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Kinetics Release of *Cymbopogon Citatrus* Essential Oil from Starch Beads: For Food Security in Nigeria

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Abstract

Purpose: The essential oil of *Cymbopogon citratus* was extracted using a green method (i.e. essential oil still), encapsulated into cassava starch matrix using sodium alginate as a binder.

Materials and methods: The encapsulated sodium alginate starch beads were characterized using Scanning Electron Microscope (SEM) and Fourier Transform Infrared Spectrophotometer (FTIR), the release kinetics of the oil was monitored by UV-Visible Spectrophotometry from two (2) aqueous media, buffer of pH 4 and pH 9, and analysed before exploring their potential for insecticide and repellant property and controlled release.

Findings: The result for SEM showed the sodium alginate starch beads were spherical in shape, average radius of 5 µm, modal area of 0.10 μm^2 and average size of 852.28 nm and were good absorbent of essential oil in which its volatility can be entrapped. Release kinetics showed its maximum peak at 2 hours and data obtained spectrophotometrically were further fitted to the various Kinetics Models such as First Order, Pseudo Second Order, Zero Order and Higuchi Order Kinetics Model from which their equations gave (R^2= 0.9991, 0.9188) (K= 0.1547, 0.0738), (R^2= 0.9962, 0.9201) (K= -0.0071, -0.0038), (R^2= 0.9929, 0.8059) (K= 0.0065, 0.0247) and (R^2= 0.8389, 0.7216) (K= 0.033, 0.0392), respectively, from the two (2) aqueous media, buffer of pH 4 and pH 9, and it followed the First Order Kinetic Model, which indicated the absorption of the encapsulated essential oil into the porous material of the sodium alginate starch beads and its FTIR showed the functionalities such as OH, CO, CHO, C=C, =C-H and C-H present conformed to the structure of citral, starch and algin.

Implications to Theory, Practice and Policy: The green method of the extraction of essential oils as shown in Appendix viii (i.e. the locally made essential oil still) should be improved upon to ease extraction of essential oil from barks, roots, seeds and leaves of plants.

Keywords: *Cymbopogon Citatrus, Kinetics, Starch Beads, Oil*

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1.0 INTRODUCTION

According to FAO (2002), a pesticide is defined as any substance or mixture of substances used in controlling pests. They are substances that can be apply to animals to prevent insects, and other parasite in or on their bodies. Pests are vectors of animal disease, unwanted species of plants or animals causing harm during production, processing, storage, transport. In other words, pests destroy agricultural commodities, wood products and animal feedstuffs (FAO, 2002). Lemon grass (*Cymbopogon citratus*), is a perennial plant which has long thin leaves, it is a medicinal plant and because of its essential oils, is largely cultivated in parts of tropical and subtropical areas of Asia, Africa and America (Chanthal *et al*., 2012). The partly dried lemon grass contains 1-2 % essential oil. The chemical composition of the essential oil varies widely upon genetic diversity, habitat and agronomic treatment of the culture (Paviani *et al*., 2006). The leaves of lemon grass possess a lemon-like odour due to its main content, citral which presents great importance to the pharmaceutical industry. Citral has two isomers known as neral and geranial. These two isomers are used as raw material for the production of ionone, vitamin A and beta-carotene (Carlson *et al*., 2001; Wang *et al*., 2022).

Essential oils are extracts of various plant materials, which can be sourced from flowers, herbs, trees and various other plant materials. It has been estimated that about 10 % of plant species contain essential oils in quantities that could be utilized as sources of their production (Ashgari *et al*., 2010; Vieira, *et al*., 2020). Essential oils are formed by combining diverse and complex volatile mixtures of chemical compounds, in which terpenes with aldehyde, alcohol, and ketone moieties predominate (Tajidin *et al*., 2012; Tian *et al*., 2022). These essential oils are accumulated in various structures plants. The typical method of extraction of essential oils from partly dried and fresh leaves of lemon grass is by hydro-distillation (Tian *et al*., 2022).

Release kinetics is the monitoring of the concentration effects (either positive or negative) of substance(s) on living organism(s) overtime. It is seeing how the concentration of that particular named substance(s) could affect positively or negatively the existence or the life span of the so named organism(s) overtime (Tian *et al*., 2022). In this work, the effect of essential oil extracted from *Cymbopogon citratus* on pests, will be monitored. Considering the reliance of many farmers on pesticides, and the increasing cases of fake and toxic products in the market. The aim of the work was to study the release kinetics of *Cymbopogon citratus* essential oil from starch beads.

2.0 MATERIALS AND METHODS

Materials

Cymbopogon citratus leaves were collected from various homes randomly in Adoka District Otukpo LGA, Benue State. The plant samples were freshly cut 10 cm from the root, in the morning. Essential oil still, Peristaltic pump (welch)-A Gardner Denver Product Model 3200, IKA High shear Mixer-Vortex Genie-2, Buchner funnel, conical flasks, volumetric flasks, beakers, stop watch, separating funnel, weighing balance-METTLER TOLEDO AB5A-S, UV Spectrophotometer-Jenway 7305, FTIR-8400S Fourier Transform Infrared Spectrophotometer, and Scanning Electron Microscope (SEM)-10KV-Image BSD Full. Deionised or distilled Water, Anhydrous sodium sulphate, Starch, Alginic acid, Calcium chloride ($CaCl_2$), and already made commercial Buffer Tablets.

Methods

Steam and Hydro-Distillation

Two kilograms of freshly or partly dried lemon grass (*Cymbopogon citratus*) leaves were put in the cooking chamber of the distiller while water in the heating chamber and then heat was applied. The water boiled at the temperature of 100 ºC. At this stage, the steam generated passed through the leaves and extracted the oil contained in the leaves, and then the steam together with the oil moved to the condenser and the condenser re-liquefied the mixture to make the oil float on the top. The water and the oil (in a sol) were separated by the separator (separating funnel). The water portion is referred to as hydrosol, hydrolat, flower or floral water, and the oil portion is "the essential oil" (Ashgari *el al*., 2010).

Encapsulation in Starch Beads

Zero point three millilitres (0.3 mL) of the essential oil of lemon grass was added to aqueous slurry of 15 g cassava starch into which 1 g of alginic acid has been added. The slurry was stirred for 30 min using a high shear mixer until it was homogeneous. Starch beads of the slurry were produced using a peristaltic pump, the beads dropped into 1 % $CaCl_2$ solution (i.e. 1 g of $CaCl_2$ in 10 mL of distilled water) as they formed. The beads were allowed to set in the $CaCl_2$ for 10 min and filtered by vacuum through a Buchner funnel. These beads were left to air dry in a desiccator for 7 days and weighed. This experiment was carried out in the Centre for Agrochemical Technology Laboratory, Federal University of Agriculture Makurdi, Benue State.

Release Kinetics

Five grams of the dried starch alginate beads were introduced into 200 mL buffer (pH 4 and 9) maintained at room temperature. Aliquots of 3 mL of the solution were withdrawn at pre-determined times and replaced by fresh buffer to maintain sink conditions. The withdrawn samples were analysed for essential oil concentration using UV spectrophotometry at the pre-determined maximum wavelength of 650 nm. These measurements were carried out in triplicate.

Starch Beads Characterization

The starch beads were evaluated for SEM, FTIR and size and uniformity.

3.0 FINDINGS

The under listed results comprise the UV Spectrophometery of the Release Kinetics of the Sodium Alginate Starch Beads. While the SEM and FTIR results of the same Sodium Alginate Starch Beads are captured in the appendices. In Table 1, the serial dilution of the extracted essential oil of lemon grass using essential oil still in appendix viii was done and run in UV spectrophotometer to obtain various absorbances which was then processed in the MS Excel to give standard curve in Figure 3. The withdrawn samples at predetermined wavelength of the release from the two media pH 4 & 9 at different time intervals were also run in the UV spectrophotometer to obtain various mean absorbances as shown in Tables 2 & 3, respectively.

The straight line equation established from the standard curve in Figure 3 was used to calculate (Appendix ix) for various concentrations at different time intervals as shown in Tables 4 & 5 and the data obtained were further processed in the MS Excel to give parabolic type of graphs in Figures 4 & 5. The calculated concentrations in Tables 4 & 5 were further fitted to various Kinetics Models equations such as First Order, Pseudo Second Order, Zero

Order and Higuchi Model calculated and processed in the MS Excel to give the various release graphs with their correlation coefficients (R^2) and release constant (K) displayed, respectively, as shown Figures 6-13. From these Figures 6-13, the correlation coefficient (R^2) and release constant (K) values for each Model were abstracted and displayed in Table 6. The scanning electron microscopy of the dried sodium alginate starch beads was run using SEM to know their shape and size at different magnifications as shown in appendices i-vi and FTIR of the same dried sodium alginate starch beads was carried out using FTIR spectrophotometer to know their functional groups as shown in appendix vii.

Sample Calculations

Equation of straight line obtained from calibration curve on Figure 3 was used to calculate the various concentrations in Tables 5 and 6. The data obtained were used plot Figures 4 and 5, respectively. The equation: y = 0.0266x-0.0264, where y were the absorbances obtained, x the unknown concentrations. This was substituted, using this equation.

Sample calculations:

a). y = -0.0177, x =? Using the above equation, y = 0.0266x-0.0264

-0.0177 = 0.0266x-0.0264, make x subject of the formula, then

$$x = \frac{-0.0177 + 0.0264}{0.0266} = 0.327 \text{ for pH 9}$$

b). y = -0.0203, x =? Using the above equation, y = 0.0266x-0.0264

$$x = \frac{-0.0203 + 0.0264}{0.0266} = 0.229 \text{ for pH 4}$$

Table 1: Calibration Result for the Serial Dilution of Essential Oil of Cymbopogon citratus at Maximum Wavelength of 650 nm

Concentration (ppm)	Absorbance (A)
0.5	-0.0093
1.0	0.0043
1.5	0.0071
2.0	0.0121
2.5	0.0534

Table 2: Absorbances (at 650 nm) of the Release from Buffer Solution of pH 4

Absorbance(A)				Time(hr)
First	Second	Third	Mean	
-0.026	-0.024	-0.063	-0.0203 ±0.005	1
0.003	0.005	0.006	0.0046 ±0.005	2
-0.009	-0.013	-0.007	-0.0097 ±0.005	3
-0.024	-0.026	-0.034	-0.0280 ±0.005	4
-0.010	-0.015	-0.017	-0.0140 ±0.005	5
-0.001	-0.002	-0.003	-0.0014 ±0.005	6

Table 3: Absorbances (at 650 nm) of the Release from Buffer Solution of pH 9

Absorbance(A)				Time (hr)
First	**Second**	**Third**	**Mean**	
-0.016	-0.023	-0.014	-0.0177 ±0.005	1
0.030	0.027	0.042	0.0330 ±0.005	2
-0.024	-0.034	-0.022	-0.0266 ±0.005	3
-0.016	-0.015	-0.018	-0.0163 ±0.005	4
-0.026	-0.033	-0.020	-0.0263 ±0.005	5
-0.011	-0.010	-0.015	-0.0126 ±0.005	6

Table 4: Calculated Mean Concentration of Buffer of pH 4

Concentration (ppm)	Time (hr)
0.229	1
1.165	2
0.628	3
-0.060	4
0.466	5
0.941	6

Table 5: Calculated Mean Concentration of Buffer of pH 9

Concentration (ppm)	Time (hr)
0.327	1
2.233	2
-0.007	3
0.379	4
0.004	5
0.519	6

Table 6: Release Kinetic Equation Values of the Encapsulation from the Two Media

Order Release Models	pH 4	pH 9
First:		
R^2	0.9991	0.9188
K_1	0.1547	0.0738
Pseudo Second:		
R^2	0.9962	0.9201
K_2	-0.0071	-0.0038
Zero:		
R^2	0.9929	0.8059
K_0	0.0065	0.0247
Higuchi:		
R^2	0.8389	0.7216
K	0.033	0.0392

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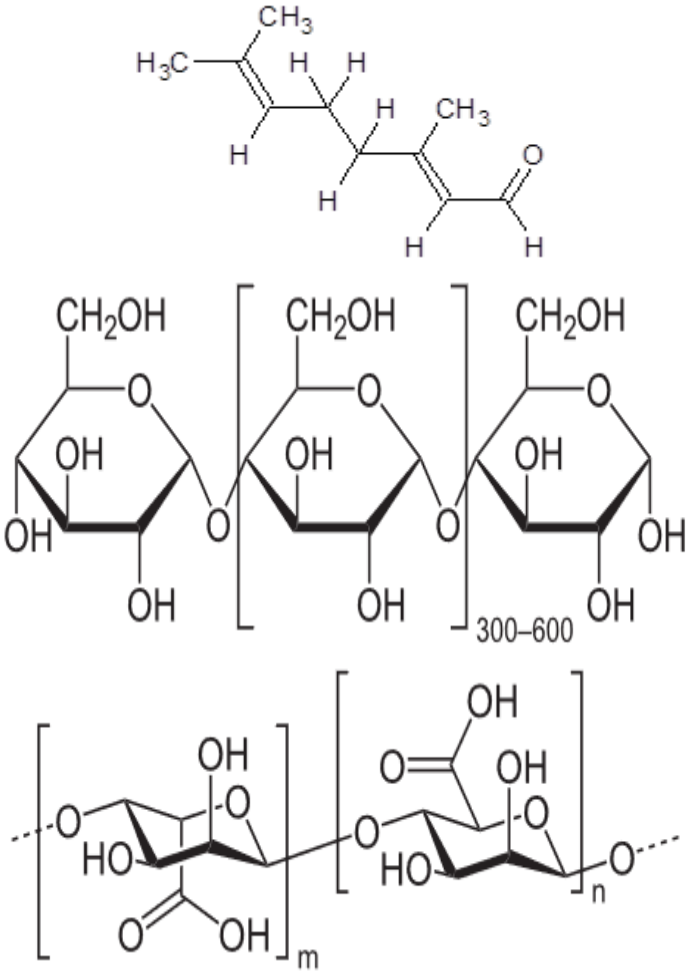

Figure 2: Chemical Structure of Citral, Starch and Algin, Respectively

m, n = 2, 3, 4, …

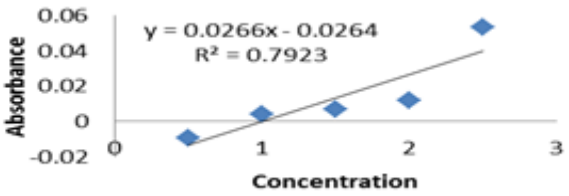

Figure 3: Calibration Curve of Essential Oil of Cymbopogon citratus

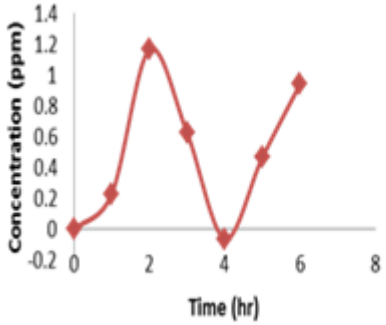

Figure 4: Graph of Release from Buffer Solution of pH 4

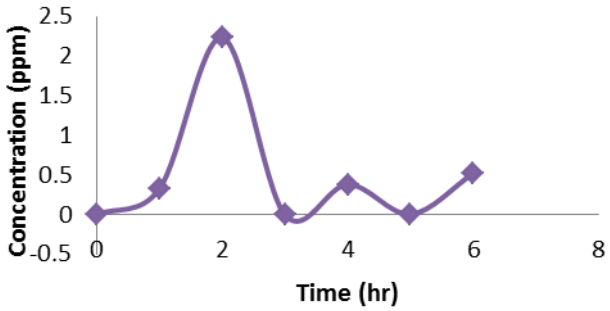

Figure 5: Graph of Release from Buffer Solution of pH 9

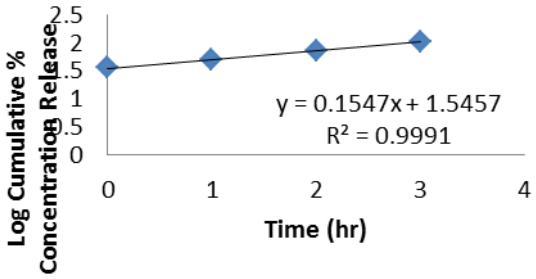

Figure 6: First Order Release Kinetics at pH 4

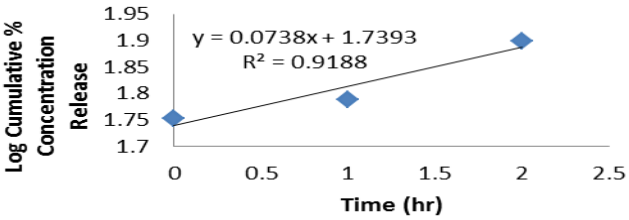

Figure 7: First Order Release Kinetics at pH 9

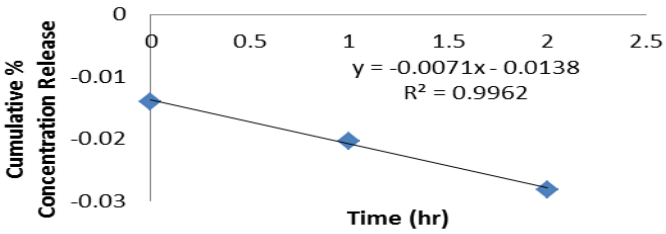

Figure 8: Pseudo Second Order Kinetics at pH 4

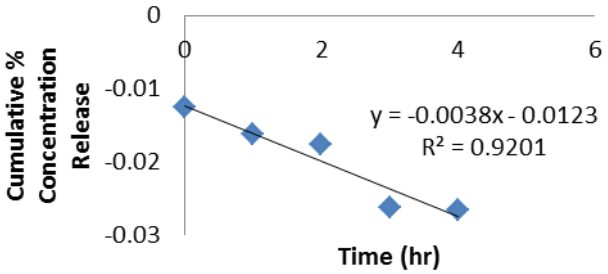

Figure 9: Pseudo Second Order Kinetics at pH 9

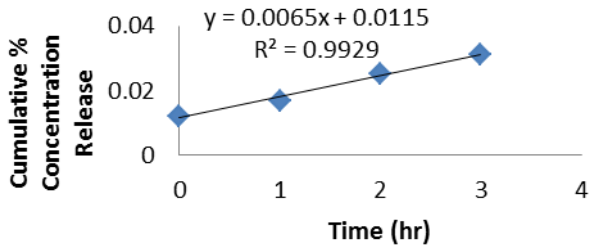

Figure 10: Zero Order Release Kinetics at pH 4

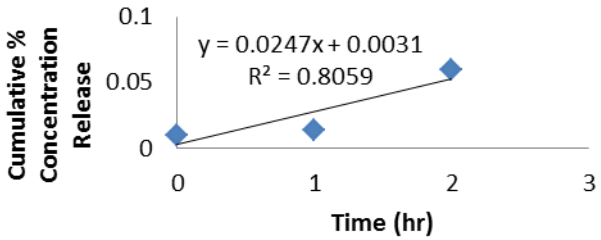

Figure 11: Zero Order Release Kinetics at pH 9

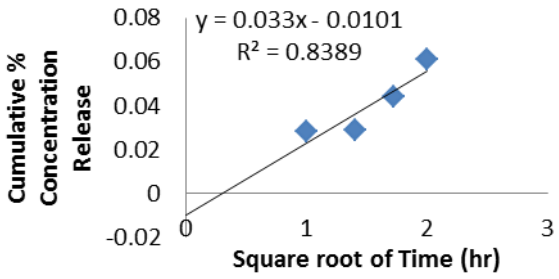

Figure 12: Higuchi Order Release Kinetic at pH 4

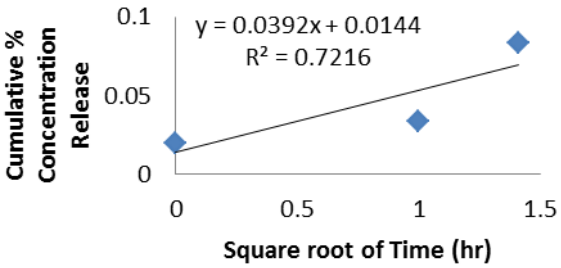

Figure 13: Higuchi Order Release Kinetics at pH 9

4.0 DISCUSSION

Scanning Electron Microscopy

Morphological examination of the surface structure of the dried Sodium alginate starch beads was carried out using Scanning Electron Microscope as shown in Appendices. Appendices ii, iii and iv showed that the starch alginate beads were spherical in shape. Higher magnification

of 3500 (Appendix v) revealed average radius of 5 µm and pentagonal creases on the faces of the spherical beads. White spots also became visible at this higher magnification. These may be due to some starch particles not fully gelled by the sodium alginate. From Appendices i and vi, the starch alginate beads have a modal area of 0.10 μm^2 and average size of 852.28 nm

Size and Uniformity

The sizes of the beads were not the same and uniform due to homogenization, Viscosity, rate of falling of drops, stirring rate and distance between syringe and gelation medium were constantly maintained.

Release Kinetics

The information in Table 1 was processed in the MS excel to give a calibration curve as shown in Figure 3. R^2 value shows perfection of the serial dilution from the stock sample of essential oil of *Cymbopogon citratus* according to Beer-Lambert's law. The straight line equation established here was used to calculate for the mean concentration of the oil content of the 1st, 2nd, and 3rd absorbance from the two (2) media, buffer of pH 4 and pH 9 as shown in Table 2 and 3. The calculated mean concentration of the oil content from the two (2) media were processed in the MS excel which give a parabolic shape of graph as shown in Figures 4 and 5, which indicated the release of the essential oil in aqueous buffers pH 4 and 9 rose gradually to a peak at 2 hours after which there was a drop in amount released. This is similar to release of active ingredient from matrices reported by (Hina *et al*., 2015).

The data were fitted to the Kinetic Models such as First Order Release, Pseudo Second Order Release, Zero Order Release and Higuchi Order Release with the various equations $\log c = \log c_0 - kt/2.303$, $t/q_t = 1/k_2q^2_e + t/q_e$, $Q_t = Q_0 - K_0t$ and $Q = A\sqrt{D(2C-Cs)Cst}$ respectively, calculated and processed in the MS EXCEL as shown in Figures 6-13, gave the various Correlation coefficients (R^2) and Rate constant (K) as shown in Table 6. From the Correlation Coefficient Applicability according to Beer-Lambert's law, the values for ($R^{2)}$ were high enough to explain the release which followed the First Order Release Model, with the various values (R^2 = 0.9991, 0.9188) and (K= 0.1547, 0.0738) respectively from the two (2) media buffer of pH 4 and pH 9, which may indicate the absorption of the encapsulated essential oil into the porous material of the sodium alginate starch beads (Hina *et al.*, 2015).

FTIR of the Sodium Alginate Starch Beads as shown in Appendix vii

i. There is a strong broad band absorption at 3436.30 cm^{-1} which indicated the presence of OH stretch, H-bonded of alcohols or phenols.

ii. Near to this broad band, there is C-H band absorption at 2927.08 cm^{-1} in which to its right there two (2) weak absorption bands at 2397.60 cm^{-1} and 2274.15 cm^{-1} and these are caused by the C-H bond that is part of the CHO aldehyde functional group.

iii. There is a strong absorption band at 1641.46 cm^{-1} which showed or indicated the presence C=O stretch of α, β unsaturated aldehydes

iv. There is a strong absorption band at 1018.45 cm^{-1} which indicated the presence of =C-H bend.

v. There is a medium absorption band at 1431.23 cm^{-1} which indicated the presence of C-H bend.

Source https://en.wikipedia.org/wiki/Infrared_spectroscopy_correlation_table

5.0 CONCLUSION AND RECOMMENDATION

Conclusion

The FTIR characterisation of the sodium alginate starch beads conforms to the chemical structure of citral, starch and alginic acid, since all the functionalities such as OH, CO, CHO, C=C, =C-H and C-H of the compounds (citral, starch and algin) are present. SEM images of the dried sodium alginate starch beads shows they are spherical in shape, average radius of 5 µm, modal area of 0.10 μm^2 and average size of 852.28 nm. The pore-histogram shows that the beads are "good absorbent" of essential oils in which the volatility of essential oils can be entrapped.

Finally, the release kinetics of the sodium alginate starch beads was at its maximum peak at 2 hours and followed first order release kinetics model which indicated the absorption of the encapsulated essential oil into the porous material of the sodium alginate starch beads.

Recommendations

The green method of the extraction of essential oils as shown in Appendix viii (i.e. the locally made essential oil still) should be improved upon to ease extraction of essential oil from barks, roots, seeds and leaves of plants. The pesticidal activity of the encapsulated sodium alginate starch beads should be carried out by other researchers to ascertain its effectiveness on pest.

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