofillers is justified by their biocompatibilities, their availability and by the nanometric size of their sheets. They can also be used for their physicochemical properties such as large specific surfaces (Siafu et al., 2015). Type 2: 1 clays, such as montmorillonites, are in common use because of their ability to exfoliate easily. Kaolinites, type 1: 1 clays, remain very little used despite the great abundance and great purity of kaolinic deposits (Murray, 1988). It is therefore logical to consider that, if the adequate dispersion conditions are achieved, the use of kaolinites as filler in polymer-clay composites may be of real technical and economic interest. Red mud is the solid waste by product resulting from the production of alumina by the Bayer Process. On average globally, between 1 and 1.5 tons of red mud is generated per ton of alumina produced and over 150 million tons of this residue is produced (Evans et al., 2016). Unfortunately, it is estimated only about 2-4.5 tons are used annually in some way (cement-500,000 to 1,500,000 tons; raw

material/additive in iron and steel production-200,000-1,500,000 tons; roads/landfill capping/soil amelioration—200,000-500,000 tons; (bricks (tiles, ceramics, construction materials etc.)—100,000–300,000 tons; other (refractory, adsorbent, acid mine drainage, catalyst, etc.))-300,000 tons) (Evans et al., 2016). This means over 145 million tons are being dumped in different parts of the world with the risk of causing another disaster like the Ajka, Hungary red mud spill that killed ten people, injured 200, and eliminated all aquatic life in a 71 km stretch (Mayes et al.). Currently, most red mud produced from alumina plants is disposed in landfills or dumped at sea. The cost of disposal is very high, constituting for approximately 5% of alumina production. Hence the question arises what to do with these large volumes of red mud with high polluting character. Because it contains compounds of Na, Ca, Si and K it can act as filler to fix some compound. In view of its physicochemical composition, this sludge can be used as a support to produce a bio insecticide to replace the clay generally used in previous work. In this sense, the polymer such as the association limonene and eugenol in contact with the red bauxite mud could form ideal bio composites with applications as bio insecticides. Indeed, previous work carried out on gum arabic has shown that it allows the interfoliar spaces of clays to be widened. Moreover, the literature has also shown that gum arabic also has antimicrobial and insecticidal activities (Maria et al., 1992; Elhassan et al., 2016 and Hussien et al., 2019; Zaccheus et al., 2019). Thus, composite materials based on kaolinite and gum arabic have proven to be good candidates for the formulation of a powdered insecticide. So far no work has been done on the use of red bauxite sludge and limonene and eugenol extracts in the preservation of cereals. Essential oil clove has very interesting properties as an antibacterial thanks to its composition of eugenol and eugenyl acetate. It could thus be an alternative to the use of antibiotics to fight against the appearance of

resistant strains (Barbelet, 2015). Orange peels constitute a rich deposit in nutritional ingredients (water, proteins, sugars and minerals) and in functional ingredients (essential oils, fibers, carotenoids, vitamin C, phenolic compounds). However, the most widespread industrial recovery route remains the extraction of essences and essential oils which can be used as an alternative to synthetic fungicides (Tian et al., 2001; Fisher & Phillips, 2006; Ververis et al., 2007; Fisher & Phillips, 2008; Virot et al., 2008; Chutia et al., 2009; Singh et al., 2010). Clove essential oil is distinguished by its antibacterial, antifungal and acaricidal powers. While it is not the most powerful of the phenolic essential oils, it is above all the least toxic. Barbelet (2015) asserts that essential oil extracts of cloves have been used in the past for the protection of orange trees against pests, insects and fungi, this assertion leads us to believe that we are making the hypothesis according to which the association of extracts of eugenol and limonene could constitute precursors for the development of bio-insecticides. The general objective of this work will be to develop a powdery formulation from the combination of red bauxite sludge and extracts of limonene and eugenol with insecticidal properties. More specifically, it will be a question of producing the precursors, characterizing them, carrying out formulation tests, testing their bio-insecticidal activity, carrying out a test on two samples of voandzou and cowpea and evaluating the physicochemical and nutritional properties in order to assess the influence of bio-insecticides on shelf life.

# 2. Experimental Procedures

2.1 Bauxite Red Mud, Eugenol and Limonene Preparation and Characterization

## 2.1.1 Bauxite Red Mud

Due to the fact that Cameroon is not yet processing its bauxite, the bauxite collected at Ngaoundal  $(6^{\circ}27'55'' \text{ N}, 13^{\circ}16'16'' \text{ E})$  was used in producing the red mud in laboratory as described in our previous studies (Tsamo et al., 2017 et Ze et al., 2018). The large blocks obtained were crushed in a porcelain mortar with a porcelain pestle until small blocks were obtained. Then, it was dried at 105 °C in an oven (HERAEUS type VT 5042 EK) for least 24 h. After which it was introduced into a jar with alumina balls closed and placed on a ball crusher and rotated for 4 h. The powder obtained was sieved at 75  $\mu$ m, stored and used for characterization.

The chemical composition of red mud was determined using the X-Ray Fluorescence (XRF) technique (AXIOS PANalytical, Dy 1680) while the surface functional groups were determined by Fourier transform infrared spectroscopy (FTIR Spectrophotometer (Bruker Make), Model: ALPHA-P). The presence of mineral compounds in red mud was determined by X-ray Diffraction (XRD) using a Philips X'Pert PRO diffractometer. The different diffraction peaks recorded were compared with those of similar samples reported in literature for identification. The differential scanning calorimetry-thermgravimetric (DSC-TG) technique (SDT Q600 V20.9 Build 20) was used to identify the different phase transformations involved in red mud sample with heating while Scanning Electron Microscopy (SEM) was used to determine the surface morphology of the solid materials.

2.1.2 Eugenol and Limonene Preparation and Characterization

(1) Plant Material

The floral buds of *S. aromaticum* and eugenol were purchased from the central market of the city of Ngaoundere town, Cameroon (7°19'39.5" N, 13°35'05" E). For the extraction of oil, dry flower buds were selected in an oven with air circulation at 37 °C/48 h, and soon afterwards they were sprayed in a knife mill in the Physics-Chemistry Laboratory of the Food and Water Quality Control Program (PCQA), University Institute of Technology of Ngaoundere.

(2) Essential Oil Extraction

The extraction of the essential oil of S. aromaticum

and eugenol was carried out with 300 g of the ground vegetable product, and it was diluted in water in the proportion of 1:10 by hydrodistillation using the Clevenger system for 3 h at 100 °C. The essential oil collected was dried with anhydrous sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>), and the final volume found was used to determine the yield through the mass/volume ratio by measuring the density. Mass/volume ratios were calculated from the mass (g) of the initial vegetal material and the volume (mL) of essential oil obtained after extraction. The essential oil samples were kept at 25 °C and then weighed.

(3) Physical-Chemical Analysis of Essential Oil

Some physical-chemical analyses were performed on the essential oil of *Syzygium aromaticum* and eugenol for density measured with a glass pycnometer, refractive index calculated with ABBE 2WAJ refractometer (PCE Instruments, Southampton, United Kingdom), color and appearance that were visually verified by three different people, and determination of solubility that is carried out through the ratio of 1:1 of oil and ethanol 80% until its complete solubilization.

# 2.2 Method of Absorption of Essential Oils by Materials

2.2.1 Extraction and Preparation of the Modified Red Mud

After extraction of the alumina, the red mud was washed, then soaked in 30% hydrogen peroxide to remove the organic matter and dry in an oven for 24 h at 105 °C then introduced into a solution of sodium chloride (NaCl) to replace all cations exchangeable by sodium ions (Na<sup>+</sup>). The method applied is that used by Mbouga et al. (2010). To do this, 50 g of treated red sludge is introduced into 2 L of 1 M NaCl solution. The mixture is left stirring for 24 h using a magnetic stirrer. The suspension is then centrifuged and washed with

deionized water until the complete elimination of chloride ions (Cl<sup>-</sup>). The complete absence of Cl<sup>-</sup> ions is proven by a negative test for silver nitrate (AgNO<sub>3</sub>). The red sludge devoid of chloride ions is dried at 70 °C for 24 h, pulverized in a porcelain mortar and then sieved at 75  $\mu$ m.

Table 1 shows the eugenol: limonene ratio used for the adsorption of the extracts to the material. Five (5) mL of each mixture was introduced into a burette at a flow rate of 0.2 mL/min in a 250 mL Erlenmeyer flask containing 50 g of previously treated red mud and 150 mL of distilled water, the mixture is stirred at 800 trs/min for 24 h, filtered and dried in an oven at 70 °C for 24 h. The filtrate recovered was assayed with the Libermann-Burchard reagent to be analyzed by UV-visible spectroscopy. The solid phase was the formulated insecticide.

2.2.2 Methods of Assaying Lupeol

The reference compound used to establish the calibration curve in this work is lupeol, a pentacyclic triterpene isolated from the ethyl acetate extract of the leaves of *Prosopis africana* at the hexane/ethyl acetate ratio (95:5).

The demonstration of this compound was carried out by the reagent of Liberman Buchard. Therefore, 5 mL of each of dilute lupeol solution (20, 40, 60, 80 and 100 mg/L), 5 mL of the mixture of acetic anhydrous/chloroform (5/5: V/V) is added; then add a few drops of concentrated sulfuric acid until a green-purple color appears. Their absorbance is measured by UV-Visible spectrophotometry of the Spectrumlab 22 Spectrophotometer brand at 530 nm against the blank consisting of the supernatant obtained from a suspension of red mud previously treated in methanol.

The absorbance allowed us to establish the calibration curve. The adsorbed quantities  $(4_e)$  are given by the following expressions:

Table 1 % Eugenol-limonene ratio.

% Eugenol	0	10	30	50	100
% Limonene	100	90	70	50	0

$$Q_e = (C_o - C_e) V/m$$

where V is the volume of the solution used and m, the mass of adsorbent.

2.3 Study of the Parameters Influencing the Adsorption of Essential Oils

### 2.3.1 Influence of Stirring Time on Adsorption

We placed 0.02 g of red mud in beakers containing 30 mL of an extract solution of 20 mg/L strength. The mixture was brought to stirring with an INTLLAB brand magnetic stirrer for times of: 3; 5; 10; 15; 20; 25; 30; 35; 40 and 45 min, then strain. Each filtrate was assayed by UV spectroscopy to determine residual extract concentrations.

2.3.2 Influence of the Mass of Materials

In 5 different beakers containing 0.02; 0.04; 0.06; 0.08 and 0.1 g of red mud, we introduced 30 mL of extract solution of 20 mg/L concentration. The whole was stirred for 10 min, and then filtered. Each filtrate was assayed by UV spectroscopy to determine the residual concentrations of extract from the leaves.

## 2.3.3 Influence of pH

We introduced 0.02 g of composite in beakers containing 30 mL of extract 100 mg/L of pH respectively: 3; 5; 7; 9; 11. After stirring for the equilibrium time, the solutions are filtered and the filtrates assayed.

## 2.3.4 Influence of Temperature

We introduced 0.02 g of composite into beakers containing 30 mL of a 100 mg/L methanol leaf extract solution. The mixture was stirred using an SH-2 MAGNETIC STIRRER brand heating magnetic stirrer while varying the temperature from 25 to 50 in steps of 10 °C, then filtering after 10 min. Each filtrate was assayed by UV spectroscopy to determine the residual concentrations of extract from the leaves.

## 2.4 Insect Rearing

Intensive rearing of *Sitophilus zeamais* bruchids has been developed at the Chemical Engineering Laboratory of the University of Ngaoundéré in order to obtain sufficient insects for bioassays. Infected maize was purchased at the Ngaoundéré market in the Adamaoua region. In order to have a homogeneous test population, all insects present in the collected maize samples were eliminated. The insect-free samples were kept in jars at optimum conditions of 25-27 °C and 70-75% relative humidity which favor the multiplication of the insects for a period of 15 to 21 days. The tests were carried out on insects older than 21 days. The adults were fasted for 24 h before the test was performed. The corn kernels used are healthy ones sorted and washed in deionized water and then dried.

2.4.1 Poudrox (Malathion)

Malathion is an organophosphate which acts by contact, ingestion and inhalation on beetles, aphids and mites. Powrox or malathion (Malagrain DP5) is used as a reference (positive control) in our work.

2.4.2 Study of Insecticidal Activity

The formulations of 1%, 2%, 3%; 4% to 5% of formulation weight per weight of corn seed was used.

Each dose of insecticidal powders contained in a box received a quantity of 20 g of corn grain and then infested with 10 insects previously starved for 48 h and then stored at room temperature. The number of dead insects is calculated every 24 h for 5 days after treatment (Nukenine et al., 2010). The negative control is the composite alone and the positive control is Malathion which is an insecticide generally used by farmers to protect cereals and the corrected mortality rate is expressed according to the formula of Abbott (1925).

#### $Mc = (Mo-Mt)/(100-Mt) \times 100$

With Mc: mortality corrected in %; Mt: mortality observed in the negative control; Mo: mortality observed in the test.

# 2.5 Assessment of the Physico-Chemical Quality of Corn Seeds during Storage

The best inhibitory concentration obtained during previous treatment will allow us to evaluate the

physicochemical properties of the seeds during storage and to estimate the shelf life of our seeds. These tests will be carried out at 0.30; 60; 90; 120; 150 and 180 days. To carry out this study, the ash content, protein content, fat content and sugar content will be carried out on the flour obtained from the seeds. The principle of the analysis is as follows: crude ash is obtained after high temperature calcination of a dry material. The sample to be analyzed of known dry weight is heated in a muffle furnace until its complete calcination to ash, as regards the determination of proteins, the method is based on the determination of the fraction of total proteins soluble in the water. The total protein content was determined by the spectrophotometric method described by Dubois et al. (1956). It is based on the gradual solubilization of the fat in a sample using an organic solvent. The total carbohydrate content was determined by the spectrophotometric method described by Dubois et al. (1956). In fact, in a hot sulfuric medium, neutral oses produce furfural derivatives (aldehyde derivatives of furan) which condense with resorcinol to give a vellow-orange complex absorbing at a wavelength of 450 nm.

## 3. Results and Discussion

XRF method has been used to identify the major minerals and chemical compounds present in the clays. The main oxides are SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, CaO, and MgO. The kaolin was rich in SiO<sub>2</sub> (7.68%), in Fe<sub>2</sub>O<sub>3</sub> (27.21%); and in Al<sub>2</sub>O<sub>3</sub> (29.6%) and contained only small amounts of CaO, MgO, K<sub>2</sub>O, Na<sub>2</sub>O, SO<sub>3</sub> and P<sub>2</sub>O<sub>5</sub> (Table 2). Loss on ignition (LOI) was 20.26%. The chemical composition indicated the presence of considerable amounts of silica-and iron-bearing impurities

The average size of the gains is  $34.75 \ \mu m$ .

The particle size distribution of a material is an

important physical property because it has an impact on the reactivity of the material (Diaz et al., 2010). The finer the particles, the higher the reactivity of this material. This reactivity will allow better adsorption of the principle, which is active within the pores of the red mud. The analysis of the spectrum of the red mud (Fig. 3) consists of the absorption bands of 3,600-3,900 cm<sup>-1</sup> (3,653; 3,735; 3,648; 3,335 cm<sup>-1</sup> are characteristic of the vibration of the OH bonds of the hydroxyl group of gibbsite (Kloprogge et al., 2002, Yasiharni and Gilkes, 2012), basic mineral of bauxite. This confirms the results shown by the chemical composition. 1,068 and 973 correspond to the different modes of vibration of the OH bond deformation of the water molecule (Yasiharni and Gilkes, 2012). And 1,450-1,550 cm<sup>-1</sup> are characteristic of the valence vibration of the C = O bond (Mistry, 2009), on the other hand those around  $1,397 \text{ cm}^{-1}$ correspond to the vibration of the Si-O-bond (Vo-Dinh et al., 2001). The peak around 983 cm<sup>-1</sup> corresponds to the vibration of the Si-O bond (Sahu et al., 2010; Castaldi et al., 2008), 733 cm<sup>-1</sup> corresponds to the Fe-OH group (Lefevre, 2014).

# 3.1 Chemical Composition of S. aromaticum Essential Oil

The essential oil presented a yield of 3.8%, a density of 0.989 g/mL at 25 °C, and a refractive index (ND 25) of 1.595. It was soluble in 90% (v/v) ethanol at a ratio of 1:2 and exhibited a transparent yellow color with a clear appearance in all samples. Chemical compounds in *S. aromaticum* essential oil evaluated by GC-MS are shown in Fig. 3. Five major compounds were determined in the *S. aromaticum* essential oil and enumerated in order of elution and retention time. The major constituent was eugenol, representing 52.53% (Table 4).

Table 2Chemical composition of treated bauxite red mud.

Sample	LOI	SiO <sub>2</sub>	$Al_2O_3$	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	SO <sub>3</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	$P_2O_5$	PbO	TiO <sub>2</sub>
Treated red-mud	20.26	7.68	29.6	27.21	2.75	0.08	0.08	4.71	0.06	0.12	-	1.89

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Fig. 1 IRTF spectrum of bauxite red mud.



Fig. 2 Red mud particle size distribution.

D <sub>10</sub>		D <sub>50</sub>	D <sub>90</sub>		
0.775 (μm)		19.250 (µm)	84.226 (	μm)	
Table 4	Chemical composition of Syzy	gium aromaticum essenti	ial oil.		
Peak	Compounds	RT (min) <sup>-1</sup>	Kovats index	PA (%)2	
1	Eugenol	26.55	1,402	51.89	
2	Copaene	27.04	1,379	1.67	
3	Caryophyllene	30.01	1,498	36.43	
4	Humulene	29.95	1,507	4.43	
5	Eugenyl acetate	33.12	1,535	3.78	

Table 3	Numerical values of	the particle size	e distribution of red	l sludge analvzed	by ultrasound.
				sidage anal, sea	



Fig. 3 Chromatogram of Syzygium aromaticum essential oil.



Fig. 4 Inhibition of the ABTS radical by Syzygium aromaticum essential oil and eugenol.

*Syzygium aromaticum* essential oil and eugenol presented concentration-dependent antioxidant activity, as observed in the graph that relates *S. aromaticum* essential oil and eugenol concentration to the percentage of inhibition of the ABTS radical (Fig.

4). The calculated  $EC_{50}$  was 78.98 µg/mL for *S. aromaticum* essential oil and 12.66 µg/mL for eugenol.

Table 5 gives us the average corrected mortalities of insects subjected to the different formulations (Eu/Li

ratio) as a function of the concentration of the formulated bio-insecticide. The insecticidal activity of the two essential oil samples or their combination (eugenol and limonene) varies very significantly with the concentration applied and the duration of exposure. It emerges from this table, a mortality rate ranging from 17.28; 21.58; 34.45; 20.16 and 35.12% for a dose of 1% to 84.35; 87.22; 95.88; 87.58; 83.54 for a dose of 5% for 2; 4; 6 and 8 days of exposure. This analysis lets know the mortality rate with the dose of the bio composite and the contact time. Eugenol alone contributes to good bio-insecticidal activity as does limonene. But the combination of these two essential oils provides the best results. However, all the concentrations tested were found to be relatively toxic to insects. It can be remembered from this table that the 30/70 ration formulation (Eu/Li) makes it possible to deduce a mortality rate of 95.88%, i.e. a high insecticidal effect. This value is close to Malathion which is a commercial insecticide. In view of this reduction rate, we can deduce that it is possible to develop a bio-insecticide from a composite (red mud-eugenol-limonene). The results obtained are in agreement with those of Tamgno et al. (2018).

The work of Boni et al. (2017) carried out on pesticide plants and market garden crop protection was able to demonstrate that certain extracts can have the same efficacy as synthetic insecticides. The increase in insect mortality rates is believed to be due to respiratory poisoning linked to volatile compounds contained in the formulations.

These results are similar to those found by Boeke et al. (2004); Aïboud (2012); Kayombo et al. (2014) whose work has shown that certain plant species reduce the lifespan of adults of *Sitophilus zeamais* and that the mortality rates increase with the doses used.

The mortality observed in the different formulations showed that the latter did not have the same effects on *Sitophilus zeamais*. Indeed, previous work carried out on phytochemical tests of the plant has shown the presence of a multitude of families of compounds such as alkaloids, steroids, phenolic compounds, saponins, tannins, glucosides and triterpenoids (Olivier et al., 2016, Annapoorani et al., 2013). These plant active substances have a broad spectrum of action and do not spare non-target organisms, and do not cause the development of resistance in insects treated like chemical insecticides. The use of plant extracts as an insecticide has long been known. Moreover, it has been noted that pyrethrum, nicotine, rotenone have already been used as agents for controlling insects. However, polyphenols as well as alkaloids are toxic metabolites used as an insecticide in insect control (Abdul-Malik, 2018; Victor et al., 2020). Alkaloids induce toxic effects towards insects (Víctor et al., 2020). Triterpenoids have an anti-nutritional effect. According to Victor et al. (2020), triterpenoids inhibit food intake by phytophagous insects and cause death and malformations in future generations. Thus, we note that a synergistic reaction of the families of compounds within our insecticide formulations has made it possible to explain the high efficiencies against cowpea pests. It is worth noting that the insecticidal effect resulting from the methanol formulation is better than that of hexane and ethyl acetate. In our work, it should be noted that the natural substances used have a good insecticidal action against corn weevil (Sitophilus zeamais), their toxicities were functions of the dose used. It would therefore be interesting to continue research in order to demonstrate the synergistic action in the fight against the corn weevil and other pests of stored seeds.

To assess the physicochemical properties of corn kernels in contact with the composite (red mud-eugenol-limonene) in the proportion 30/70, a kinetic study was carried out over a period of 180 days. It is noted after 180 days of contact of the composite and the corn, a loss of 14.90% of quality in the ash content, 12.5% in protein, 16.14% in lipids and 13.63% in total sugars. These values show that there is an average loss of 15% in the physicochemical quality of the grains. Studies should be carried out on the

Mortality average corrected (%)							
	Concentration (%	6)					Malathion
Eugenol/limonene 0/100	Exposure time in (day)	1	2	3	4	5	1
	2	$17.28\pm4.11$	$25.12\pm3.17$	$27.25\pm5.41$	$26.31 \pm 3.29$	$30.32\pm7.43$	$75.12\pm8.32$
	4	$25.57\pm3.89$	$47.30\pm10.14$	$41.74 \pm 13.23$	$50.15 \pm 16.31$	$51.93 \pm 15.38$	$88.24\pm18.35$
	6	$38.65 \pm 12.35$	$67.09 \pm 17.56$	$60.21 \pm 13.25$	$69.33 \pm 21.07$	$70.15\pm7.85$	$90.68 \pm 11.47$
	8	$62.19 \pm 17.32$	$78.19 \pm 18.74$	$70.49 \pm 15.18$	$82.67 \pm 21.65$	$84.35\pm27.27$	$98.72\pm7.87$
	Exposure time in (day)	1	2	3	4	5	
Eugenol/limonene	2	$21.58 \pm 15.25$	$30.24\pm4.13$	$27.35 \pm 11.68$	$31.43 \pm 12.42$	$34.76\pm15.55$	
10/90	4	$32.36\pm13.18$	$48.93 \pm 5.36$	$43.78\pm23.86$	$51.44 \pm 17.78$	$53.54 \pm 19.73$	
	6	$47.63 \pm 19.87$	$65.82 \pm 18.24$	$60.45\pm9.37$	$75.73\pm15.65$	$79.46 \pm 15.79$	
	8	$60.88 \pm 9.98$	$77.59 \pm 25.21$	$73.32\pm16.62$	$86.65\pm23.44$	$87.22 \pm 18.26$	
	Exposure time in (day)	1	2	3	4	5	
Eugenol/limonene	2	$34.45\pm19.84$	$56.44 \pm 8.12$	$53.79 \pm 22.13$	$60.04\pm15.33$	$65.73 \pm 17.28$	
30/70	4	$47.36 \pm 18.78$	$70.33 \pm 19.21$	$65.09 \pm 15.11$	$78.19 \pm 13.25$	$80.45\pm21.65$	
	6	$60.41 \pm 12.32$	$82.46 \pm 10.54$	$78.76\pm23.67$	$86.56\pm25.78$	$88.13\pm27.31$	
	8	$70.23\pm24.58$	$86.78\pm31.72$	$87.70\pm28.54$	$91.45\pm34.65$	$95.88\pm32.45$	
	Exposure time in (day)	1	2	3	4	5	
Eugenol/limonene	2	$20.16\pm9.45$	$46.46 \pm 15.19$	$44.73\pm17.49$	$57.56\pm21.67$	$60.27\pm5.13$	
50/50	4	$51.28 \pm 11.65$	$63.24\pm12.46$	$60.88 \pm 23.17$	$66.47 \pm 18.35$	$69.35\pm17.57$	
	6	$58.47 \pm 18.34$	$68.54 \pm 14.67$	$69.23 \pm 17.45$	$75.37\pm21.33$	$78.45\pm22.48$	
	8	$65.79\pm21.89$	$76.41\pm24.59$	$79.85\pm28.34$	$81.44\pm21.98$	$87.58 \pm 17.99$	
	Exposure time in (day)	1	2	3	4	5	
Eugenol/limonene	2	$35.12\pm9.87$	$44.45\pm25.56$	$45.19\pm21.28$	$48.12\pm14.25$	$51.32\pm20.38$	
100/0	4	$40.77\pm16.46$	$49.66\pm24.68$	$56.86 \pm 19.45$	$59.38\pm26.75$	$61.32\pm23.15$	
	6	$49.17\pm18.23$	$56.56\pm28.16$	$58.79 \pm 18.83$	$68.89 \pm 27.96$	$72.61\pm22.21$	
	8	$68.28 \pm 19.87$	$69.32\pm18.67$	$70.95\pm6.78$	$79.16\pm22.76$	$83.54\pm27.33$	

#### Table 5 Mortality average corrected with eugenol extract, limonene or their combination.

#### Table 6Corn quality loss rate.

Time (J)	Parameters								
	Ash content	Protein content	Sugar content	Lipid content					
0	2.08	8.56	2.86	4.5					
30	2.08	8.54	2.88	4.38					
60	2.00	8.47	2.79	4.25					
90	1.99	8.23	2.64	4.99					
120	1.87	7.87	2.56	4.57					
150	1.84	7.57	2.50	4.21					
180	1.77	7.49	2.47	3.48					
Quality loss rate (%)	14.90	12.50	13.63	16.14					

nutritional, functional and toxicological quality in order to assess the toxicity in the short and medium term. Microbiological tests will also have to be done in order to assess the microbiological quality of the flour from this storage. These results are in agreement with the work carried out by Nguemtchouin et al. (2012) on the conservation of corn seeds. From its extract, it was able to guarantee storage and the physicochemical and functional properties in the standards for a period of 90 days. The powders tested

showed a positive effect on the conservation of corn seeds with regard to the shelf life of previous work carried out by Scholl (2002). The variation in ash content can be explained by harvesting processes, agricultural techniques and mainly the characteristic soil type and climate (Hamid et al., 2015). The results obtained with regard to the protein content during storage in contact with our composite are similar to those of Health Canada in 2015. This study showed that the treatments have no effect on the protein content of maize, which is in accordance with the observations of Trèche et al. (1996) who showed that the treatments of corn with bio-insecticides do not modify the protein quality. With regard to lipids, from the results recorded, we can conclude that there is a great change in the lipid contents of cowpea after the treatment of the order with 16% loss in nutritional quality. The results obtained remain inferior to those of Khalid et al. (2012). In fact, the slight differences in content may be due to environmental conditions, cultural practices and the genetic factor (Hamid et al., 2015). These results therefore show the effectiveness of powdery insecticides used in preserving corn.

# 3.2 Study of the Influence of the Contact Time of the Extract on the Red Bauxite Mud



strong adsorption of essential oil extracts from the first minutes of contact, followed by a slow increase until reaching a state of equilibrium mainly around 6 p.m. The rapid adsorption observed during the first hours of contact can be interpreted by the availability of the number of vacant active sites on the surface of the adsorbent material followed by the formation of one after their saturation by the adsorbate after the 18 h (Chikwe et al., 2018). The equilibrium time is globally estimated at 18 h for the two materials used.

Fig. 6 shows that the retention efficiency of extracts decreases with increasing concentration. It can be seen from this figure that the adsorbed quantity of essential oil extract increases considerably with the increase in the adsorbate concentration. The increase in the concentration causes the driving force of the concentration gradient to increase, thus increasing the molecular diffusion of the monotriterpene in solution at the surface of the adsorbent. The maximum adsorbed amounts for initial concentrations of 2, 4, 6, 8 and 10 mg/L are of the order of 0 and 1.8 mg/g. Thus, the more the concentration of the solution increases, the more extract molecules are in the solution and the more they are in contact with the adsorbent, then the adsorption percentage increases (Zohra et al., 2006, Uday et al., 2006; Mohamed et al., 2014).



Fig. 5 Influence of the contact time on the adsorption of the extract.



Fig. 6 Influence of adsorbent mass on extract adsorption.



Fig. 7 Influence of the extract concentration.

It can be seen from Fig. 7 that the amount of essential oil extracts absorbed increases with the amount of adsorbent. This can be explained by the fact that by increasing the mass of adsorbent, there is a decrease in the number of active adsorption sites due to the superposition of the different adsorbent particles and by ricochet that of the adsorbed quantity. Essential oil extracts. This result could also be justified by the phenomenon of normalization of the adsorbed quantities. The less saturated the medium is with the particles of the red mud, the more easily the molecules of the oil extract migrate to the adsorption sites.

From this kinetic study, it emerges that the mixture of ratio 30/70 (Eu/Li) exhibits a strong adsorption during 24 h; this will probably make it possible to formulate a bio-insecticide against insect pests of foodstuffs.

## 4. Conclusion

The study of the adsorption of limonene and eugenol extract by the red bauxite mud is a contribution to the formulation of a bio-insecticide. The general objective of this work was to develop a bio-composite from the red mud of bauxite and extracts of limonene and eugenol. The physicochemical properties of the precursors namely red mud of bauxite, limonene and eugenol allowed us to understand that their association would have insecticidal properties. The powder formulation of Eu/Li combination (30/70) is the most effective with a fatality rate of 70.23; 86.78; 87.70; 91.45; 95.88% respectively for concentrations 1, 2, 3, 4 and 5% for 8 days. The results show that corn preserved from our bio-composite gradually loses quality in the order of 13.63% carbohydrate content, 16.14% fat, 14.90% ash and 12.50% protein over 180 days. As a perspective, it is necessary to make a toxicological evaluation, conduct an in-depth study on the adsorption of extracts and test the insecticidal power of bio-composites on other insect pests of post-harvest foodstuffs.

## Data Availability

The data used to support the findings of this study are included within the article.

# **Conflicts of Interest**

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

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