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## RESEARCH ARTICLE

### SYNTHESIS AND LARVICIDAL ACTIVITY OF COPPER COMPLEXES CONTAINING AMINO ACID AND PYRIDINE/TRIPHENYLPHOSPHINE

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#### ABSTRACT

Various species of mosquitoes transmit different types of disease like malaria, yellow fever, dengue fever, chikungunya, filariasis and encephalitis to more than 700 million people annually with millions of resultant deaths. The aim of the work is to evaluate larvicidal activity of Cu(II) complex against mosquito. These reduced Schiff base pyridine/ triphenyl phosphine Cu(II) complex were found to have effective against anopheles and Culex mosquito.

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## INTRODUCTION

Mosquitoes are very wide spread, occurring in all regions of the world. In warm and humid tropical regions, they are active for the entire year. The female mosquitoes are blood eating pests and transmit extremely harmful human and livestock diseases. Some mosquitoes that bite humans routinely act as vectors for a number of infectious diseases causing millions of death every year (Mittal and Subbarao, 2003). *Aedes aegypti* is known for the transmission of viral diseases such as yellow fever, dengue fever and chikungunya. *Anopheles* is an important vector, which caused parasitic disease malaria (Fonteuillel *et al.*, 1997). *Culex* and *Culiseta* are vectors of Bacterial tularemia. The chemicals used in insecticidal sprays will affect the natural environmental conditions (Lokesh *et al.*, 2010). Chloro fluoro carbon which is released by mosquito coil and liquidators depletes ozone layer. Some insecticides possess a substantial hazard to a variety of animal life and the environment in the form of biomagnifications (Wej Choochote *et al.*, 2005). The resistance capacity of mosquitoes increased against insecticides (Kamaraj *et al.*, 2011). Thus there is an urgent need for new, effective and eco- friendly mosquito control agents (John odda *et al.*, 2008). Schiff base ligand and their copper complexes possess significant antimalarial activities (John *et al.*, 1982). It has shown that the presence of 2-pyridylalkylidene essential for antimalarial activities (Vinod *et al.*, 2003). Heterocyclic ligands based on ferrocene, pyridine

containing transition metal complexes exhibit antimalarial activity (Rishu Katwal *et al.*, 2013; Bakir *et al.*, 2005). Chanadrakeka *et al.* (2011) reported the larvicidal activity of copper (II) complexes of amino acid derived Schiff base with 1,10 Phenanthroline and 2,2' bipyridyl. The aim of the present work is synthesise of copper (II) complexes of amino acid derived Schiff base containing pyridine/ triphenylphosphine and evaluate the larvicidal activity of copper complexes.

## MATERIALS AND METHODS

### Materials

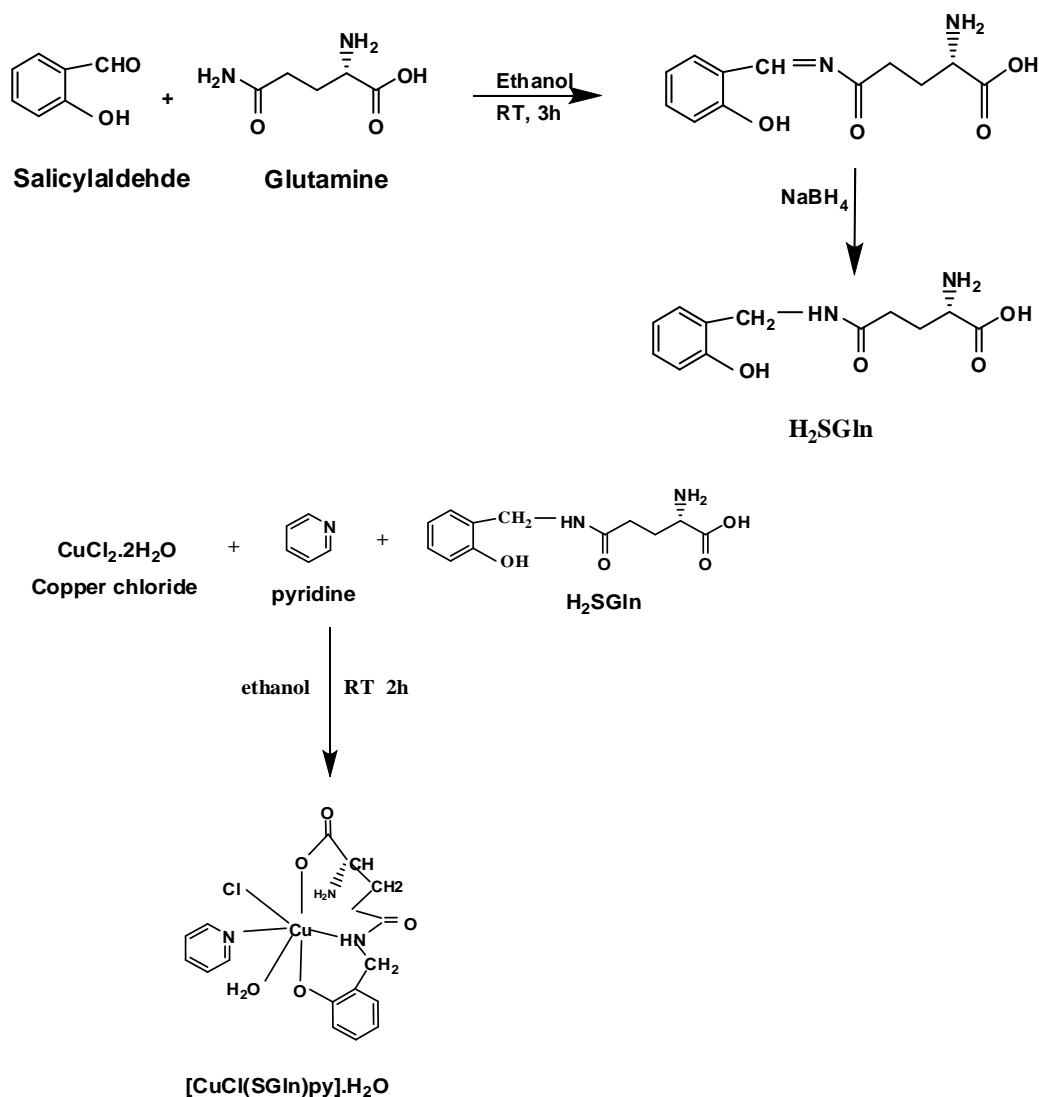
All the chemicals used were pure and analytical grade. Glutamine, Cysteine, Aspartic acid, Methionine, Salicylaldehyde, Pyridine, Triphenyl phosphine, Cu(II) chloride dihydrate and common reagents such as NaOH, KOH, Sodium boro hydride were purchased from Merck Specialties Private Limited.

### Preparation of Ligands

#### Preparation of H<sub>2</sub>SGln from 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 path. The intermediate Schiff base that had formed was

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reduced with 5 mL of Sodium borohydride (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 H<sub>2</sub>SCys from Cysteine, H<sub>2</sub>SAsp from Aspartic acid, H<sub>2</sub>SMet from Methionine were prepared.

#### 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 copperchloride solution. It was stirred for 10 minutes. The corresponding ligand (0.01M) was dissolved in 10 mL sterile water with KOH (1 mL, 0.01M). 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

Copperchloride 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. The similar methodology was followed to synthesize copper(II) complexes using four ligands with pyridine and triphenylphosphine. They include copper-salicylaldehyde-Glutamine-pyridine, copper-salicylaldehyde-Glutamine- triphenylphosphine, copper-salicylaldehyde-cysteine-pyridine, copper-salicylaldehyde-cysteine - triphenylphosphine, copper-salicylaldehyde-Aspartic acid-pyridine, copper-salicylaldehyde- Aspartic acid - triphenylphosphine, copper-salicylaldehyde- Methionine - pyridine, copper-salicylaldehyde- Methionine – triphenylphosphine.

#### Larvicidal assay

The mosquito larvae were collected from stagnant water, open sewage path and pond in Thiruvavur district. In the larvicidal assay, 5 nos of early instar larvae of Culex and Anopheles species were taken in clean sterile test tubes in 1 mL of water. They were exposed to 1 µg/ml concentration of synthesized complex in aqueous and ethanol extracts. Five instar larvae

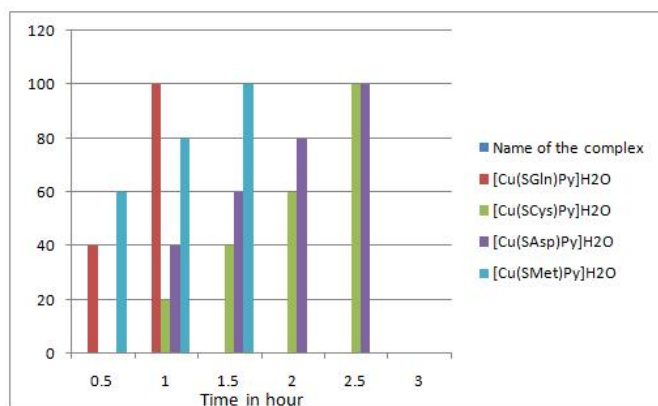
**Table 1. Mosquito larvicidal activity against Culex observed at a concentration of 1 µg/ml (aqueous)**

S. No	Name of the complex	Time(hour)						Mortality time in hour	% of larvicidal activity
		1/2	1	1 1/2	2	2 1/2	3		
1.	[CuCl(SGln)Py]H <sub>2</sub> O	2	5					After 1 h	100
2.	[CuCl(SGln)PPh <sub>3</sub> ]H <sub>2</sub> O	0	1	3	4	4	5	After 3 h	100
3.	[CuCl(SCys)Py]H <sub>2</sub> O	0	1	2	3	5		After 2 1/2 h	100
4.	[CuCl(SCys)PPh <sub>3</sub> ]H <sub>2</sub> O	1	3	4	5			After 2 h	100
5.	[CuCl(SAsp)Py]H <sub>2</sub> O	0	2	3	4	5		After 2 1/2 h	100
6.	[CuCl(SAsp)PPh <sub>3</sub> ]H <sub>2</sub> O	2	3	4	5			After 2h	100
7.	[CuCl(SMet)Py]H <sub>2</sub> O	3	4	5				After 1 1/2 h	100
8.	[CuCl(SMet)PPh <sub>3</sub> ]H <sub>2</sub> O	3	5					After 1h	100

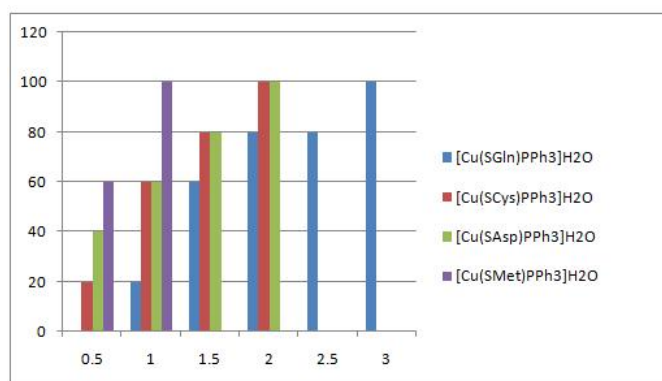
(5 nos) taken in distilled water and ethanol served as control. The tubes were kept for 5 h and observed for mortality of the larvae.

## RESULTS AND DISCUSSION

The larvicidal activity of copper complexes in aqueous extract against culex mosquito was tabulated (Table 1) and the percentage mortality of complexes were shown in Fig.1 and Fig.2. All copper complexes (aqueous extract) showed 100% mortality after 3 h.



**Fig. 1. Percentage mortality of copper pyridine complexes at different time interval**



**Fig. 2. Percentage mortality of copper triphenylphosphine complexes at different time interval**

When compared to pyridine complexes, triphenylphosphine complexes showed effective larvicidal activity. It was found that all copper complexes in ethanol extract showed 100% mortality within 1 h. Table 1: Mosquito larvicidal activity against Culex observed at a concentration of 1 µg/ml (aqueous).

## Conclusion

These synthesized copper complexes can be used as a new insecticidal agent for mosquito control.

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