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Chitosan active coating on paperboard surface forming an anti-insect grain-based food packaging

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In the food chain, the packaging is an intermediary product between the food industry and consumers. It represents a critical step in food quality preservation and the ultimate protection against insect pests. Insects may infest grain-based food products during their packed life. Active packaging systems involve intentional interaction with food or its surroundings, and a few studies have been focused on anti-insect packaging materials for food. The aim of this work was to develop a sustainable and active packaging material based on chitosan coating on paperboard surface incorporating lemongrass essential oil with potential action against weevil infestation in cereal-based food packed products. The innovation is the eco-friendly packaging development by applying natural and biodegradable compounds to prevent stored product insect infestation in the grain-based product. The effects of chitosan and lemongrass essential oil (LG) concentrations and the number of coating layers were studied. The active packaging material was anti-insect efficient against adult weevils in wheat grain and pasta package. The active coating reduced the air and water vapour transmission rate, and water absorption capacity improved the fat barrier while maintaining a microbial impermeability. The proposed active packaging material presented a potential application to extend the shelf life of grain-based food products against weevil infestation.

KEYWORDS
active packaging, anti-insect packaging, chitosan, weevil

1 | INTRODUCTION

Paper and paperboard are biodegradable and renewable materials commonly used as food packaging and are considered susceptible to insect attacks.[1] Bio-based coatings can be a safe and sustainable alternative to improve mechanical and barrier properties.[2] Chitosan is a cationic polymer obtained from the alkaline deacetylation of chitin. Chitosan comprises a linear sequence of monomeric sugars β-(1,4)-2-acetamido-2-deoxy-D-glucose (*N*-acetylglucosamine) and glucosamine, characterized by molecular weight (Mw) and degree of acetylation (DA).[3] Incorporating lipid molecules into a chitosan matrix, forming an emulsion, could improve barrier and resistance to water.[4] Chitosan could be used as an additive or coatings in the papermaking process. The chitosan structure similarity with cellulose favors the strong bonding, improving the mechanical, electrical, printing, barrier, and antibacterial paper properties.[5] Chitosan presents a cationic character, while cellulose presents an anionic character, in which there is an attraction between the polymers, providing good adhesion between chitosan coating and cellulose surface.[6] The amino groups in chitosan can form hydrogen bonds

with the hydroxyl group of cellulose fibres giving extra strength to paper.[7] The easy biodegradability of chitosan films is an advantage as compared with synthetic polymers.[8] A study involving chitosan coating noticed an improvement in the barrier properties of the paper-based matrix without decreasing the inherent biodegradability of paper.

The biodegradability of the polymers depends on the molecular structure, length of the polymer chain, crystallinity, and the complexity of the polymer formula. Furthermore, temperature, oxygen supply, and test duration can also influence the biodegradability of the system.[9] The effect of chitosan coating on cellulose biodegradation was significant, indicating that chitosan film and emulsified chitosan-based coatings acted as a substrate for microorganisms, inducing their growth.[10] Fast biodegradation of Kraft paper coated with chitosan solution was demonstrated by Kato et al.[11] Moreover, it demonstrated that the DA could impact the biodegradation process where lower Mw and DA improved the biodegradation.[12]

Active packaging refers to incorporating active compounds into packaging material to maintain the quality or extend the shelf life of food products. The development of active anti-insect packaging materials is an innovative technology for food preservation during storage, incorporating botanical insecticides. Infestations of insects are highly unpleasant for the consumer. The stored food products of agriculture are affected by different species of coleopteran beetles causing quantitative and qualitative losses, altering the final quality control and nutritional and market values.[13] The weevil *Sitophilus zeamais* (Motschulsky, 1885) (Coleoptera: Curculionidae) is the main worldwide pest with economic relevance to stored products with significant losses of stored grains such as wheat, rice and maize.[14] It is one of the main stored food pests that can affect cereal-based dry foods, such as pasta, rice, flour, cake, and others, causing economic impacts due to product rejection by manufacturers and consumers.[15] The insect attack can occur during the production or distribution (transportation and storage). The control infestation depends heavily on synthetic insecticides, which cause environmental damage, the resistance of insect populations to chemicals of different toxicological classes, chemical food contamination[16] and health risks.[17]

The eco-friendly alternatives to synthetic pesticides are active compounds secondary metabolites associated with a defensive or protective function against insect attack, classified as botanical insecticides.[18] It is an important phytosanitary protection strategy, well accepted by food regulators and the general consumer public.[13] The botanical insecticides include alkaloids, phenolics, polyacetates and terpenoids. The mechanism of action depends on the active compounds, which could be toxic or repellent to the target organisms and cause changes in sterility, have an antifeedant and growth inhibitory effect or alter the insect's behaviour.[19] Lemongrass *Cymbopogon citratus* Stapf is an herb widely used in tropical countries. It was verified antioxidant, bactericidal, fungicidal, antidepressant, antiseptic, astringent, nervine and sedative properties of LG.[20] LG is composed mainly of monoterpenes of remarkable insecticidal efficacy, among them citral, and has proven successful against *S. zeamais* weevil and most grain pests.[21] Different active anti-insect bioactive compounds were studied, such as turmeric extract[22]; citronella, oregano and rosemary essential oils against *Tribolium castaneum*[13]; biopesticide (1-octen-3-oil) in polyethylene films against *S. zeamais*[23]; eugenol with insecticide/insectifuge in paper-based active packaging using polycarboxylic against *T. castaneum*.[1]

This work aimed to develop, evaluate the anti-insect efficiency and characterize an innovative, sustainable and active chitosan coating paperboard packaging material containing LG against one of the worldwide pests stored grain-based food products, *S. zeamais*.

2 | MATERIALS AND METHODS

2.1 | Materials

S. zeamais adults from the Laboratory of Entomology and Acarology, University of São Paulo (Brazil). Paperboard duplex (240 $g.m^{-2}$) from Suzano Papel e Celulose SA (Brazil). Chitosan ChitoClear, degree of deacetylation 95% e $Mw = 1.26 \times 10^5$ $g.mol^{-1}$ (Primex, Iceland). Acetic acid PA (Synth, Brazil) and lemongrass (*C. citratus*) essential oil (Quinarí, Brazil).

2.2 | Insect rearing

Weevil generations were kept in glass pots (3 L) with perforated polyethylene cover and internally lined with voile. Organic wheat grains (200 g) were decontaminated by freezing and were used as food and substrate for weevil oviposition. Climate-controlled temperature (25 ± 2°C, 60 ± 10% relative humidity [RH] 14:10 h L:D photoperiod) in an incubator BOD (model TE391, Tecnal, Brazil). Adult weevils were confined to wheat grains in glass pots for 15 days and removed from the oviposited wheat grains and kept in a new glass container and stored under the same controlled conditions until the emergence of a new insect generation, 10–20 days old, to assure the number of adults of adequate age in the bioassays.

2.3 | Active coating paperboard

Chitosan filmogenic suspensions were prepared in aqueous acetic acid added stoichiometric[24] and kept under continuous magnetic agitation for 1 h (IKA, Topolino, Germany) at 150 rpm. The LG was added (%, v/w, based on the total weight of the emulsion) under rigorous homogenization at 20 000 rpm for 10 min (Ultra-Turrax IKA T25-Digital, Staufen, Germany). The paperboard sheet was coated with 3 g of film-forming solution, per layer, at a speed of 10 $mm.s^{-1}$ on the automatic spreader (Zehntner®, Switzerland). The coated paperboard was dried at 120°C for 90 s in an air convection oven (MA 035/100, Marconi, Brazil).

2.4 | Factorial experimental design

A factorial experimental design (2^3) was adopted to determine the influence of three independent variables, namely, chitosan concentration (C_{chi}), lemongrass essential oil (C_{oil}) and the number of layers of coating (Cl_{ayer}), on an active chitosan–LG paperboard system formation. Independent variable levels were chosen from preliminary studies (Table 1). The response of factorial experimental design evaluated were anti-insect efficiency, total solids, thickness, grammage, coating homogeneity, microstructure, water vapour transmission rate, air permeance, water absorption capacity, grease resistance and microbial permeance. Uncoated paperboard was used as a control sample.

2.5 | Sample preconditioning

Samples of uncoated and coated paperboard were previously conditioned at (25 ± 2°C) and 50 ± 2% RH before being analysed according to ASTM D 685-93.[25]

2.6 | Characterization

2.6.1 | Coating evaluation: Coloured solution penetration

The methodology described by Marcy[26] was adapted. Erythrosine solution (0.5%, w/v, in isopropanol) was applied to uncoated (matte side of paper) and coated paperboard samples (coated side) covering the entire surface (150 × 200 mm) with cotton and metallic tweezers. Samples were held upright for 60 min and dried at 50°C for 30 min in an air circulation oven (MA 035/100, Marconi, Brazil). The specimens were visually examined on the opposite side of the coating. The analysis was performed in triplicate.

2.6.2 | Scanning electron microscopy

Uncoated and coated paperboard were cut in square shape 1 × 1 cm and kept for 48 h at 25°C in a desiccator containing silica. The surface and cross-sectional microstructures of the active systems were analysed.[27] The metallic coating was done on a Sputter Coater Model K450 (Emitech, France), gold layer thickness estimated at 200 Å. Microscopic images of samples were obtained using an LEO 440i scanning electron microscope (LEO Electron Microscopy, Oxford, Cambridge, England) at 15 kV.

TABLE 1 Values of coded levels of independent variables used in factorial experimental design: concentration of chitosan (C_{chi}), lemongrass essential oil concentration (C_{oil}) and the number of layers (C_{layer}) forming active chitosan–oil paperboard material

	Level		
Independent variables	**−1**	**0**	**+1**
C_{chi} (%, w/w)	1	2	3
C_{oil} (%, v/w)	20	30	40
C_{layer} (unit)	1	3	5

2.6.3 | Moisture content

The moisture content evaluation experiment was executed in triplicate. The coated and uncoated paperboard sheets were cut into a square of dimensions 1 × 1 cm and dried at 105 ± 2°C in a TE 393 forced air circulation oven (MA 035/100, Marconi, Brazil) until constant weight according to the standard method ASTM D 644-99.[25] The moisture content *Xbs* (g H_2O.100 g^{-1} paper) was calculated according to Equation 1. M_u and M_s are humid and dried weight, respectively.

$$Xbs = \left(\frac{M_u - M_s}{M_s}\right) \times 100. \quad (1)$$

2.6.4 | Evaluation of anti-insect efficiency

The bottom of the plastic Petri dish (6.1 cm in diameter) was covered with the active paperboard material[28] and added decontaminated organic wheat grains (10 g). After loading the recipients, 20 unsexed adult weevils (10–20 days old) were released into a plastic Petri dish and stored in an incubator BOD (25 ± 2°C, 60 ± 10% RH, 14:10 h L:D photoperiod, TE 391 Tecnal, Brazil).[29] Anti-insect efficiency was evaluated periodically during 360 h (15 days) by counting the dead weevils. Ten replications were performed for each treatment. The anti-insect efficiency was calculated as presented in Equation 2.

$$\%Anti-insect\ efficiency = \frac{final\ dead\ weevils}{initial\ live\ weevils} \times 100 \quad (2)$$

2.6.5 | Total solids, thickness and grammage

Total solids were estimated by active solution weight (g) in the coating area (m^2). The average thickness (μm) was calculated by 10 replicates using a digital micrometer Mitutoyo (MDC-25M, Japan). For each sample, six measurements were carried out. The grammage ($g.m^{-2}$) determination was the average of 10 replicates. The coated and uncoated paperboard samples (12.5 × 12.5 cm) were weighed on an analytical balance (Shimadzu®, AUY220, Japan), according to ASTM D 646-96.[30] Grammage *G* ($g.m^{-2}$) was calculated by Equation 3, where *M* (g) is paperweight and *A* (m^2) is the area.

$$G = M/A \quad (3)$$

2.6.6 | Water vapour transmission rate

The water vapour transmission rate (WVTR) was determined based on ASTM E 96/E96M-16.[25] Five coated and uncoated paperboard samples were cut in a disc shape and fixed on the top of permeation cells containing silica gel. Cells were conditioned in desiccators at 50 ± 5% RH and kept in a temperature-controlled chamber (25.0 ± 0.2°C). Mass gain of the system (cell and sample) was determined at 24-h intervals over 120 h using an analytical balance (Shimadzu®, AUY220, Japan). WVTR (g $H_2O.m^{-2}.day^{-1}$) was calculated by Equation 4, where G (g) is the weight gained by the system, A (m^2) is the area exposed to vapour transmission and t (day) is incubation time.

$$WVTR = G/(A.t) \quad (4)$$

2.6.7 | Air permeance

The air permeance was determined based on ASTM D 726-94.[25] The uncoated and coated paperboard samples (preconditioned in a chamber at room temperature (25 ± 2°C) and 50 ± 2% RH) were fixed in the porosimeter (air pressure 1.47 kPa) and the time required to pass 100 ml of air through the paper surface in Gurley-type apparatus (model PGH-T, Regmed®, Brazil) was measured. Five replications were performed, and the results were expressed in $\mu m.Pa^{-1}.s^{-1}$.

2.6.8 | Water absorption capacity (Cobb test)

Cobb test was executed following ASTM D3285-93.[25] Ten samples (12.5 × 12.5 cm) of uncoated and coated paperboard were preconditioned (described in Section 2.5). Samples were individually weighed on a semi-analytical scale with a precision of 0.01 g and attached to the equipment for Cobb (Regmed, Brazil). Distilled water of 100 ml were added in contact with the surface delimited by the apparatus ring (internal diameter of 11.28 ± 0.02 cm) for 120 s. Soon after, the specimen was removed. The excess water was removed from the surface, placing it between two sheets of absorbent paper and passing a conditioning roller (Regmed, Brazil) on top. Samples were weighed using an analytical balance (Shimadzu, AUY220, Japan). Water absorption capacity (Abs, $g.m^{-2}$) was determined by Equation 5. M_f and M_i (g) are final and initial sample weights, respectively.

$$Abs = \frac{(M_f - M_i)}{A} \times 100 \quad (5)$$

2.6.9 | Fat barrier

The methodology was based on Ham-Pichavant et al.[31] Uncoated and coated paperboard samples were tested with a series of solutions (Kit numbers 1–12) containing specific proportions of three reagents: castor oil, toluene and *n*-heptane. One drop of test solution was applied to the paper sample surface and kept for 15 s. The excessive solution was removed, and the appearance of the opposite surface was observed. The adopted value of fat repellence was the solution with the highest kit number that did not cause blemishes. Kit n°1 and Kit n°12 are the least and the most aggressive solutions for producing a stain on the opposite surface.

2.6.10 | Microbial permeation

Uncoated and coated paperboard discs (4.6 cm diameter) were sterilized by UV light exposure for 15 min on each side and fixed at the top of sterilized permeation cells with sterile TSB broth (Tryptic Soy Broth Soybean-Casein Digest Bacto®).[8] The positive control was the permeation device completely sealed with parafilm, and the negative control was the permeation device without a seal. The permeation devices were incubated at room temperature (25 ± 5°C) for 15 days. The visual turbidity of the TSB media indicated the microbial permeation through the active paperboard system. Four replications were performed.

2.6.11 | Consumer acceptability test

The active chitosan–LG paperboard system that provided the best anti-insect efficiency (1% chitosan (w/v), 40% LG (v/w) and 5 layers) was chosen to coat the inner surface of pouch paperboard that was used as anti-insect grain-based food packaging. *Talharim* pasta was packaged in pouch paperboard uncoated (control) and coated with the active formulation. Packages were stored at controlled room temperature (25 ± 2°C) and RH (50 ± 5%) for 40 days. The shelf life of 40 days was based on the previous experiment of bioactivity of the coating as an anti-insect effect on living *S. zeamais* weevils carried out in the first coating tests. The biological cycle of the eggs until adults' emergence is approximately 34 days.[32] Pasta from each treatment was evaluated by untrained and healthy consumers ($n = 116$). Potential consumers (age: 18–59; gender: male and female) were recruited from students and staff from the Federal University of São Paulo based on their frequency of pasta consumption. No participant had any known food allergy. At the entrance to the testing area, the panellists were informed of all details of the study, signing the Informed Consent Form. Pasta samples were prepared according to typical cooking recommendations for the commercial pasta products. Samples were removed from pouch paperboard and were immersed in salted boiling water for 7 min.[33] The pasta was then removed, mixed with butter, placed in styrofoam cups and immediately served 20 g per sample to

panelists. The pasta samples were coded with random three-digit numbers and presented according to a balanced complete block design. The presentation order was randomized for each panellist. Mineral water and unsalted crackers were provided to the panellists to palate cleansing between sample tastes. The sensory attributes of odour, flavour and overall impression were evaluated using a 9-point hedonic scale (1: *dislike very much*, 9: *like very much*). It was also informed to the panellists of a second part of the test: a purchase intention test, performed by using a 5-point scale ranging from 1 = *would certainly not buy* to 5 = *would certainly buy*. Panellists could add any additional comments. This study obtained a favourable ethical opinion from the Research Ethics Committee of Federal University of São Paulo (CAAE: 73690617.3.0000.5505).

2.7 | Statistical analysis

Statistical analyses were carried out using Statistica® software 12.7 (Statistica®, USA). Differences between the averages were identified by ANOVA and Tukey's test ($p < 0.05$).

3 | RESULTS AND DISCUSSION

The active chitosan–LG coating was characterized by visual uniformity and homogeneity on the paperboard surface in all formulations studied in the factorial experimental design. After vigorous handling, no delamination of the layers was observed, which indicates good adhesion and compatibility between the active coating and the paperboard. The homogeneity and stability of the chitosan–LG emulsion could be associated with the electrostatic interactions in the interfacial area between the anionic lipid molecules and cationic groups of chitosan glucosamine residues.[31] Higher rigidity was observed with increasing chitosan concentration in the film-forming coating solution.

The chitosan emulsion coating was observed in scanning electron microscopy (SEM) images of active coated paperboard (Figure 1). The heterogeneous surface of the uncoated paperboard was characterized by the porous structure of the cellulose fibre interlacement (Figure 1A). Applying only one layer ($C_{layer} = 1$), the active coating solution filled the void spaces between the cellulose fibers (Figure 1C, E,G,I). Increasing $C_{layer} = 5$, it was possible to verify the film formation on the paperboard surface (Figure 1B,D,F,H). A more homogeneous surface was obtained by increasing the C_{chi} from 1% to 3% (w/w), related to the higher solids deposited on the paperboard surface. The increment of C_{oil} from 20% (Figure 1D,E,H,I) to 40% (v/w) (Figure 1D, E,H,I) changed the surface coating microstructure, promoting an amorphous structure. The micrographs indicated porous and void spaces present in the coating filmogenic matrix. Similar results were observed by Yoshida et al.[4] when comparing the micrographs of chitosan and emulsion chitosan–palmitic acid films. Chitosan film coatings on bleached Kraft pulp paper sheets (74 $g.m^{-2}$) had a considerable effect by increasing three or more chitosan layers, resulting in a smooth and uniform surface-coated paper.[34] However, the chitosan did not fill the

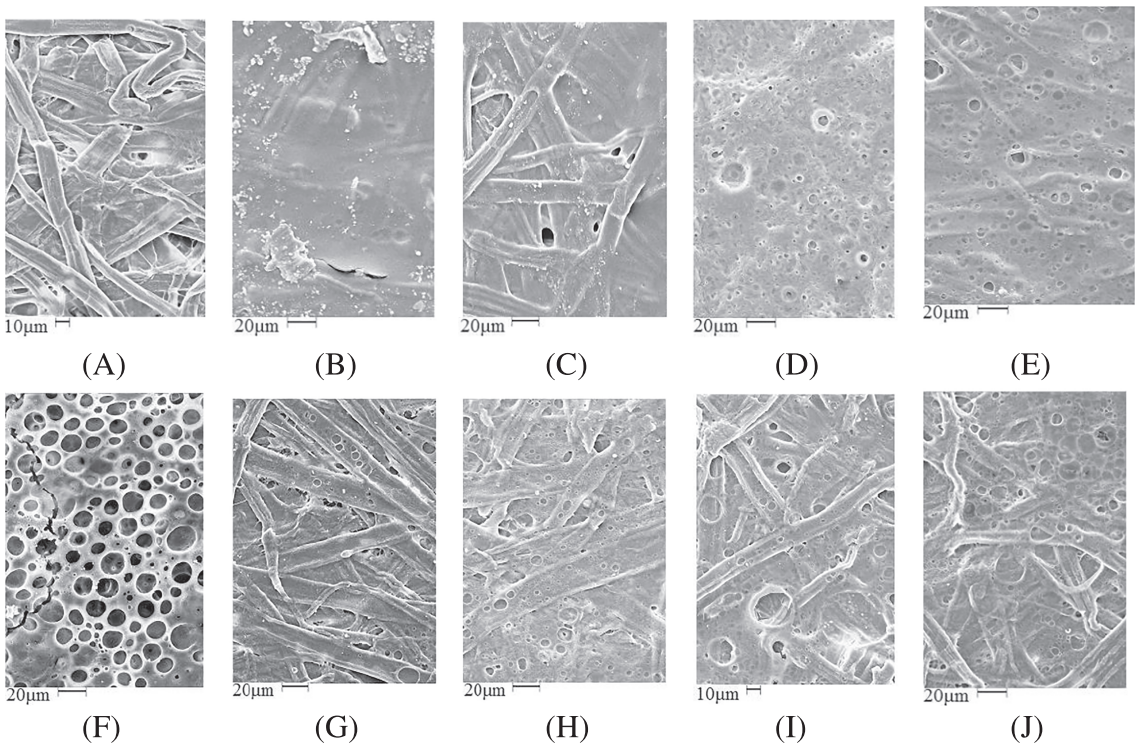

FIGURE 1 Surface micrographs of paperboard (1000×): (A) uncoated; and coated with formulations from factorial experimental design treatments: (B) Run 1; (C) Run 2; (D) Run 3; (E) Run 4; (F) Run 5; (G) Run 6; (H) Run 7; (I) Run 8; (J) Runs 9–11

paper pores in the three-dimensional structure, even after five layers.[35]

All coatings adhered well to the paperboard surface as revealed that it was not observed coloured spots on the back surface of the coated surface in all formulations (Runs 1–11) after the coloured solution of erythrosine application (Figure 2). The uncoated paper showed coloured spots on the back surface (Figure 2A). The filling of the cellulose interfibrillar spaces on paperboard by the active chitosan solution was observed, which could be associated with a good uniformity coating. The chitosan–LG filmogenic solution could fill the pores of the cellulose fibrous structure, forming a film barrier, improving paper surface properties. Similar results were obtained by Reis et al[27] in chitosan (4%, w/w) coated Kraft paper.

The coating formulations of the active paperboard systems and the final properties for each dependent variable response are shown in Table 2: moisture content, anti-insect efficiency, average thickness, grammage, WVTR, air permeance, water capacity absorption (Cobb test) and fat barrier. The total solids of coatings were estimated for each formulation. Table 3 summarizes the mean effects and ANOVA results for each dependent variable response. The anti-insect efficiency, thickness, grammage and Cobb test models were significant at 5% level ($p < 0.05$), considering $F_{calc}/F_{tab} > 1$, but they were not predictive for the conditions of this study. For a regression model to be statistically predictive, the F ratio (F_{cal}/F_{tab}) must be at least four or five times higher than the F_{tab}.[36] In our study, the main objective was to evaluate the effect of independent variables on the final properties of the developed material. The high values of lack of fit could be associated with the living biological assays, such as weevil, which resulted in statistically non-predictive models. Similar results were verified by Lee et al,[37] studying multilayered films containing different essential oils.

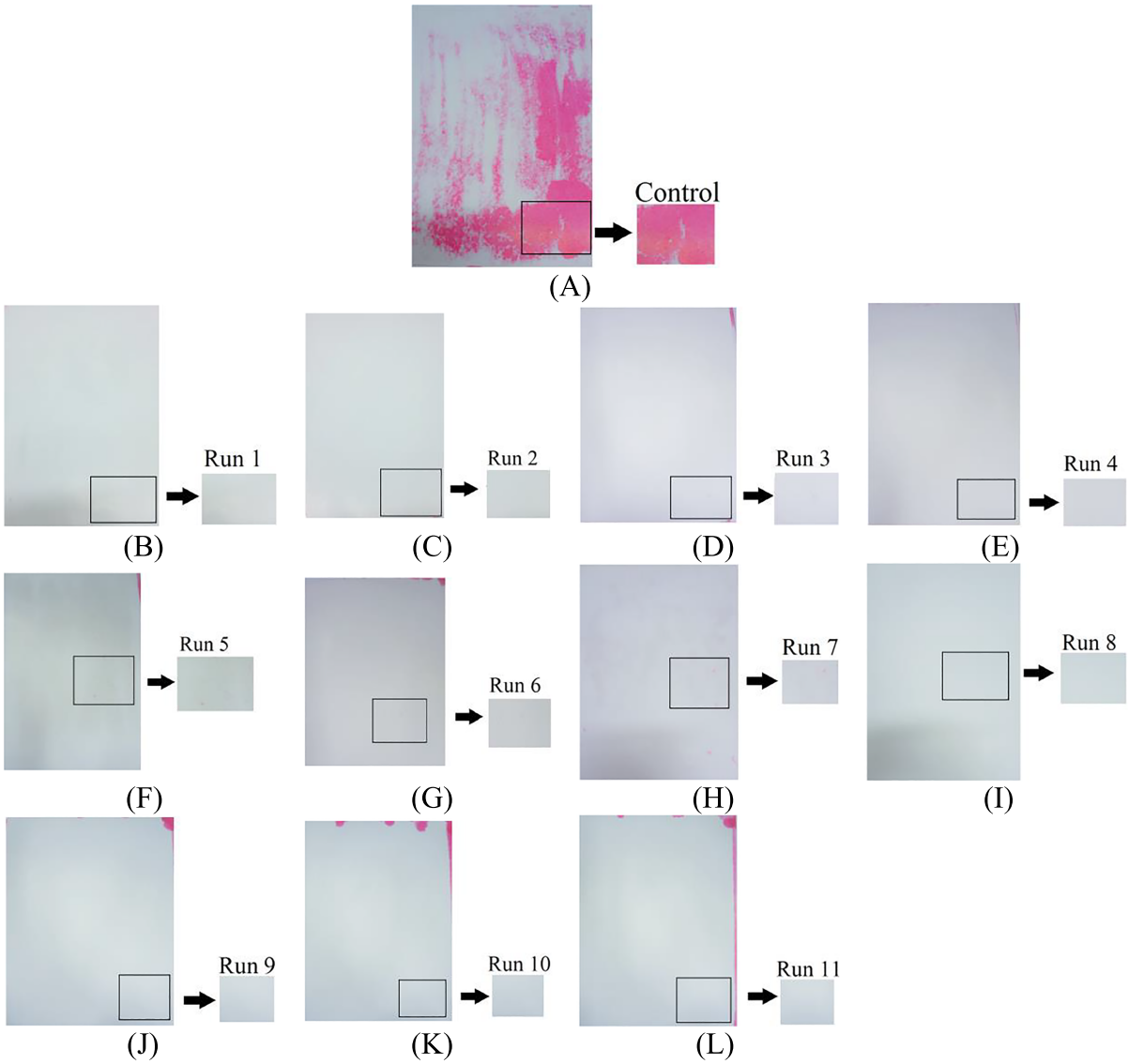

FIGURE 2 Visual uniformity coating analysis of active chitosan–oil paperboard systems based on factorial experimental design formulations: (A) control (without coating); (B) Run 1; (C) Run 2; (D) Run 3; (E) Run 4; (F) Run 5; (G) Run 6; (H) Run 7; (I) Run 8; (J) Run 9; (K) Run 10; (L) Run 11

TABLE 2 Estimated total solids, moisture content, anti-insect efficiency, thickness, grammage, WVTR, air permeance, Cobb test (water absorption capacity) and fat barrier of active chitosan–LG paperboard systems based on the formulations of factorial experimental design 2^3 + central points

	Independent variables (coded values)				Dependent variables							
Run	C_{chi}[a]	C_{oil}[b]	C_{layer}[c]	Total solids[d] ($g.m^{-2}$)	Moisture content (%)	Anti-insect efficiency (%)	Thickness (μm)	Grammage ($g.m^{-2}$)	WVTR ($gH2O\ m^{-2}\ day^{-1}$)	Air permeance ($\mu m.\ Pa^{-1}.s^{-1}$)	Cobb test ($g.m^{-2}$)	Fat barrier
1	+1	+1	+1	134	7.7 ± 0.3	78 ± 18	430 ± 10	269	252 ± 19	0.01	32 ± 7	Total barrier
2	+1	+1	−1	27	6.2 ± 0.1	66 ± 22	400 ± 0	245	256 ± 12	0.02	42 ± 6	10
3	+1	−1	+1	72	8.6 ± 0.3	61 ± 18	480 ± 30	274	238 ± 12	0.01	16 ± 3	Total barrier
4	+1	−1	−1	14	7.3 ± 0.1	1 ± 1	410 ± 0	247	332 ± 11	0.02	43 ± 4	10
5	−1	+1	+1	128	7.7 ± 0.7	100	440 ± 10	289	235 ± 28	0.02	22 ± 3	11
6	−1	+1	−1	26	7.2 ± 0.2	2 ± 1	410 ± 0	245	326 ± 11	0.21	47 ± 8	6
7	−1	−1	+1	66	6.2 ± 0.2	69 ± 19	420 ± 10	251	289 ± 9	0.02	22 ± 2	11
8	−1	−1	−1	13	5.6 ± 0.5	3 ± 1	400 ± 0	241	314 ± 7	0.28	43 ± 2	5
9	0	0	0	60	7.1 ± 0.4	30 ± 10	425 ± 10	255	289 ± 9	0.01	22 ± 4	11
10	0	0	0	60	6.9 ± 0.3	31 ± 9	420 ± 10	254	295 ± 8	0.01	26 ± 4	11
11	0	0	0	60	6.9 ± 0.5	33 ± 12	430 ± 10	254	279 ± 11	0.01	24 ± 4	11

[a]C_{chi}: concentration of chitosan.
[b]C_{oil}: lemongrass essential oil concentration.
[c]C_{layer}: number of layers.
[d]Theoretical estimated value.

The moisture content values of the active chitosan–LG paperboard systems ranged from 5.6 ± 0.5 to 8.6 ± 0.3%. For uncoated paper was 5.4 ± 0.4%, indicating that the coating does not decrease the moisture sorption. Similar results were obtained by Reis et al[27] for Kraft paper coated with chitosan, which could be associated with the hydrophilicity of chitosan molecules.

3.1 | Anti-insect efficiency of active paperboard

The main characteristic to establish the active paperboard formulation coating was based on the anti-insect efficiency. The anti-insect efficiency (%) was evaluated in different formulations of active material in contact with the adult weevils (Figure 3). The uncoated paperboard (control) did not present anti-insect efficiency after 360 h. The increment of C_{chi} and C_{oil} did not show a significant effect on the anti-insect efficiency. The C_{layer} promoted a positive and significant effect on the anti-insect efficiency. Increasing the number of layers from −1 (1 layer) to +1 (5 layers) increased the anti-insect efficiency by 59%. In Run 5, after 360 h, it was observed 100% of anti-insect efficiency. The anti-insect difference between Run 1 (78%) and Run 5 (100%) is the Cchi. The reduction in anti-insect efficiency in Run 1, which presents higher chitosan concentration (Cchi = 3%) when compared with Run 5 (Cchi = 1%), may be associated with the formation of an emulsion of chitosan and LG entrapping the lipid particles and reducing the availability of oil against the weevils.

Essential oils are composed mainly of volatile components such as benzene derivatives, hydrocarbons, terpenes and other varied compounds.[31] Citral is a monoterpenoid predominantly found in LG,[38] which might explain the high anti-insect efficiency observed in our work. According to Ramawat and Mérillon,[39] the presence of terpenes in essential oils has been associated with anti-insect properties. The physiological action of essential oils on insects is still little known. However, it is believed that they would act on the nervous system, which could result in a neurotoxic action mode.[40]

Repellent packages containing commercial citronella applied on the cardboard surface (0.2 $g.m^{-2}$) significantly reduced *T. castaneum* infestation by approximately 50% in 2 weeks (360 h).[41]

Theoretical estimation of the total solids in the active chitosan–LG paperboard systems was calculated considering the chitosan and LG in each formulation. The total solids deposition ranged from 13 g. m^{-2} for one-layer up to 134 $g.m^{-2}$ for a five-layer coating. The higher anti-insect efficiency was at a higher C_{layer} associated with a higher LG entrapped into the chitosan coating surface. Consequently, a higher citral concentration is the main anti-insect component. The toxic effect of LG on the life cycle of insects was associated with the volatile natural compounds, such as citral (neral + geranial), that change the morphological reproductive system, leading to alterations in oviposition.[42]

The development of anti-insect effect packaging may require high doses of the insecticidal compound, and this could be attained by adding outer or inner layers, with shaving barrier properties to volatiles,

TABLE 3 Mean effects of C_{chi}, C_{oil}, C_{layer} on dependent variables and analysis of variance of the polynomial models on active chitosan–oil paperboard systems at 95% confidence level using the *F* test of the factorial experimental design

	Effects					
Factor	Anti-insect efficiency (%)	Thickness (μm)	Grammage ($g.m^{-2}$)	WVTR ($g_{H2O}\ m^{-2}\ day^{-1}$)	Air permeance ($\mu m\ Pa^{-1}.s^{-1}$)	Cobb test ($g.m^{-2}$)
Mean	43*	420*	257*	282*	0.06*	31*
C_{chi}	8	100	2	−21	−0.11*	0
C_{oil}	28	0	8	−27	−0.02	5
C_{layer}	59*	40*	26*	−53	−0.12*	−21*
$C_{chi} \times C_{oil}$	13	−30*	−12	−4	0.02	3
$C_{chi} \times C_{layer}$	−23	10	−1	4	0.11*	2
$C_{oil} \times C_{layer}$	−4	−10	8	7	0.02	3
Coefficient of determination (R^2)	0.88	0.83	0.78	0.72	0.90	0.77
Model significance (*p*)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Lack of fit (*F* test)	4516.16	0.00	765.80	3061.98	0.06	364.49
Fcal	13.78*	15.53*	15.88*	1.70	4.46	20.45*
Ftab (5%)	5.12	5.12	5.12	5.12	5.12	5.12

*Statistically significant ($p < 0.05$).

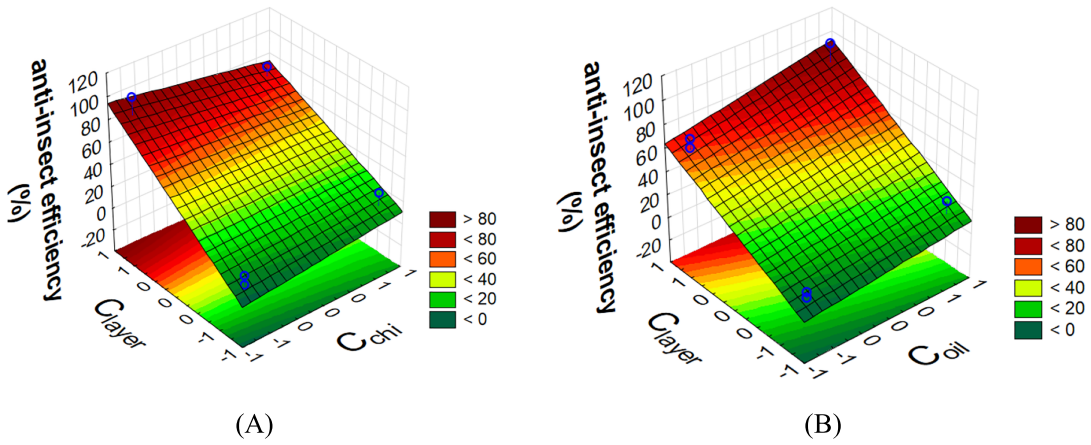

FIGURE 3 Response surfaces for anti-insect efficiency after 360 h in contact with active chitosan–oil paperboard systems: (A) Cchi × Clayer, (B) Coil × Clayer

or using techniques of immobilization of the active compounds, such as microencapsulation, nanocomposite materials and others that could extend the effectiveness of active packaging material.[13]

3.2 | Characterization of active chitosan–LG paperboard systems

The thickness and grammage of uncoated paperboard were 380 μm and 239 $g.m^{-2}$, respectively. The thickness and grammage were directly related to the total solids coating applied on the paperboard surface. The C_{layer} and the interaction of $C_{chi} \times C_{oil}$ presented a significant effect on the thickness. Increasing the C_{layer} from −1 (1 layer) to +1 (5 layers), the thickness increased, on average of 40 μm, and the interaction of $C_{chi} \times C_{oil}$ reduced the thickness, on average of 20 μm. In grammage, only the C_{layer} presented a positive and significant effect, on average, of 26 $g.m^{-2}$.

Thickness increased significantly with chitosan–glycerol coating on paper packaging (grammage 79 $g.m^{-2}$ and thickness 99 ± 1 μm).[43] Similar results were obtained using chitosan and sodium caseinate to coat paper that increased the grammage function of the solid's deposition.[44]

One of the alternatives to control pest infestation in food packaging is to apply a selective barrier. Paper and paperboard are frequently wax coated, aiming to enhance their water resistance and consequently improve the shelf life of the products.[45] Our study added LG in chitosan suspensions in layers, which reduced the WVTR of active chitosan–LG paperboard systems by up to 30% compared with uncoated paperboard (369 ± 11 g $H_2O.m^{-2}.day^{-1}$). All variables (C_{chi},

C_{oil}, C_{layer}) effects were negative and not statistically significant, reducing the WVTR of the active systems. The water vapour barrier was improved by adding palm oil[46] and beeswax,[47] associating with the lipid presence that increased the hydrophobicity of the chitosan coating matrix. Jeong and Yoo[48] evaluated the effect of the beeswax addition in whey protein concentrate–sucrose coating on paperboard's moisture and oil-barrier properties. In general, the transport mechanism of water vapour through porous paper structure is based on vapour-phase diffusion in the inter-fibre pore space (diameter of pores less than 100 Å—Knudsen diffusion), surface diffusion over the fibre surfaces, bulk-solid diffusion within the fibers and capillary transport.[49]

The air permeance of the active chitosan–LG paperboard systems ranged from 0.01 to 0.28 μm $Pa^{-1}.s^{-1}$. It was reduced in all treatments compared with the control (0.38 μm $Pa^{-1}.s^{-1}$). This range of results was not conclusive for air permeance. However, the effects of the independent variables on air permeance were analysed to verify the tendency in different coating formulations. The effects of C_{chi}, C_{layer} and the interaction of $C_{chi} \times C_{layer}$ had a significant and negative effect on the air permeance. Increasing the C_{chi} from −1 (1%) to +1 (3%) and increasing the C_{layer} from −1 (1 layer) to +1 (5 layers) reduced the air permeance, on average of 0.11 and 0.12 μm $Pa^{-1}.s^{-1}$, respectively. The active chitosan–LG coating increased the air resistance of the paperboard, confirming that the active suspension filled the pores between the cellulose fibber networks of the paperboard. It is an interesting property for the active anti-insect material, which increases the total solids deposition on the paperboard structure, retaining the LG volatiles and expanding the potential fumigation action. The greater resistance to air permeance (0.01 μm $Pa^{-1}.s^{-1}$) was observed in Run 1, which contains the highest solids deposition (chitosan and LG) on the paperboard surface (134 $g.m^{-2}$). The lowest air resistance (0.28 μm. $Pa^{-1}.s^{-1}$) was observed with the lowest total solids (13 $g.m^{-2}$) coating, corresponding to Run 8. According to Wang et al,[50] dense and homogeneous chitosan films provide a gas barrier with potential use in low-moisture products packaging, which was observed in the present study after deposition of chitosan–LG solution on the paperboard surface. A natural multilayer material based on chitosan (7 $g.m^{-2}$) and a hydrophobic material (carnauba wax) applied as a coating on a paper sheet improved the gas barrier (water vapour, O_2 and CO_2).[51]

It is well known that cellulose has an affinity to water molecules due to high amounts of hydroxyl groups and depends on the fibre type and surface modification to decrease the hydrophilicity.[52] The water absorption capacity (Cobb test) for uncoated paperboard was 47 ± 4 $g.m^{-2}$ and for the active coated paperboard ranged between 16 $g.m^{-2}$ ($C_{quit} = 3\%$, $C_{oil} = 20\%$, $C_{layer} = 5$) and 43 $g.m^{-2}$ ($C_{quit} = 1\%$, $C_{oil} = 20\%$, $Cl_{ayer} = 1$). The water absorption capacity values obtained by Muratore et al[1] were in the same range as this work. The Cobb value decreased in starch- and chitosan-coated papers (42 and 26 $g.m^{-2}$, respectively) compared with uncoated paper (105 $g.m^{-2}$) was associated with the clogging effect coating the pores in the paper sheet, reducing the water molecule penetration into the pores.[53]

Paper and paperboard packaging exhibits moisture absorption from the surrounding air, which negatively influences the products' quality and usefulnes.[1] The considerable reduction of water absorption obtained in our work (from around 47 to 16 $g.m^{-2}$) could indicate that the proposed material (Table 2) presents the potential to be used as a packaging material.

The water absorption capacity is reduced in the function of the number of coating layers. Increasing the C_{layer} from −1 (1 layer) to +1 (5 layers) significantly reduced the water absorption capacity to 21 $g.m^{-2}$. It was discussed in the SEM images that the emulsion chitosan–LG filled the cellulose interfibrillar spaces, improving the hydrophobicity of the material. The water transport through paper material occurs by diffusion and/or flow process: penetration in the capillaries of the paper sheet, followed by surface diffusion along the capillary walls and diffusion through the cellulose fibres. Mechanical properties and shape of the paperboard packaging are dependent on control over the wetting and barrier properties.[54] The addition of hydrophobic or less hydrophilic materials into cellulose-based materials is an interesting alternative to improve the water and water vapor barrier.

Fat barrier is an important property for packaging foods containing high concentrations of fats or oils directly related to the product's appearance. The Kit test used in our study was employed to investigate papers' grease/oil resistance in different researches.[31,53,55,56] Polymeric coatings improved fat barrier on paperboard surfaces.[57] The active chitosan–LG paperboard systems displayed a fat barrier. A Kit test number of 2 was obtained with the uncoated paperboard. Runs 1 and 3 were fat impermeable, containing a higher chitosan concentration (3%) and a higher number of layers (five layers) in the coating formulations. Kit test numbers of 10–11 were obtained with coating formulations of Runs 2, 4, 5, 7, and 8–11, showing good resistance to fat transfer through the active paperboard systems. A Kit test number of 5 was obtained in coating formulation containing the lowest chitosan (1%) and LG (20%) concentrations (Run 8). A similar result was obtained by Ham-Pichavant et al[31] using a chitosan coating (1.5%, w/w) on a 40 $g.m^{-2}$ Kraft paper surface. The grease resistance improved with increasing the coat weight.[58] Jung et al[53] developed functional antimicrobial papers using chitosan/starch-silver nanoparticles. It verified that all coated papers exhibited a higher Kit number than the uncoated ones, suggesting improvements in grease resistance properties. Basu et al[59] observed grease resistance property in paperboard coated with polysaccharide polyelectrolyte complex material composed of nanostructured fibrous hydrocolloids of chitosan and carboxymethyl cellulose. Chi and Catchmark[55] developed eco-friendly barrier materials based on crystalline nanocellulose/chitosan/carboxymethyl cellulose polyelectrolyte complexes and obtained excellent grease barrier performance, suggesting the food packaging and handling application. A fat barrier was also found by Tyagi et al[60] in nanocellulose-based multilayer barrier coatings as sustainable packaging materials.

Food packaging material should protect the packed product from external microbial contamination. The microbial permeability test was applied on active chitosan–LG paperboard systems. After two days, all negative controls showed turbidity of the TSB broth (Figure 4A). After

(A) (B) (C)

FIGURE 4 Images of microbial permeability of active chitosan–LG paperboard system after 15 days of the experiment: (A) negative control, (B) positive control, (C) examples of the final aspect of chitosan–oil paperboard systems (Runs 1–11)

Packaging type	Odour	Flavour	Overall
Control	6.54 ± 1.86[a]	6.89 ± 1.68[a]	6.97 ± 1.60[a]
Active chitosan–LG paperboard	6.85 ± 1.96[a]	6.45 ± 1.93[a]	6.77 ± 1.88[a]

[a]Media with different letters in the same column are significantly different by Tukey's test ($p < 0.05$).

TABLE 4 Odour, flavour and overall attributes evaluated by panellists for the pasta packaged in control (uncoated paperboard) and active chitosan–LG paperboard pouch

7 days, fungal agglomerates were formed, and at 10 days of the experiment, a fungal mycelia formation was observed. Positive controls did not present turbidity of the broth, as expected, due to the total isolation of the system (Figure 4B). All active chitosan–LG paperboard systems (Runs 1–11) did not show turbidity of the broth, indicating microbial impermeability after 15 days (Figure 4C). Fungal and insect contamination of food products is a problem related to not good agricultural practices. These spoilage agents lead to the deterioration of the food products entailing prejudices in their nutritional value.[61]

The barrier of cellulose fibres, even with the rough and porous surface on an uncoated paperboard, may have restrained the entrance of microorganisms. The antimicrobial and antifungal activities of chitosan could be a beneficial characteristic to active food packaging.[62] de Silva et al[8] verified that chitosan films containing or not buriti oil exhibited a total bacterial barrier.

3.3 | Consumer acceptability test

The main focus of the sensory test was to examine the impact of active chitosan coating on paperboard surface incorporating LG on acceptability and purchase probability of pasta. The use of essential oils as natural preservatives in food systems can negatively influence the organoleptic properties.[63] Before sensory analysis, the paste samples were analysed to assess safety for consumption. The samples had counts within limits established by the National Health Surveillance Agency[64] for cooked meat products (thermotolerant coliforms: 105 NMP.g^{-1}, *Clostridium* sulfite reducer <5 × 10^2 CFU.g^{-1}, coagulase-positive *Staphylococci*: <5 × 10^3 CFU.g^{-1} and *Salmonella*: absence/25 g). The means for odour, colour, flavour and overall impression of the pasta packaged in control (uncoated paperboard) and active chitosan–LG paperboard pouch are reported in Table 4.

The results showed that there were no significant differences between the sensory attributes of the pasta packaged in control (without coating paperboard) and active paperboard pouch ($p > 0.05$). Mani-Lopez et al[65] reported that incorporating LG (500–10 000 ppm) inhibited fungal activity for 3 weeks without negatively affecting the sensory evaluation results of bread samples. Similarly, Lee et al[66] reported that an anti-insect film incorporated with star anise essential oil did not reduce the panellist's acceptance of the material. On the other hand, it should be noted that the sensory characteristic of the real food system can be negatively affected by essential oils, even when they are used as a component of active packaging.[67] Oregano essential oil-containing sachets inhibited the growth of *Penicillium* sp. on sliced bread. However, these impart an unpleasant bitter taste and strong odour, which reduced the overall acceptance of the product.[68] In our study, the overall acceptance of *talharim* samples was 77% and 75% for control and active paperboard packaging, respectively. According to Stone and Sidel,[69] an acceptance ratio higher than or equal 70% is considered acceptable. Regarding the purchase intention, there was no difference between the sample means (without coating 3.96 ± 1.23 and active material 3.66 ± 1.17) among 116 judges, indicating that the active material packaging has market potential. Our results confirmed that the active chitosan film (1% chitosan (w/w), 40% LG (v/w) and five layers) seems to be an acceptable method in sensory terms, sustainable and safe for anti-insect grain-based food packaging as t*alharim* pasta.

Consumer demands will force the industry into environmental awareness. The search for food sustainable materials packaging that promote less worldwide pollution and reduce the chemical pesticides in agriculture has improved a new market in the food packaging sector, following the environmental and food safety consumers' concerns. However, it is still necessary to give more information on these new natural additives before introducing them in daily food packaging.

4 | CONCLUSION

Incorporating LG in a chitosan coating on the paperboard surface resulted in innovative and active packaging material for food

preservation. The active packaging material (C_{chi} = 1.0%, C_{oil} = 30%, C_{layer} = 5) was efficient against weevils, presenting 100% of anti-insect efficiency against *S. zeamais* in wheat grains packaged. The active coating formulations were compatible with the cellulose fibres of the paperboard, forming a homogeneous system. The addition of LG as an active compound in chitosan coatings improved the final properties of paperboard when compared with uncoated paperboard, reducing the WVTR, air permeability, water absorption capacity and increasing the fat and microbial barriers. *Talharim* pasta packed in an active paperboard pouch was sensory acceptable. The active chitosan–LG paperboard system has potential applications as an active material for food packaging to reduce the weevil infestations in wheat, maize grains, farinaceous and other food products, such as pasta and semolina. It could be an alternative to reduce the harmful pesticide substance in the agriculture sector and a proposal of sustainable packaging using raw materials with low impact on the environment.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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