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RESEARCH ARTICLE

SYNTHESIS OF COPPER COMPLEXES AND THEIR PESTICIDAL ACTIVITY

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ABSTRACT

A series of new Cu(II) complexes were synthesized by the addition of Copper(II) chloride to ligand derived from Salicylaldehyde and amino acid with Pyridine/triphenylphosphine. The complexes were tested for pesticidal activity against papaya mealybug. All eight Cu(II) complexes show significant pesticidal activity against all stage instars of mealybug.

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INTRODUCTION

The papaya mealybug, *Paracoccus marginatus* Williams and Granara de Willink is a small polyphagous sucking pest insect. This caused damage to several genera host plants, including economically important tropical fruits, vegetables and ornamentals. It attacks Parthenium hysterophorus, Tapioca, Guava, Teak, Brinjal, Pigeonpea, Jatropha and Hibiscus with varying levels of severity. Papaya mealybug has caused havoc in agricultural and horticultural crops¹. In 2009 it destroyed economically important crops and huge losses to farmers in western part of Tamilnadu². The bug can damage upto 80% of host plants like cotton, egg-plants and sunflower³. Early instars (called crawlers) are highly active which get readily dispersed by wind, rain, and irrigation water, birds, ants, clothing and vehicle etc⁴. It covered with powdery white wax and deposits a waxy layer on the leaf surface reducing the productivity of the plant⁵. The waxy coating sticks to equipments, animals and man while moving. Ants carry the bug and spread to non-infested plants. Metal complexes play an essential role in agricultural, Pharmaceutical and industrial chemistry. Metal complexes with Schiff base derived from Salicylaldehyde show toxicities against insects⁶. Copper Schiff base complexes exhibit pesticidal activity against tribolium castaneum⁷. In the present work we report the synthesis and pesticidal activity of Cu (II) complex derived from Schiff base containing triphenylphosphine/pyridine.

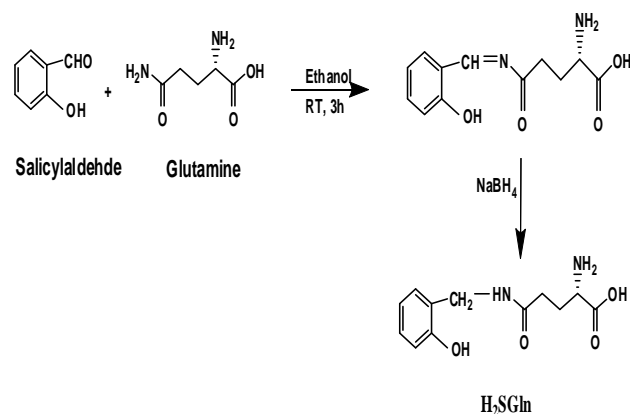
MATERIALS AND METHODS

All the chemicals used were pure and analytical grade. Glutamine, Cysteine, Aspartic acid, Methionine, Salicylaldehyde, Pyridine, Triphenyl phosphine, Cu (II) chloride dihydrate and common reagents such as NaOH, KOH, and Sodium borohydride were purchased from Merck Specialities Private Limited.

Preparation of Ligands

Preparation of Glutamine

Glutamine (1.462 g, 0.01 M) was dissolved in KOH (0.56 g, 0.01 M) in 10 mL distilled water. Salicylaldehyde (1.221 g, 0.01 M) was dissolved in 10 mL ethanol. Salicylaldehyde solution was added to Glutamine solution and allowed to stir for 3 h. The obtained yellow colour solution was cooled in an ice bath. The intermediate Schiff base that had formed was reduced with 5 mL of Sodiumborohydride (0.378 g, 0.01 M) containing few drops of NaOH solution. The yellow colour slowly discharged and the pH of the solution was adjusted to 3.5 – 6 using few drops of con HCl to obtain the solid precipitate. The obtained precipitate was then filtered and washed with Ethanol and Diethyl ether and allowed to dry completely at room temperature. In a similar manner other ligands H₂SCys from Cysteine, H₂SAsp from Aspartic acid, and H₂SMet from Methionine were prepared.



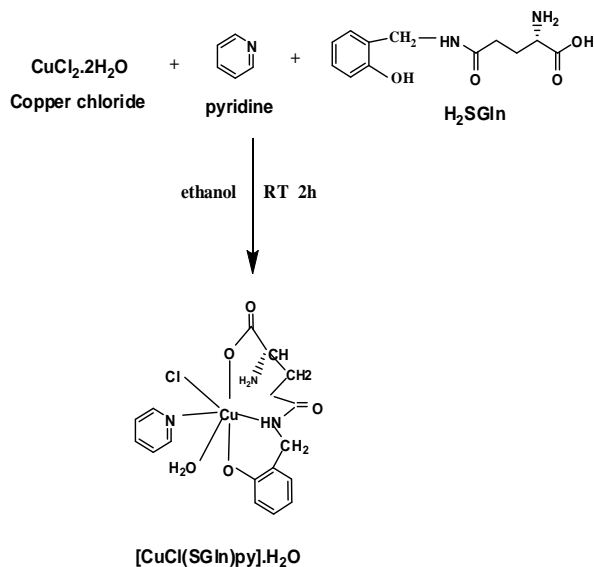
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Pesticidal Activity of Synthetic Cu(II) complexes

S.No	Name of the complexes	Concentration	Time in minutes						Mortality time	% larvicidal activity to % Mortality	
			15	30	45	60	75	90			120
1	[CuCl(SGln)py]H ₂ O	0.001M	1	2	3	3	4	5	After 90 minutes	100	
		0.002 M	2	3	3	4	5		After 75 minutes	100	
		0.003 M	2	4	4	5			After 60 minutes	100	
2	[CuCl(SGln)PPh ₃]H ₂ O	0.001M	2	2	3	5			After 60 minutes	100	
		0.002 M	2	3	4	5			After 60 minutes	100	
		0.003 M	3	4	5				After 45 minutes	100	
3	[CuCl(SCys)py]H ₂ O	0.001M	0	1	2	3	4	5	After 90 minutes	100	
		0.002 M	0	1	2	4	5		After 75 minutes	100	
		0.003 M	1	2	2	3	5		After 75 minutes	100	
4	[CuCl(SCys)PPh ₃]H ₂ O	0.001M	0	0	1	3	5		After 75 minutes	100	
		0.002 M	0	1	2	4	5		After 75 minutes	100	
		0.003 M	1	2	3	4	5		After 75 minutes	100	
5	[CuCl(SAsp)py]H ₂ O	0.001 M	0	1	2	3	4	4	5	After 120 minutes	100
		0.002 M	1	2	3	3	4	5		After 90 minutes	100
		0.003M	1	2	3	4	4	5		After 90 minutes	100
6	[CuCl(SAsp)PPh ₃]H ₂ O	0.001 M	0	1	2	3	4	5		After 90 minutes	100
		0.002 M	1	2	2	3	4	5		After 90 minutes	100
		0.003 M	1	2	3	4	5		After 75 minutes	100	
7	[CuCl(SMet)py]H ₂ O	0.001 M	2	3	4	5			After 60 minutes	100	
		0.002 M	2	4	4	5			After 60 minutes	100	
		0.003 M	3	4	5				After 45 minutes	100	
8	[CuCl(SMet)PPh ₃]H ₂ O	0.001 M	2	2	3	5			After 60 minutes	100	
		0.002 M	3	3	5				After 45 minutes	100	
		0.003 M	3	5					After 30 minutes	100	

Synthesis of copper (II) complex using pyridine

Copper chloride dihydrate (1.70 g, 0.01 M) was dissolved in 15 mL ethanol. Pyridine (0.791g, 0.01 M) was dissolved in 10 mL ethanol and transferred to copper chloride solution. It was stirred for 10 minutes. The corresponding ligand (0.01M) was dissolved in 10 mL sterile water with KOH (1 mL, 1M). The ligand solution was added to it and allowed to stir for 2h at room temperature. The reaction mass was filtered and allowed to evaporate at RT. The resulting solid precipitate was dried in a dessicator for two days.



Synthesis of copper (II) complex using Triphenylphosphine

Copper chloride dihydrate (1.70 g, 0.01 M) was dissolved in 15 mL ethanol. Triphenylphosphine (2.6 g, 0.01 M) was dissolved in 10 mL ethanol and transferred to copperchloride solution. It was stirred for 10 minutes. The corresponding ligand was dissolved in 10 mL sterile water with KOH (1mL, 0.001M).

The ligand solution was added to it and allowed to stir for 2h at room temperature. The reaction mass was filtered and allowed to evaporate at RT. The resulting solid precipitate was dried in a dessicator for two days.

Pesticidal activity

The Papaya mealybugs 1 to 5th instar larvae were collected from (Papaya tree, Guava tree and Hibiscus)plants in Thiruvapur town. Fourth stage instar larvae of papaya mealybug were exposed to test concentrations of 0.001M, 0.002M and 0.003M. The metal complexes were taken in cleaned sterilize test tubes and dissolves in 1 mL of water and were made to 0.001 M, 0.002 M and 0.003 M concentration. 5 nos larvae taken in 1 mL distilled water served as control. 5 larvae per concentration were used for all larvae experiment. The tubes were kept for 2 h and observed for mortality of larvae.

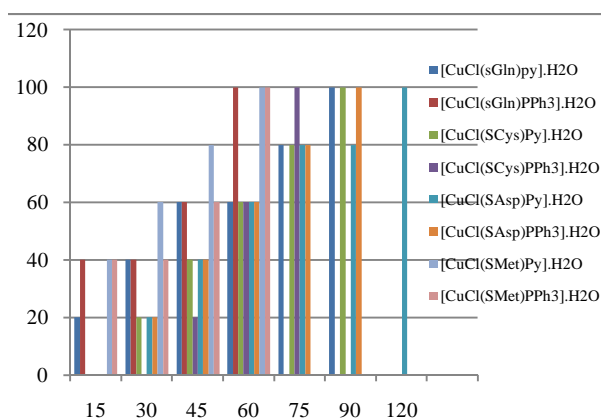


Fig 1 % Mortality of Cu(II) complexes at 0.001 M concentration

RESULTS AND DISCUSSION

The results were tabulated and the analysis data were presented in the fig.1, Fig. 2 and Fig.3. The metal complex [CuCl(SMet)PPh₃]H₂O showed maximum activity i.e 100% mortality after 30 minutes when compared to other metal

complexes. Generally Triphenyl phosphine metal complexes showed slightly higher activity than pyridine metal complexes. It was observed that all the metal complexes showed 100% mortality within 120 minutes.

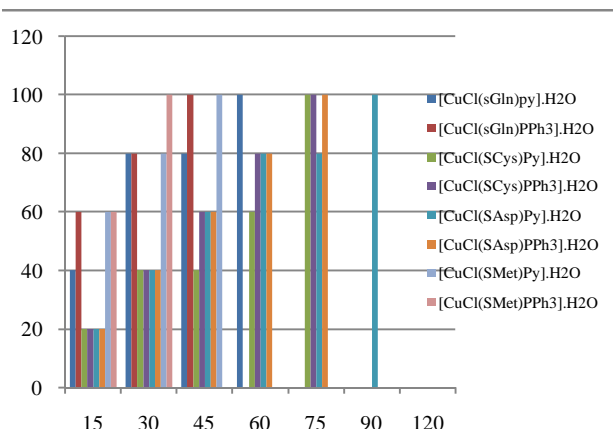


Fig 2 % Mortality of Cu(II) complexes at 0.002 M concentration

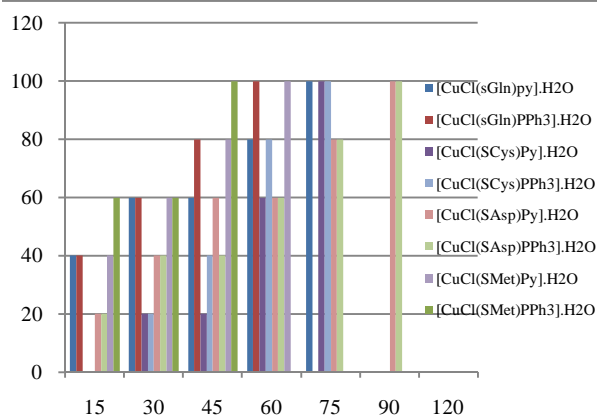


Fig 3 % Mortality of Cu(II) complexes at 0.003 M concentration

CONCLUSION

The metal complexes extracts were applied directly upon Papaya mealybug in Hibiscus plants. It removed the coated wax over mealy bug and caused to death of mealy bug. The plant was observed over one month. The metal complexes showed activity against the all kinds of instars larvae of Papaya mealy bug and not affected the plant and also the growth of plant is well. As the metal complexes were found effective in controlling Papaya mealybug, these can be used as pesticide.

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