



Synthesis, Characterization And Antimicrobial Study Of Nickel(II) Complex Of (E)-1-(2,6-Dihydroxyphenyl)-3-(5-Methylfuran-2-Yl)Prop-2-En-1-One

Shankar N. Ipper^{1*}, Satish A. Dake¹, Gopal K. Kakade²

¹Department of Chemistry, Sunderrao Solanke Mahavidyalaya, Majalgaon Dist. Beed (India),

²Department of Chemistry, A.C.S. College, Kille-Dharur, Dist. Beed (India)

Corresponding email address: isn13@rediffmail.com

Abstract:

The synthesis of Nickel (II) metal complex has been synthesized by using novel (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one ligand. The ligand was prepared by the Claisen-Schmidt condensation method of 2,6-dihydroxy acetophenone and 5-methylfurfural. The structure of the complex has been characterized by the analytical data, conductivity measurement, magnetic moment, UV-Vis spectra, thermal studies and XRD analysis. Analytical data shows 1:2 stoichiometry and the magnetic moment, TG-DTA suggests that Ni(II) complex has octahedral geometry. The presence of coordinated water molecules in Ni (II) complex is confirmed by thermal studies. The conductivity data reveals that the complex is non electrolyte. Antimicrobial study of complex with selected bacterial strain and fungal strain carried out and the results have been compared with commercial standards. The Ni (II) complex shows moderate to good Antibacterial and Antifungal activity.

Keywords: Antimicrobial activities, TG-DTA, XRD study, Physico-chemical property, Magnetic Susceptibility and Conductivity.

1. Introduction:

Chalcones constitute an important group of natural products, which has two aromatic rings joined by α , β unsaturated carbonyl system. The name chalcone is given by Kostanecki and Tambar [1]. The $>CO-C=C<$ moiety imparts biological characteristics to the Chalcones. Such α , β -unsaturated carbonyl group in chalcone is found to be responsible for their antimicrobial activity [2]. The metal complexes possess interesting biochemical properties, such as antitumor, antioxidant, and antimalarial, anti-fungal and antimicrobial activities [3]. The magnetic moment, TG-DTA supports the octahedral



geometry of the metal complex of chalcone. All crystals of a substance possess the same elements of symmetry. The computer program, used for indexing data was powder-X [4]. The X-ray powder diffractogram of the metal complex was used for the structural characterization and determination of lattice dimensions.

2. Materials and Methods:

2.1 Synthesis of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one ligand:

The reagents used for preparation of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one are of A.R. grade. The mixture of 2,6-dihydroxy acetophenone (0.01 mol) and 5-methylfuraldehyde (0.01 mol) is dissolved in ethanol (20 mL) and then sodium hydroxide 10 mL (40%) was added to it. The mixture was stirred for overnight till brown color ppt was observed. The progress of the reaction was monitored by TLC, from thin layer chromatography the completion of the reaction is observed. After completion of the reaction, the contents were poured into ice cold water and then acidified by dil. HCl. The solid obtained was filtered and crude product was recrystallized from ethanol to give the chalcone [5].

2.2 Synthesis of Metal Complex:

The solution of 0.02 mole of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one was taken in round bottom flask containing 30 ml of anhydrous methanolic solution and boiled for 10 minutes. A hot solution of 0.01 mole, of Nickel Nitrate in 20 ml of methanol was added drop wise to the solution of the chalcone of 5-methylfurfural to this reaction mixture, 10% alcoholic ammonia was added up to slightly alkaline pH. The complex was precipitated at 8 pH range. The pH 8-10 range was definite for these complexes [6]. The content was stirred on magnetic stirrer for one hour. The solid metal complex separated out and washed with methanol three to four times. The melting point of the complex was determined by Thiele's melting apparatus. The reactions of formation of Ni(II) complex is shown in **Figure-1**.

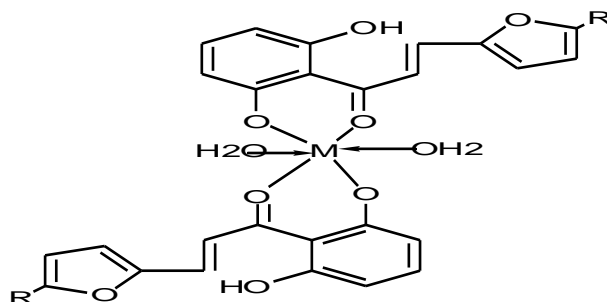
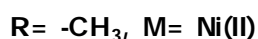
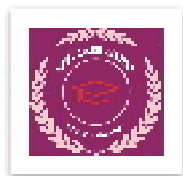


Figure-1: Metal complex of Nickel (II) with (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one





3. Results and Discussion:

3.1 Physical parameters:

Metal complex of Nickel (II) with (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one was reddish brown in color. The complex was precipitated at 8 pH range, having Melting point 280°C. The complex is insoluble in water and soluble in DMSO, DMF [7].

3.2 CHO analysis:

The carbon, hydrogen, oxygen, Nickel metal percentage in Ni (II) complex of chalcone measured at SAIF Cochin, Kerala. The calculated and measured values of CHO analysis are matching and are given in the **Table-1**.

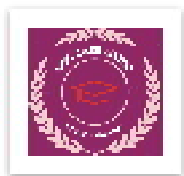
Table-1: Study CHO analysis synthesized Ni (II) complex

Metal complex	Chemical formula	Mol. Wt.	Elemental analysis : % found (calculated)						
			C	H	N	O	S	X(Br)	M
Nickel (II)	[C ₂₈ H ₂₆ O ₁₀ Ni]	581	57.86 (65.02)	4.50 (5.06)	-	27.52 (18.56)	-	-	10.09 (11.34)

3.3 Magnetic susceptibility, solution conductivity and electronic absorption spectral data

Magnetic susceptibility:

The magnetic moment values of octahedral Ni(II) complex fall in the range 2.9 to 3.4 B.M. These values of magnetic moment are slightly higher than expected from spin only values of 2.83 B.M. due to spin-orbit coupling and contribution from the ³A_{2g} And ³T_{2g} state. These values are in good agreement with the moment reported for octahedral Ni (II) complex by earlier workers [8].



Solution conductivity and electronic absorption spectral data:

The solution conductivities of 10^{-3} M solution of metal complex in DMSO were measured on EQUIPTRONICS digital conductivity meter EQ - 660 with $20 \mu\Omega$ to $200 \mu\Omega$ at 298K temperature. They are insoluble in water and soluble in DMSO, DMF. The low conductivity values in DMSO solution (10^{-3} M) are indicates non-electrolytic nature. The Solution conductivity **Ni (II) complex** was (6.00), **Figure-2**.

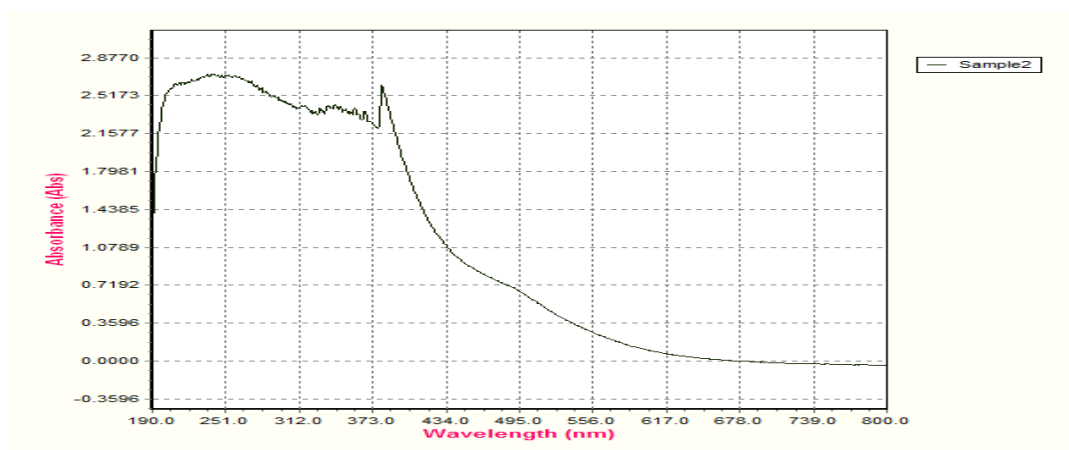


Figure-2: Electronic absorption spectrum

The magnetic moment of Ni(II) complex was (2.92 BM) and The electronic absorption spectra of Ni(II) complex was showed two bands at $26315 (380) ^3A_{2g} (F) \rightarrow ^3T_{1g} (P)$ and $29761(336) \text{ cm}^{-1} (\text{nm})$ Charge Transfer band indicating distorted octahedral geometry around the Ni(II) ion [9-10].

3.4 Infra red spectrum:

The IR spectrum of α, β -unsaturated carbonyl group has characteristic bands of chalcone at prominent bands between 1625 to 1650 per cm. The characteristic peaks in infra red spectrum give the presence of particular functional group. The region at which other absorption bands appear depends on the type of aromatic / hetero-aromatic rings as well as the substituent present on these rings. The infrared spectrum of metal complex of Nickel (II) with (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-



2-en-1-one was recorded on a Perkin- Elmer Spectrum RX-IFTIR Spectrophotometer in the range 4000-400 cm^{-1} (Table-2) using potassium bromide pellet at CIL, Chandigarh, Punjab. The stretching frequency of metal complex of Nickel (II) with (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one is represented in table number (2) and the IR spectrum in Figure-3.

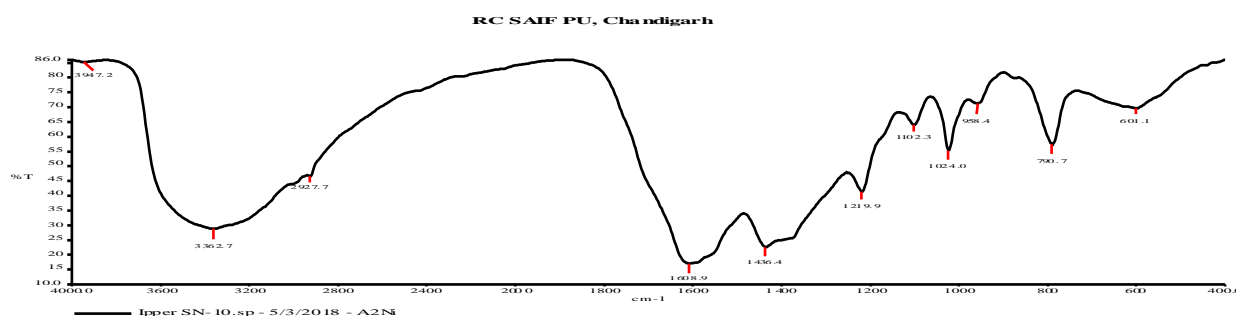


Figure-3: IR spectrum of metal complex of Nickel (II) with (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one

Table-2: IR spectral data Ni(II) complex of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one:

Ligand/ Metal complexes	ν (OH) cm^{-1}	ν (H ₂ O) cm^{-1}	ν (-CO- CH=CH-) cm^{-1}	ν (-C=O in pyron ring) cm^{-1}	ν (C- O-C) cm^{-1}	ν (C=C) cm^{-1}	Aromat ic Ring (C=C) cm^{-1}	ν (M-O) cm^{-1}
[Ni(A ₂) ₂]	2927	3362	1608	-	1024	1436	1219	601

3.5 Thermal analysis Ni(II) complex of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one:



The simultaneous thermo gravimetric, differential thermal analysis of Ni(II) complex (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one was performed in an inert nitrogen atmosphere on Perkin Elmer STA 6000 at SAIF, Cochin, Kerala. The heating rate was 10°/min and flow rate of nitrogen 50 ml/min. The reference substance used was α Al₂O₃ in platinum crucible and sample weighed in the range of 4-12 mg. The thermogram of Ni(II) complex (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one is presented in figure-2. This curve reveals that there is presence of lattice as well as coordinated water in the complex.

The thermogram of Ni(II) complex of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one (**Figure-4**) shows high thermal stability and two-step decomposition. The first step within temperature range 240-330°C with mass loss of 40.36% (calc. wt. loss 41.03%) due to two fragments of C₆H₅O₂ take place. This is confirmed by an endothermic peak at 275.86°C in DTA curve. The second step decomposition observed within 380-420°C with a weight loss of 46.32%. This may be due to the loss of two fragment of 5-methyl-furan and enone part of chalcone of metal complex. Beyond that TG curve attains a constant weight loss 14.44% level corresponding to their metal oxide that is NiO.

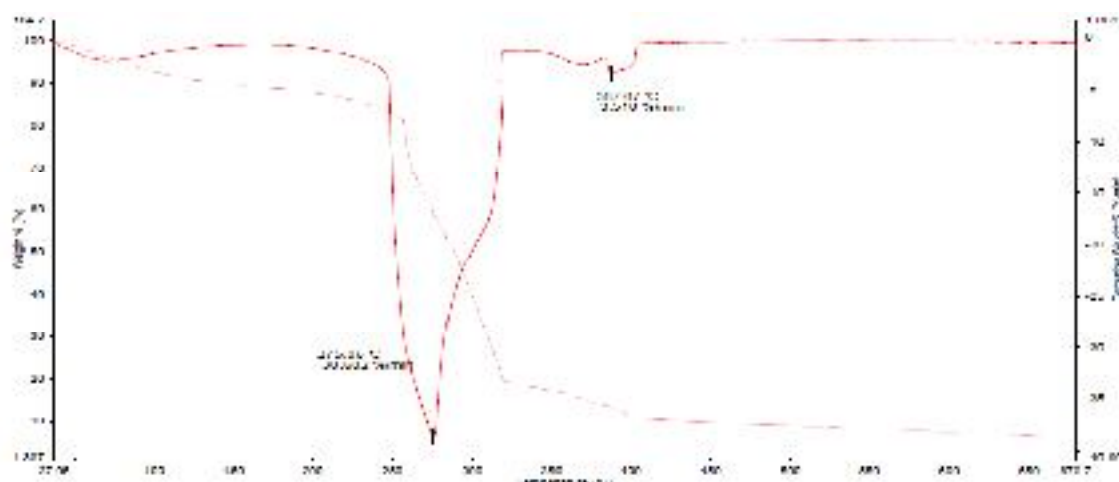
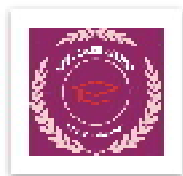


Figure-4: TG-DTA curve of Ni(II) complex of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one

Thermodynamic and Kinetic Parameters



Akahira [11], first introduced that decomposition and kinetic studies of thermal reactions are useful in determining thermodynamic and kinetic parameters like free energy, entropy change, activation energy, pre-exponential factor. Thermal decomposition studies of materials are useful in predicting thermal stability (**Table-3**).

The negative values of the entropy of activation (ΔS) indicate that the metal complex is thermally stable. ΔG is positive for the complexes revealing that the free energy of the final residue is higher than that of the initial complex, and all decomposition steps are non-spontaneous processes. Also, the value of free energy of activation, ΔG increases significantly for the subsequent decomposition stages of a given complex [12].

Table-3: Thermodynamic and Kinetic Parameters of Ni(II) complex of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one:

Metal complex	Method	Step	Decomp. Temp.	Order of Reaction	Ea(KJ mol ⁻¹)	ΔS (KJ mol ⁻¹)	ΔG (KJ mol ⁻¹)	Z (S ⁻¹)	Correlation Coefficient (r)
A ₂ Ni	H-M C-R	I	290	0.9	13.42 70.96	-161.70 -138.30	24.89 80.81	47529.95 738243	0.999 0.998
	H-M C-R	II	580	0.9	6.73 8.69	-171.09 -86.94	18.93 14.89	14335.18 355917865.6	0.987 0.996

3.6 X-ray diffraction spectral studies of metal complex of Ni (II) complex of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methylfuran-2-yl)prop-2-en-1-one:

The XRD spectral study has been done at SAIF, Cochin Kerala. The standard deviation observed for A₂Ni is 0.023% which is within the permissible limit of 2%. The observed and calculated densities are 1.9583 gcm⁻³ and 1.7392 gcm⁻³ respectively. The volume is found to be 339.43 Å³ and complex crystallizes in the monoclinic system with 1 atom per unit cell. The lattice parameters are a=13.345 Å, b=8.252 Å, c= 9.756 Å, $\alpha=90^\circ$, $\beta= 109.78^\circ$, $\gamma=90^\circ$.

Unit cell data and crystal lattice parameters for Ni (II):



Unit cell data and crystal lattice parameters

a (Å) = 13.345, b (Å) = 8.252, c (Å) = 9.765, $\alpha = 90^\circ$, $\beta = 109.78^\circ$, $\gamma = 90^\circ$, Standard deviation (%) = 0.23, Volume (V) = 339.43 Å³, Density (obs.) = 1.9583 gcm⁻³, Density (cal.) = 1.7392 gcm⁻³, Z = 4
Crystal system = Monoclinic, Space group = P2/m. (Figure-5)

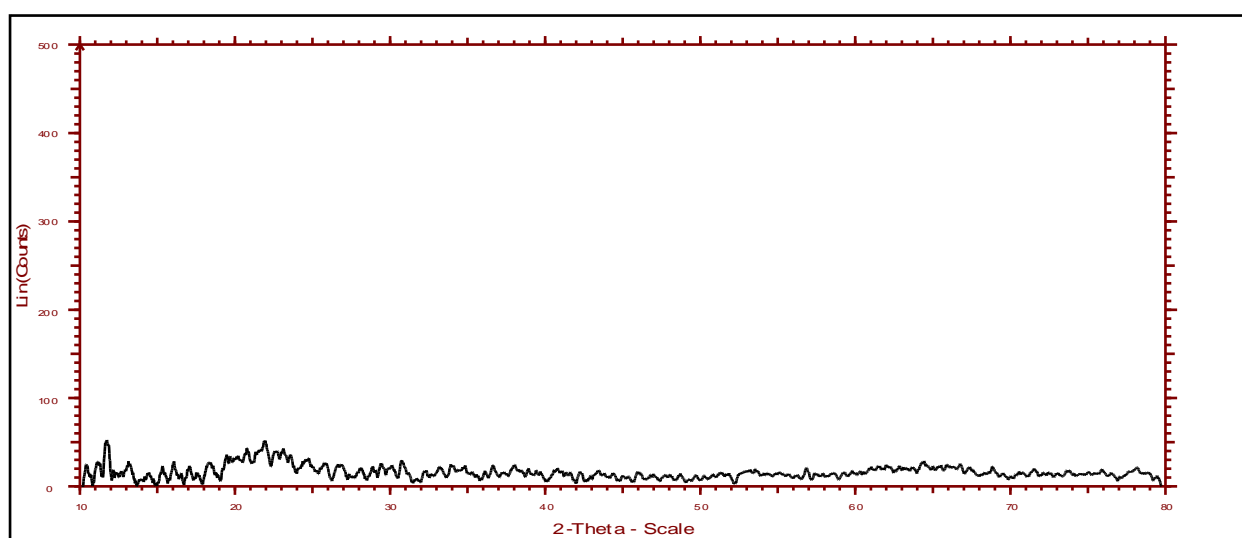


Figure-5: X-ray diffractogram of Ni (II) complex of (E)-1-(2,6-dihydroxyphenyl)-3-(5-methyl furan-2-yl)prop-2-en-1-one

3.7 Antimicrobial activity:

Antimicrobial activity was assayed by cup plate agar diffusion method by measuring inhibition zones in mm. In vitro antimicrobial activity of all synthesized compounds and standard have been evaluated against strains of The fungal toxicity of Ni(II) complex was studied *in vitro* against *Aspergillus niger* ATCC 16404, *Saccharomyces cerevisiae* ATCC 9763, *Candida albicans* ATCC10231 fungal pathogens at fixed 1% concentration.

The antibacterial activity of Ni(II) complex was studied, for evaluating antibacterial activity Gram positive and Gram negative bacterial pathogens were used. *Staphylococcus aureus* ATCC 6538, *Bacillus megaterium* ATCC 2326, *Bacillus subtilis* ATCC 6633 were Gram positive pathogens used in this study. *Escherichia coli* ATCC8739, *Salmonella typhi* ATCC9207, *Shigella boydii* ATCC 12034,



Enterobacter aerogenes ATCC13048, *Pseudomonas aerogenosa* ATCC9027, *Salmonella abony* NCTC6017 were the Gram-negative pathogens used in this study.

From the results of antimicrobial activity of ligands and complex it is clear that the complex shows enhanced activity than ligand. The increase in antimicrobial activity is due to faster diffusion of metal complexes as a whole through the cell membrane or due to the combined activity of the metal and ligands [13].

Conclusion:

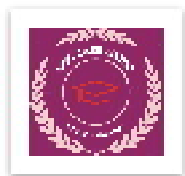
The Ni (II) complex was colored, soluble in most of the organic solvent. The stoichiometry ratios of the metal complexes are obtained has been found to be 1:2. Solution conductivity of this metal complex reveals nonelectrolytic nature. The electronic spectral data, magnetic moment, TG-DTA suggests that Ni(II) has Octahedral geometry. The CHO analysis gives C, H, and O percentage in the metal complex. The XRD parameters shows that the structure of Ni(II) is Monoclinic and has space group = P2/m. From the antimicrobial activity of ligand and complex it is clear that the complex shows enhanced antimicrobial activity than ligand.

Acknowledgement:

Authors are thankful to Principal of Sunderrao Solanke Mahavidyalaya, Majalgaon Dist. Beed (India) for providing laboratory facilities.

References:

- [1] S. V. Kostanecki and Tambar, J. Chem. Ber., 32, 1921 (1899)
- [2] N. V. Damme, V. Zalisky, A. J. Lough, M. T. Lemaire, Polyhedron, 89, 155-159 (2015)
- [3] Omar. H. Al-Obaidi, Study of the cytotoxicity effect of new Co(II), Mn (II) Ni(II), and Cu(II) complexes of chalcone on cancer and antimicrobial activity, PCE, 1(2),14-18 (2014)
- [4] S. I. Habib, P. A. Kulkarni, XRD analysis of transition metal complexes of substituted heterocyclic chalcones. World Journal of Pharmaceutical Research, 6(7), 650-657 (2017)



- [5] Omar. H. Al-Obaidi, Synthesis, spectral characterization, theoretical evaluation of new Co(II) and Mn(II) complexes of flavones., *Journal of Applicable Chemistry*, 3(1), 157-163 (2014)
- [6] S. N. Ipper, G. K. Sanap and G. K. Kakade, Synthesis, spectral Study, characterization and antimicrobial activity of Zinc(II) Complex of chalcone of pyridine-2-Carbaldehyde. *IJRASET*, 7(IX), 453-458 (2019)
- [7] S. N. Ipper, N. K. Raut and G. K. Kakde, Physico-chemical properties of furan chalcone. *Ind. J. Appl. Res.* 7(6), 683-684 (2017)
- [8] K.G. Karamunge, Y.B. Vibhute, Synthesis and structural studies of complexes of Cu(II), Ni(II) and Co(II) with 2-hydroxy chalcones; *Int. J. Chem. Sci.*, 10(1), 490-500 (2012)
- [9] M.S. Suresh, V. Prakash, Preparation and characterization of Cr(III), Mn(II), Co(II), Ni(II), Cu(II), Zn(II) and Cd(II) chelates of schiffs bases derived from vanillin and 4-amino antipyrine. *Int. J. phy. Sci.*, 5(14), 2203-2211 (2010)
- [10] Muneerah M. Al-Mogren and Abdel-Nasser M.A. Alaghaz, Synthesis, spectral and quantum chemical calculations of mononuclear Nickel(II), Copper(II) and Cadmium(II) complexes of new Schiff-base ligand. *Int. J. Electrochem. Sci.*, 8, 8669-8685 (2013)
- [11] T. Akaharia, *Sci. Papers Inst. Phy. Chem. Res.*, 9, 165 (1928)
- [12] A. A. Frost, R. G. Pearson, "Kinetics and Mechanism", John Wiley, New York. (1961)
- [13] S. K. Sengupta, B. K. Pandey, B. K. Shrivastava and V. K. Sharma, *Transition Metal Chem.*, 23, 4, 349-353 (1998).



Aliquat 366: An Efficient Catalyst For Knoevenagel Condensation Under Solvent- Free Condition

Gopal K. Kakade^{1*}, Shankar N. Ipper¹

¹*Department of Chemistry, ACS College, Kille-Dharur Dist. Beed (MS)*

²*Department of Chemistry, Sunderrao Solanke Mahavidyalaya, Majalgaon Dist. Beed (India)*

E-mail: gkkakade27@gmail.com

Abstract:

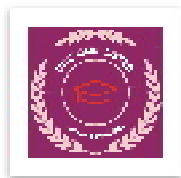
Aliquat 366 phase transfer catalyst has been utilized as a novel and efficient catalyst for the Knoevenagel condensation of aldehydes with active methylene compounds such as ethyl cyanoacetate, cyanoacetamide and malononitrile to afford substituted olefins. The reaction proceeds smoothly under mild and solvent-free conditions at room temperature and the products are obtained in excellent yields.

Keywords: Aliquat 366, Knoevenagel condensation, Grinding.

Introduction:

In recent years, solid-state organic reactions have caused great interests, which have many advantages such as high efficiency and selectivity, easy separation and purification, mild reaction conditions, and benefit to industry as well as environment. Organic synthesis in the absence of solvent is a powerful tool for the generation of structurally diverse molecules, due to their special selectivity, the ease of set-up and work-up, arousing great interest [1-3]. Moreover, solvent-free reactions sometimes are faster, taking just a few minutes rather than hours to complete, because the reactants are in close contact with each other. This aspect, coupled with the lower overall costs of running a reaction without solvent and no specially needed equipment, could become a decisive factor.

Knoevenagel condensation reactions have been extensively studied as an important carbon-carbon bond forming reaction. This condensation, various catalysts are used such as Te(IV)Cl₄ [3], Ammonium acetate [4], ZnCl₂ [5], KSF-montmorillonite [6], KF-Al₂O₃ [7], Ethylenediammonium diacetate [8], CaO [9], Mg-Al-*t*-Bu-Hydroxalcite [10], Phosphene [11],



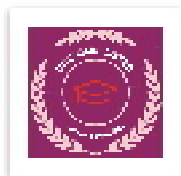
ionic liquid [bmim][BF₄] [12], PEG400 and anhydrous K₂CO₃ [13]. In recent years, chemists paid more and more attention to the clean synthesis of alkenes by condensation reactions.

Therefore, the introduction of efficient and new methods based on green methodology is still in demand. In continuation of our ongoing research to develop newer, environmentally benign methods Aliquat 336 is used as efficient catalyst for the Knoevenagel condensation of aldehydes with active methylene compounds such as ethyl cyanoacetate, cyanoacetamide and malononitrile to afford substituted olefins. Aliquat 336 is a water insoluble quaternary ammonium salt made by the methylation of tri octyl/decyl amine, which is capable of forming oil soluble salts of anionic species at neutral or slightly alkaline pH. Aliquat 336 is a versatile and affordable cation source for an entirely new family of hydrophobic ionic liquids [14]. It can form salts with anions over a wider pH range than primary, secondary or tertiary amines. For this reason Aliquat 336 finds application in environments from acid to slightly alkaline pH. It is used as a phase transfer catalyst [15].

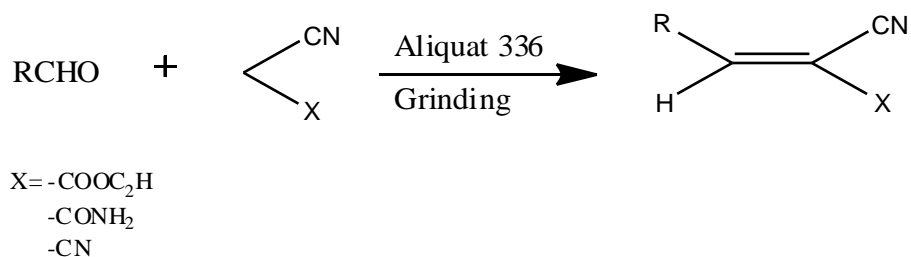
Results and Discussion:

To investigate the role of catalyst and effect of grinding, first the reaction of p-chlorobenzaldehyde, ethylcyanoacetate and catalytic amount of Aliquat 336 are ground together in mortar and pestle at room temperature, the reaction starts immediately usually with gentle heat production, the reaction mixture turned into brownish viscous liquid then to thick brownish mass and finally to free flowing powder. Surprisingly, reaction mixture turned into the desired product within a short period.

On the other hand, indication of softening for some seconds followed by immediate hardening were visually observed only in syntheses with P-chlorobenzaldehyde with malononitrile (**entry 3r**). Subsequently, the scope of the Knoevenagel condensation of other aldehydes with various active methylene compounds catalyzed by Aliquat 336 scheme-1 in grinding was investigated. We found that the Knoevenagel condensation of aldehydes with malononitrile occurred easily in the presence of grinding to form corresponding products. Both electron-rich and electron-deficient aldehydes worked well to give the corresponding arylidene derivatives in excellent yields (3a-s) in **Table-1**. The results are summarized in table 1. Miscibility of Aliquat 336 with water makes the workup process quite easy as the



catalyst can be easily removed from product simply by washing the product with water.



Scheme-1: Knoevenagel condensation using Aliquat 336 catalyst

Table 1: Aliquat 336 catalyzed Knoevenagel condensation under solvent-free condition

Entry ^a	-R	-X	Time	Yield ^b (%)	Melting point (°C)
3a	C ₆ H ₅	COOC ₂ H ₅	3 min.	92	52
3b	4-ClC ₆ H ₄	COOC ₂ H ₅	2 min.	96	163
3b	4-ClC ₆ H ₄	COOC ₂ H ₅	10 min	89	163
3c	2-CH ₃ OC ₆ H ₄	COOC ₂ H ₅	5 min.	92	69
3d	4-H ₃ COC ₆ H ₄	COOC ₂ H ₅	3 min.	93	99
3e	4-OHC ₆ H ₄	COOC ₂ H ₅	5 min.	90	171
3f	4-NO ₂ C ₆ H ₄	COOC ₂ H ₅	4 min.	95	171
3g	C ₆ H ₅ -CH=CH-	COOC ₂ H ₅	3 min.	92	114
3h	Vanillin	COOC ₂ H ₅	5 min.	94	112
3i	2-furyl	COOC ₂ H ₅	3 min.	96	84
3j	C ₆ H ₅	CONH ₂	2 min.	92	120
3k	4-ClC ₆ H ₄	CONH ₂	2 min.	93	210
3l	3-CH ₃ OC ₆ H ₄	CONH ₂	3 min.	95	204
3m	4-CH ₃ OC ₆ H ₄	CONH ₂	3 min.	94	215
3n	Vanillin	CONH ₂	3 min.	93	206
3o	4-NO ₂ C ₆ H ₄	CN	3 min.	95	84
3p	4-CH ₃ OC ₆ H ₄	CN	2 min.	94	80
3q	3-CH ₃ OC ₆ H ₄	CN	1 min.	94	92
3r	4-ClC ₆ H ₄	CN	2 min.	93	165
3s	2-furyl	CN	2 min.	95	64

^aAll the products are characterized by MP, IR and ¹H NMR

^bisolated yields after recrystallized



Experimental section:

Melting points were determined in open capillary tubes and are uncorrected. The purity of compounds was checked by Merck silica gel TLC plates, and the spots were detected by UV light absorption. The IR spectra were recorded in matrix of KBr with Perkin-Elmer spectrometer. ^1H NMR spectra were recorded on Varian NMR Spectrometer, Model Mercury Plus (400MHz) and the chemical shifts (δ) are given in ppm relative to signal for TMS as an internal standard. All the products were identified by comparison of analytical data (melting point, IR and ^1H NMR.) with those reported of authentic samples.

General procedure:

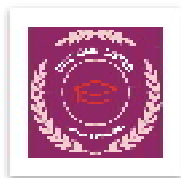
An equimolar quantity of the aldehydes (1 mmol), and active methylene compounds (1 mmol) mixed thoroughly and then 0.1 mmol of Aliquat 336 was added. The reaction mixture was ground in the mortar and pestle for 30 sec. to 5 min. at appropriate time mentioned in table 1. The reaction was monitored by TLC, on completion of reaction; the reaction mixture was treated with cold water. The product was filtered, dried, and the crude compounds were recrystallized from a mixture of ethanol and water to get the desired compound in pure form (3a-s).

Conclusion:

This new method is superior to the reported methods [5,9,10] with respect to the reaction time and simplicity of the procedure. The attractive features of this procedure are the mild and solvent-free reaction conditions, high conversions, cleaner reaction profiles and operational simplicity, all of which make it a useful and attractive strategy for the preparation of arylidene derivatives.

Acknowledgement:

Author is thankful to Principal of ACS College Kille-Dharur, Dist. Beed (MS) for providing laboratory facilities.



References:

- [1] K. Tannaka and F. Toda, Chem. Rev. 100 (2000)1025.
- [2] D. Villemain and B. Martin, Synth. Commun. 25 (1995)3135.
- [3] R. H. Khan, R. K. Mathur and A. C. Ghosh, Synth. Commun. 26(4) (1996)683.
- [4] A. K. Mitra, A. De and N. Karchaudhuri, Synth. Commun. 29(16) (1999)2731.
- [5] R. P. Shanthan and R. V. Venkataratnam, Tetrahedron Lett. 32(41) (1991)5821.
- [6] D-Q. Shi, X-S. Wang, C-S. Yao and L. Mu, J. Chem. Res. 7 (2002)344.
- [7] S-X. Wang, J-T. Li, W-Z. Yang and T-S. Li, Ultrason Sonochem. 9(3) (2002)159.
- [8] Ce. Su, Z-C. Chen and Q-G. Zheng, Synthesis. 4 (2003)555.
- [9] (a) Y. Peng and G. Song, Ind. J. Chem. 42B (2003) 924. (b) Y. Lu, Z. Ren, W. Cao, W. Tong and M. Gao, Synth Commun. 34 (2004)2047.
- [10] B. M. Choudary, M. Lakshmi Kantam, B. Kavita, Ch. Venkat Reddy and F. Figueras, Tetrahedron. 56(47) (2000) 9357.
- [11] J. S. Yadav, B. V. Subba Reddy, A. K. Basak, Boddapati Visali, Akkerala Venkat Narsaiah and Kommu Nagaiah, Eur. J. Org. Chem. (2004)546.
- [12] X. Fan, X. Hu, X. Zhang and J. Wang, Aust. J. Chem. 57(11) (2004)1067.
- [13] Y-Q. Cao, Z. Dai, R. Zhang and B. Hua, Synth. Commun. 34(16) (2004)2965.
- [14] J. Mikkola; P. Virtanen; R. Sjöholm, Green Chem., 8, 250, (2006).
- [15] K. Sato; M. Aoki; Science, 281, 1646, (1998)