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IMPROVING THE ANTIMICROBIAL ACTIVITY OF BAGASSE PACKAGING PAPER USING ORGANOPHOSPHORUS DIMERS

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ABSTRACT

The antimicrobial properties of bagasse paper sheets coated with natural polymers (chitosan, different ratios of (gelatin/glycerol) + chitosan, hemicellulose, hemicellulose + glycerol, hemicellulose+chitosan) or synthetic organophosphorus dimer compounds were evaluated in this work. Hemicelluloses showed moderate activity against *Bacillus subtilis* and *Candida albicans*, while chitosan showed weak activity against *B. subtilis*. The condition that offered the highest inhibitory activity of bagasse paper was the one coated with 1,3-diaryl-2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane (where aryl is p-chloroaniline or p-anisidine). The developed bagasse papers were evaluated against Gram-positive bacteria, Gram-negative bacteria, yeasts, and fungi. The highest inhibitory activity was obtained at a concentration of 200 mg/mL for p-chloroaniline with an inhibition zone that varied for different microbes from 6.9 mm to 26 mm. The highest inhibitory activity was obtained at 300–250 mg/mL for p-anisidine against most of the pathogenic microorganisms with an inhibition zone that varied for different microbes from 8 mm to 14.75 mm. The observed antimicrobial and antifungal activity properties for bagasse paper coated with 1,3-diaryl 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane could be attributed to the presence of Cl, P atoms, and the lone pair of electrons on N atoms in the structure of the dimers.

Keywords: Antimicrobial; Bagasse paper; Organophosphorus dimers; P-anisidine; P-chloroaniline

1. INTRODUCTION

Increasingly, paper has been used for packaging to provide commercial products with protection from different external influences, to provide ingredient food information, and to preserve food quality using a minimum of preservatives (Coles, 2013; Hakovirta et al., 2015). Many approaches have been proposed for controlling microbial growth in food. Paper and paperboard are the most widely used materials in food packaging (Song et al., 2000; Triantafyllou et al., 2007) and could be used as an antimicrobial packaging material. Antimicrobial agents incorporated into the packaging materials could migrate into the food

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through diffusion and partitioning (Han, 2000) due to the porous structure of paper. This can improve paper properties; such as physical strength, water vapor/gas permeability, surface properties, optical properties, and antimicrobial activity (Han, 2000). The constituents of the antimicrobial agents of packaging materials could diffuse into food products and these migration processes could extend the shelf life of the packaged product. In addition, these agents could improve the food safety or sensory properties while retaining the food quality (Quintavalla & Vicini, 2002; Otoni et al., 2016). Various methods have been reported on preparing antimicrobial chitosan coatings and films for food packaging applications (Basta et al., 2015). Chitosan is soluble in aqueous solutions of various acids; its own reactive amine and hydroxyl groups offer modification possibilities via ionic interactions (Kusrini et al., 2015a). Antimicrobial improvements for recycled fibers have been recorded (Nassar et al., 2015). A previous study on bagasse paper sheet coating revealed that mechanical properties, air permeability, and water absorption showed an improvement by increasing the gelatin and/or chitosan concentrations. The use of a gelatin/glycerol and chitosan blend achieved the preferred improvement on coated paper properties, followed by chitosan on its own (Nassar et al., 2014). The biological activity improvements of paper sheets could be achieved by treatment with polyaniline with or without polystyrene and sliver nanoparticles incorporated into the prepared paper sheets (Youssef et al., 2016). The silver-nanoparticle-containing paper was successfully prepared on the acrylamide grafted bagasse paper sheets under the influence of microwave radiations; the prepared paper sheets exhibited antibacterial activity (Kamel, 2012). Organophosphorus is the promising compound as a friendly flame retardant, which combines the hydro-stability and structure of high phosphorus content (Vothi et al., 2010). The aim of this work was to recycle the agricultural residues, which are normally considered to be a serious environmental problem. The present work investigated the biological activity improvement of packaging bagasse paper sheets by using natural compounds or synthetic phosphorous compounds. The antifungal and antibacterial activities of the phosphorous compounds have been demonstrated by Sharaby (2005).

2. EXPERIMENTAL

2.1. Materials

Paper sheets (100% bleached bagasse kraft pulp) having a basic weight of 80 g/m^2, and a thickness of 0.1 mm, without any surface treatment (supplied by Edfu Co., Egypt) were used as paper substrates (blank). Chitosan, (deacetylation degree 90% and average molecular weight 90,000) was provided by Oxford Laboratory Mumbai, India. Gelatin from porcine skin, type A, was supplied from SIGMA (MSDS available SL07253), USA. Glycerol (>97% purity), as a plasticizer, was purchased from Sigma Aldrich, USA.

2.2. Coating Experiments

Bagasse paper sheets were coated with different concentrations of gelatin solution (0.5, 1, 1.5, 2, and 2.5 wt./V%). Gelatin was dissolved in distilled water at 55^{o}C with stirring. Different concentrations of chitosan were prepared by dissolving the specific amount in 1% acetic acid at room temperature. Different ratios of 2% gelatin solution (containing 0.5% glycerol) and 1% chitosan solution blend were prepared by stirring the mixtures for 10 min. The ratios between gelatin/glycerol and chitosan were 1:0, 3:1, 1:1, 1:3, and 0:1. Paper sheets were coated by immersing them in coating solutions for 30 sec and dried in air for 30 min before drying on a drum (a rotating drying cylinder) at 105^{o}C.

2.3. Synthetic Organophosphorus Compounds

1,3-diaryl-2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane, where aryl is either p-chloroaniline or p-anisidine, was prepared as described elsewhere (Mohamed, 2006). Table 1 shows the equimolar ratio and the melting points of the prepared haxachlorocyclodiphosph(V)azane

compared with that recorded in the literature (Mohamed, 2006).

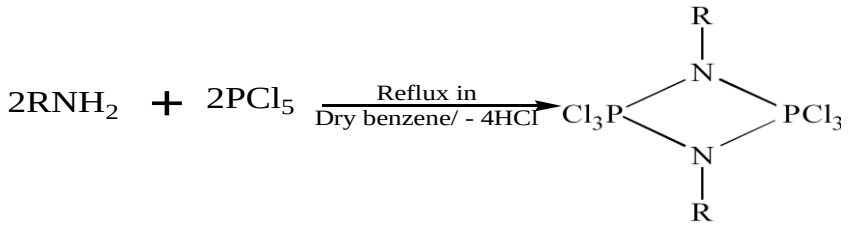


Figure 1 preparation of 1,3-diaryl-2,2,2,4,4,4-hexachlorocyclodiphosph(V) azane

2.4. Antibacterial and Antifungal Activity

Antimicrobial and antifungal activity was investigated for bagasse paper sheets after the addition of natural polymers or 1,3-diaryl-2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane where aryl is either p-chloroaniline or p-anisidine.

2.4.1. The strains used

The common pathogens and food spoilage microorganisms were selected for their relevance in bakery products. Antibacterial activities were evaluated using the Gram-positive bacteria (*Bacillus subtilis, Staphylococcus aureus*), Gram-negative bacteria (*Pseudomonas aeruginosa, Escherichia coli, Proteus vulgaris*), yeasts (*Candida albicans, Saccharomyces cerevisiae*), and fungi (*Aspergillus niger*). The inhibition zones of microbial growth produced by different compounds of cyclodiphosph(V)azane derivatives at different concentrations were observed using an overlay method (Black, 1996) and are listed in Table 2 and Figures 3–6.

2.4.2. Media used

The bacteria were slanted on nutrient agar (Merck, Darmstadt, Germany); the yeast was slanted and maintained on Sabaroud's agar medium (Lab M. Limited, Bury, Lancashire, UK); and the fungi were slanted and maintained on the potato Dextrose Agar medium (Lab M. Limited).

2.4.3. Bioassay

The antibacterial screening was investigated by the disk diffusion method (Ganapathi et al., 2016). The test compounds were dissolved in dimethylformamide. The organisms were streaked in radial patterns on the agar plates. The plates were then incubated under aerobic conditions at 37°C and 28°C for 24 h and 48 h for bacteria and fungi, respectively. In order to obtain comparable results, all prepared solutions were treated under the same conditions and under the same incubated plates. All tests were performed for three replicates. The plates were examined for evidence of antimicrobial activities, which were represented by a zone of inhibition of microorganism growth around the paper disk.

3. RESULTS

Equimolar ratios of the preferred amino compound and phosphorus pentachloride are presented in Table 1.

Table 1 Equimolar ratio and melting points of haxachlorocyclodiphosph(V)azane

Compd. No.	Amino compound (gm, mole)	Phosphorus pentachloride (gm, mole)	Empirical formula	Melting point (^oC)	
				Measured	Literature
D1	p-chloroaniline dimer (12.75; 0.1)	(20.9; 0.1)	$C_{12}H_8N_2P_2Cl_8$	180–183	181–183
D2	p-anisidine dimer (12.3; 0.1)	(20.9; 0.1)	$C_{14}H_{14}N_2P_2O_2Cl_6$	199–201	200–202

Comparable values of the measured melting point (in both D1 and D2) with that in the literature verified the formation of the desired hexachlorocyclodiphosph(V)azane with the empirical formula given in Table 1.

3.1. Antimicrobial Activity of Natural Polymers

Antimicrobial and antifungal activities were made for all samples containing natural polymers: chitosan (0.2–2%), different ratios of (2% gelatin/0.5% glycerol) + 1% chitosan, 0.215% hemicellulose, hemicellulose + 0.5% glycerol, 0.215% hemicellulose + 1% chitosan and blank bagasse paper. Activity was investigated for the Gram-positive bacteria, *B. subtilis* and *S. aureus*; the Gram-negative bacteria, *P. aeruginosa, E. coli, and P. vulgaris*; the yeasts, *C. albicans* and *S. cerevisiae*; and the fungi *A. niger*. The inhibition zones of microbial growth produced by the different compounds of chitosan or hemicellulose were measured and are listed in Table 2. Additives other than those in Table 2 showed a negative inhibition zone against microbial and fungal activity.

Table 2 Antibacterial and antifungal activity of bagasse paper with different additives

The additives for paper (100 mL)		*B. subtilis*	*C. albicans*
Chitosan (100%, 2g)		+	−ve
Chitosan 50%	gelatin/glycerol 50%	+	−ve
Hemicelluloses (100%, 0.215g)	——	++	++
Hemicelluloses (100%, 0.215g)	glycerol 0.5g	++	++
Hemicelluloses 50%, 0.215g	chitosan with reflux 50%,1g	++	−ve
Hemicelluloses 50%, 0.215g	chitosan without reflux 50%,1g	+	−ve
Blank		−ve	−ve

+ = weak; ++ = moderate; −ve = no inhibition zone

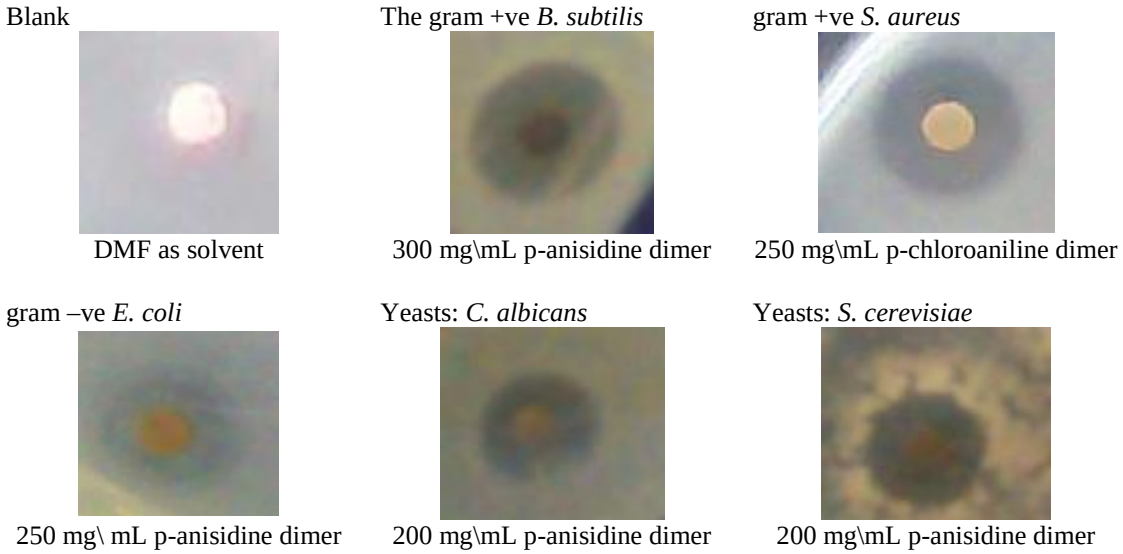


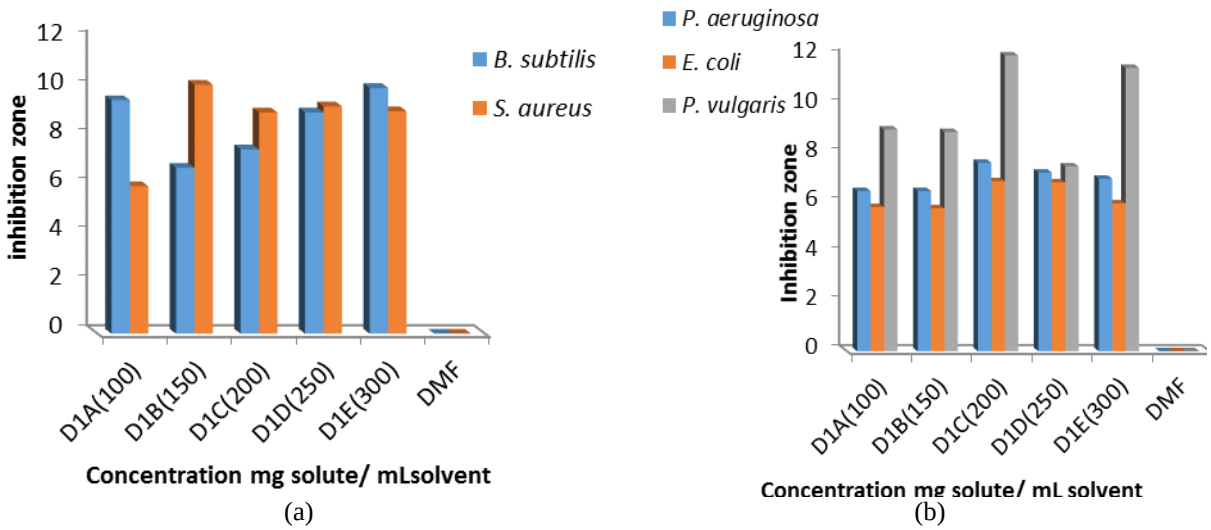
Figure 2 Some pictures for antimicrobial activities

3.2. Antimicrobial Activity of Synthetic Organophosphorus Compounds

Organophosphorus compounds have a proven biological activity (Mohamed, 2006); their compounds are used as pesticides and nerve agents. The inhibition zones of microbial growth produced by different microorganisms around the paper disk treated by organophosphorus compounds were illustrated by a diameter of clear zones expressed in millimeters. Cyclodiphosph(V)azane blocked the synthesis of the proteins and inhibited the growth of the microorganisms (Alaghaz, 2014). The most famous phosphorus–nitrogen compounds cyclodiphosphazanes with interesting properties have been intensively studied. Cyclodiphosph(V)azane blocked the synthesis of the proteins and inhibited the growth of the microorganisms (Alaghaz, 2014).

3.2.1. Antimicrobial and antifungal activity for 1,3-di-p-chloroaniline 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane on bagasse paper

Antimicrobial and antifungal activity were measured for bagasse paper with 1,3-di-p-chloroaniline 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane at different concentrations from 100 to 300 mg solute/mL solvent against Gram-positive bacteria (Figure 3a), Gram-negative bacteria (Figure 3b), and fungi (Figure 4). The inhibition zones were: 4.5–6 = weak; 6.1–7.9 = moderate; and 8–26 = significant.



(a) (b)

Figure 3 Antimicrobial activity against Gram-positive (a) and Gram-negative (b) bacteria with a p-chloroaniline dimer

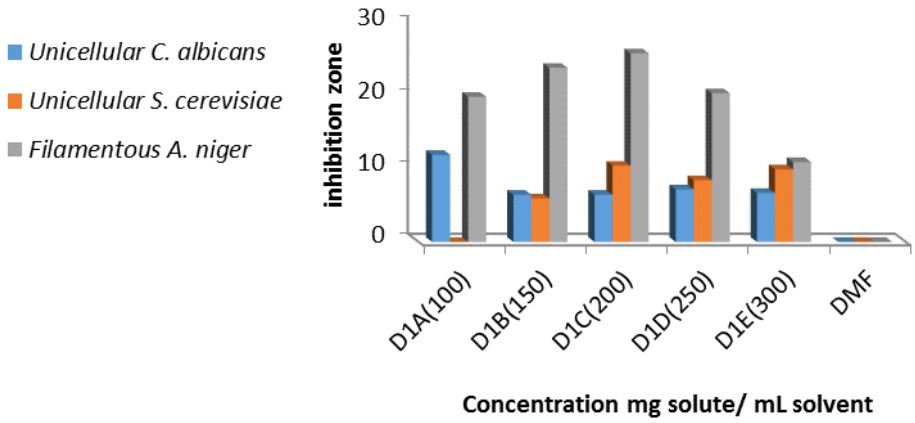
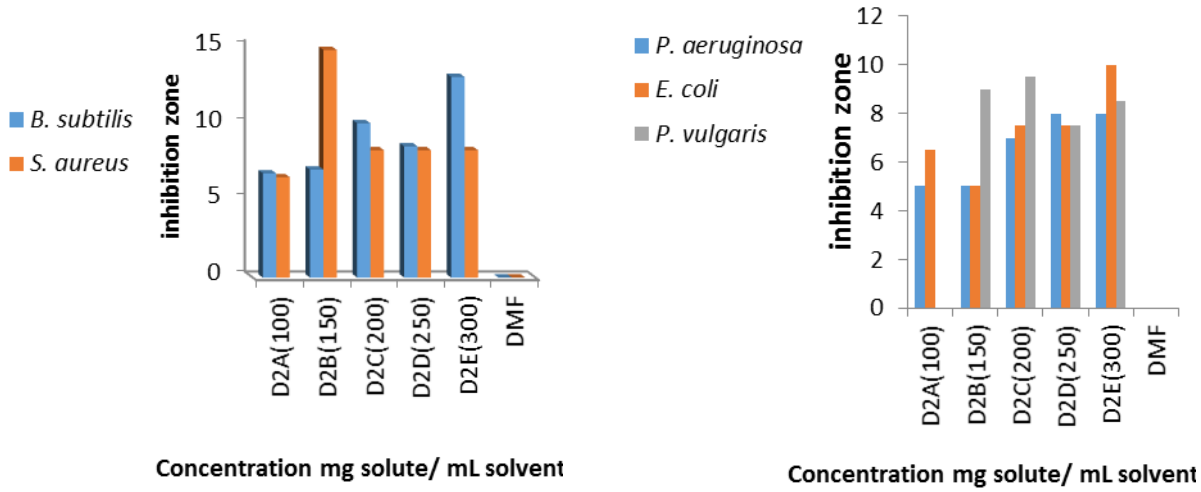


Figure 4 Activity against fungi with a p-chloroaniline dimer

3.2.2. Antimicrobial and antifungal activity for 1, 3-di-p-Anizidine 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane on bagasse paper

Antimicrobial and antifungal activity were measured for bagasse paper with 1,3-di-p- Anizidine 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane at different concentrations from 100 mg to 300 mg solute/mL solvent against Gram-positive bacteria (Figure 5a), Gram-negative bacteria (Figure 5b), and fungi (Figure 6).



(a) (b)

Figure 5 Antimicrobial activity against Gram-positive (a) and Gram-negative (b) bacteria with a p-anisidine dimer

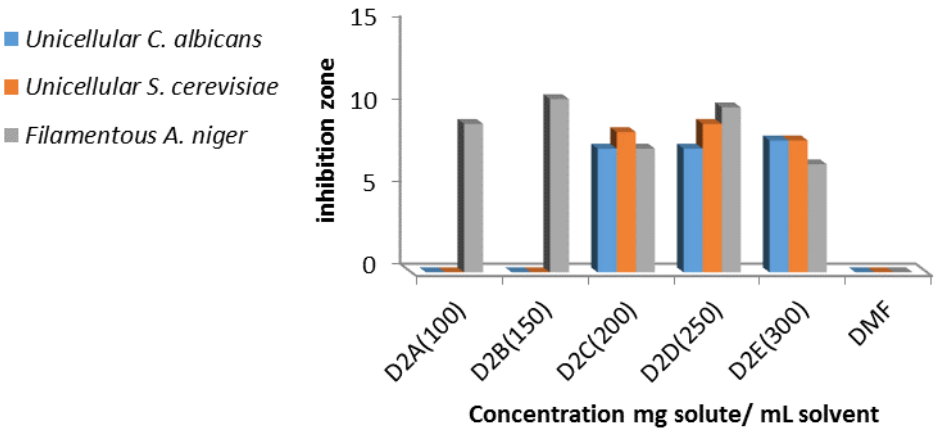


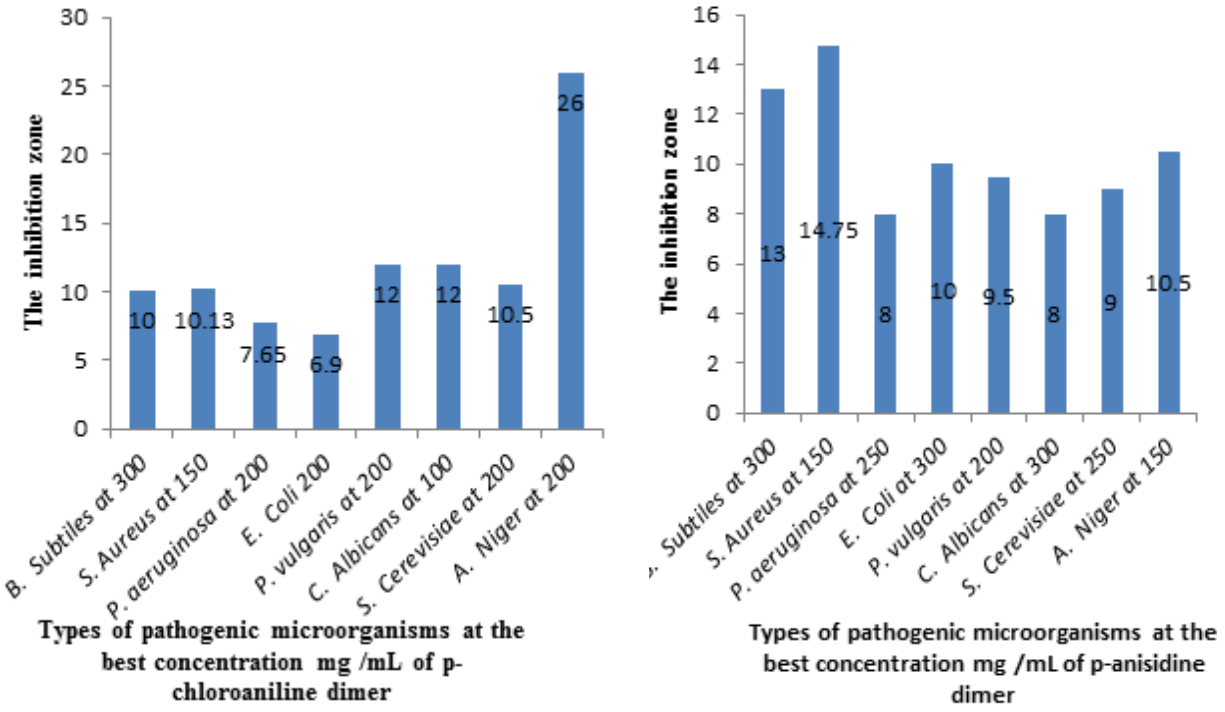
Figure 6 Activity against fungi with a p-anisidine dimer

The best concentrations of 1,3-diaryl 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane, where the aryl is D1 = p-chloroaniline or D2 = p-anisidine, could show the highest inhibition zone of bagasse paper treated with dimers against the Gram-positive bacteria *B. subtilis* and *S. aureus*; and the Gram-negative bacteria *P. aeruginosa*, *E. coli*, and *P. vulgaris*; yeasts such as *C. albicans* and *S. cerevisiae*; and fungi *A. niger*, are presented in Table 3.

Table 3 Best concentration that gave high inhibition zone of bagasse paper treated with all dimers

Microbes	P-chloroaniline		P-anisidine	
	High inhibition zone (mm)	At mg/mL conc.	High inhibition zone (mm)	At mg/mL conc.
B. subtilis	10.00	300	13.00	300
S. aureus	10.13	150	14.75	150
P. aeruginosa	7.65	200	8.00	250
E. coli	6.90	200	10.00	300
P. vulgaris	12.00	200	9.50	200
C. albicans	12.00	100	8.00	300
S. cerevisiae	10.50	200	9.00	250
A. niger	26.00	200	10.50	150

Figure 7 shows that the highest inhibition zone obtained for bagasse paper treated with p-chloroaniline (a) and p-anisidine (b) dimer at optimum concentrations against most microorganisms.



(a) (b)

Figure 7 The best inhibition zones of treated paper with: (a) p-chloroaniline dimer; and (b) p-anisidine dimer

4. DISCUSSION

4.1. Antimicrobial Activity of Natural Polymers

Table 2 shows that in terms of the inhibition zone, the control chitosan film did not show a noticeable inhibitory effect against all tested microorganisms. The positively charged amino group in chitosan is responsible for its antimicrobial character. It interacts with negatively charged microbial cell membranes and causes the leakage of intracellular constituents of the microorganisms (van den Broek et al., 2015). Chitosan films with gelatin/glycerol showed some antimicrobial effect, but it did not show an inhibitory zone towards microorganisms. This is

predictable as chitosan itself has the natural characteristic of antimicrobial activity (Goy et al., 2016). Chitosan, as a solid material, is unable to spread through the agar media, so chitosan-active sites only inhibited organisms that were in direct contact with it (Sanchez-Machado et al., 2015). Hemicellulose with/without glycerol showed a moderate antimicrobial effect against *B. subtilis* and *C. albicans*, while the hemicellulose/chitosan blend showed a weak antimicrobial effect against *B. subtilis*.

4.2. Antimicrobial Activity of Synthetic Organophosphorus Compounds

The observed antimicrobial and antifungal activity properties for bagasse paper coated with 1,3-diaryl 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane could be attributed to the presence of Cl, P atoms, and the lone pair of electrons on N atoms, in the structure of dimers. In addition, the phenolic components in the organophosphorus dimers were able to destroy the outer membrane of Gram-negative bacteria, releasing lipopolysaccharides and increasing the permeability of the cytoplasmic membrane to *Adenosine* triphosphate (Kusrini et al., 2015b).

4.2.1. Antimicrobial and antifungal activity for 1,3-di-p-chloroaniline 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane on bagasse paper

The results of antimicrobial and antifungal activity for 1,3-di-p-chloroaniline 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane bagasse paper are noted below. The results obtained using ampicillin as the reference compound (Figure 3a) indicated the following, against Gram-positive bacteria:

- For *B. subtilis*: Samples D1A, D1D, and D1E showed significant inhibition compared to the reference compound, whereas samples D1B and D1C showed moderate inhibition.
- For *S. aureus*: Samples D1B, D1C, D1D, and D1E showed significant inhibition activity to the reference compound, whereas sample D1A showed weak inhibition. This means that at least 150 mg solute/mL solvent was required for significant inhibition results, and it was considered to be the optimum concentration because higher concentrations have comparable results.

Against Gram-negative bacteria, the results obtained in Figure 3b indicated the following:

- For *P. aeruginosa*: Samples D1C and D1D showed significant inhibition compared to the reference compound, whereas samples D1A, D1B, and D1E showed moderate inhibition.
- For *E. coli*: Compounds D1C and D1D showed moderate inhibition compared to the reference compound, whereas samples D1A, D1B, and D1E showed weak inhibition.
- For *P. vulgaris*: Samples D1C and D1E showed significant inhibition compared to the reference compound, whereas samples D1A, D1B, and D1D showed moderate inhibition. The blank sample showed no inhibition compared to the reference compound. These results indicate that 200 mg solute/mL solvent was the optimum concentration for significant inhibition results against *P. aeruginosa*, *E. coli, and P. vulgaris*.

Antifungal activity;

Most of the synthesized compounds were tested for their antifungal activity using Mycostatin as a reference compound. The obtained results summarized in Figure 4 indicated that:

- For *C. albicans*: Sample D1A showed significant inhibition compared to the reference compound, whereas samples D1B, D1C, D1D, and D1E showed moderate inhibition. This means that 100 mg solute/mL solvent was the optimum concentration because higher concentrations had inferior results.
- For *S. cerevisiae*: Samples D1C and D1E showed significant inhibition compared to the reference compound, whereas sample D1D showed moderate inhibition. Sample D1B showed weak inhibition compared to the reference compound, whereas the compounds D1A and blank did not show any inhibition. These results indicated that 200 mg solute/mL solvent was the optimum concentration for significant inhibition results against *S. cerevisiae*.

- For *A. niger*: All samples showed significant inhibition compared to the reference compound. This means that 100 mg solute/mL solvent was the optimum concentration, because it gave satisfactory significant inhibition results.

4.2.2. Antimicrobial and antifungal activity for 1, 3-di-p-anizidine 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane on bagasse paper

Antimicrobial and antifungal activities were measured and are listed in Figures 5–6. The results obtained using ampicillin as the reference antibiotic (Figure 5a) indicated the following, Against Gram-positive bacteria:

- For *B. subtilis*: Samples D2C, D2D, and D2E showed significant inhibition compared to the reference antibiotic, whereas sample D2B showed moderate inhibition; sample D2A showed a weak inhibition; and the blank compound showed no inhibition. These results indicated that 200 mg solute/mL solvent was the economic concentration for an acceptable significant inhibition result against *B. subtilis*, while the maximum inhibition result was attained at 300 mg solute/mL solvent.
- For *S. aureus*: Samples D2B, D2C, D2D, and D2E, showed significant inhibition compared to the reference antibiotic, whereas sample D2A showed moderate inhibition; the blank compound showed no inhibition. It is clear that 150 mg solute/mL solvent is the optimum concentration for a significant inhibition result against *S. aureus*.

Against Gram-negative bacteria, the results obtained in Figure 5b indicate the following:

- For *P. aeruginosa*: Samples D2D and D2E showed significant inhibition compared to the reference antibiotic, whereas sample D2C showed moderate inhibition; samples D2A and D2B showed weak inhibition, and the blank compound showed no inhibition.
- For *E. coli*: Sample D2E showed significant inhibition compared to the reference antibiotic; samples D2C and D2D showed moderate inhibition; samples D2A and D2B showed weak inhibition; and the blank compound showed no inhibition. It is clear that the inhibition zones against *P. aeruginosa* and *E. coli* increased as the solute concentration increased.
- For *P. vulgaris*: Samples D2B, D2C, and D2E showed significant inhibition compared to the reference antibiotic; sample D2D showed moderate inhibition; and sample D2A and the blank showed no inhibition. This means that the 150 mg solute/mL was is the optimum concentration for significant inhibition results against *P. vulgaris*.

Antifungal activity;

Most of the synthesized compounds were tested for their antifungal activity using Mycostatin as a reference compound. The results obtained (Figure 6) indicated the following:

- For *C. albicans*: Sample D2E showed significant inhibition compared to the reference compound, whereas samples D2C and D2D showed moderate inhibition; samples D2A, D2B, and the blank showed no inhibition. An acceptable inhibition result was attained at 200 mg solute/mL solvent (7.5 mm) while the maximum inhibition result was attained at 300 mg solute/mL solvent (8.0 mm).
- For *S. cerevisiae*: Samples D2C, D2D, and D2E showed significant inhibition compared to the reference compound, whereas samples D2A, D2B, and the blank showed no inhibition. This means that the inhibition zone at 200 mg solute/mL solvent was considered to be the optimum concentration.
- For *A. niger*: samples D2A, D2B, and D2D showed significant inhibition compared to the reference compound, whereas sample D2C showed moderate inhibition; sample D2E showed weak inhibition, and the blank compound showed no inhibition. The economic and optimum inhibition zone was achieved at 100 mg solute/mL solvent.

Table 3 shows the best concentration that gave the highest inhibition zone for bagasse paper coated by 1,3-diaryl 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane, where the aryl is: D1 = p-

chloroaniline or D2 = p-anisidine. Table 3 and Figure 7a show that the highest inhibition zone was at 200 mg/mL of p-chloroaniline dimer against most microorganisms except *B. subtilis, S. aureus,* and *C. albicans,* which gave high inhibition zones at 300, 150, and 100 mg/mL, respectively.

The highest inhibition zones of the p-anisidine dimer (Figure 7b) were at 300 mg/mL against *B. subtilis*, *E. coli*, and *C. albicans*, whereas it was 250 mg/mL against *P. aeruginosa* and *S. cerevisiae*. *P. vulgaris* gave a high inhibition zone at 200 mg/mL, whereas *S. aureus* and *A. niger* gave high inhibition zones at 150 mg/mL.

5. CONCLUSION

The chitosan films showed some antimicrobial effect; however, they did not show inhibitory zones towards the microorganisms. Antimicrobial and antifungal activity properties recorded for bagasse paper coated with 1,3-diaryl 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane may be due to Cl, P, and a lone pair of electrons on N atoms in the dimers structure. The best condition that offered the strongest inhibitory activity of bagasse paper was the one coated with 1,3-diaryl 2,2,2,4,4,4-hexachlorocyclodiphosph(V)azane against the Gram-positive, Gram-negative bacteria and *A. niger* fungi. The highest inhibitory activity was at 200 mg/mL of p-chloroaniline dimer against most microorganisms except *B. subtilis, S. aureus,* and *C. albicans,* which gave high inhibitory activity at 300, 150, and 100 mg/mL, respectively.

The p-anisidine dimer gave the strongest inhibitory activity at 250-300 mg/mL against all the tested microorganisms except *P. vulgaris*, which gave a high inhibition zone at 200 mg/mL concentration. *S. aureus* and *A. niger* both gave high inhibition zones at 150 mg/mL.

6. ACKNOWLEDGEMENT

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