

BOOK OF PROCEEDINGS

GARDEN CITY 2016

CHEMICAL SOCIETY OF NIGERIA



39th

ANNUAL INTERNATIONAL CONFERENCE, WORKSHOP AND EXHIBITION



GARDEN CITY 2016

THEME:

THE ROLE OF HYDROCARBONS AND THE APPLICATION OF
CHEMISTRY IN THE EMERGING TREND OF GLOBAL ENERGY
INNOVATIONS

DATE: SUNDAY, 18TH - FRIDAY, 23RD SEPTEMBER, 2016.

VENUE: LAW FACULTY, RIVERS STATE UNIVERSITY OF SCIENCE &
TECHNOLOGY (RSUST), PORT HARCOURT, RIVERS STATE, NIGERIA.

BOOK OF PROCEEDINGS

Table of Content

S/N	Topics and Authors	Page
1	<i>In Vitro</i> Study of Isolated Compounds from the Leaf of <i>Flabellaria Paniculata</i> Against <i>Staphylococcus Aureus</i> , <i>Pseudomonas Aeuruginosa</i> and <i>Candida Albicans</i> ; Joseph Adebisi O. Olugbuyiro ^{1*} , Olugbenga Samson Taiwo ² , Olaronke Temitope Olaniyi ¹ and Francis Adolphusobi ¹	1
2	A Comparative Evaluation/Analysis of Air Quality in Some Selected Oil Producing/Bearing Communities in Delta State of Nigeria.; Ogodu Aemurerhimen Dickson (Snr.) (Chartered Chemist)	6
3	Batch Adsorption of Cd ²⁺ , Pb ²⁺ and Ni ²⁺ with Coconut (<i>cocos Nucifera L.</i>) Shell Activated Carbon Modified with 1, 2-dihydro-1, 5-dimethyl-2, Phenyl- 4- (e) - (2, 3, 4- Trihydrophenyl) -3h-pyrazol-3-one (ddptp) Ukoha, Pius. O, Anierobi, Peter. O and Nicholas, Eno-obong S.	22
4	Delonixregia Extracts as Non – Toxic Corrosion Inhibitor for Acid Corrosion of Mild Steel in HCl Solution Olusegun K. Abiola ^{1*} , Abdulraman O.C. Aliyu ² and Suleiman Muhammed ³	33
5	Assessment of Some Metals in Roadside Dust from Damaturu, Yobe State, Nigeria Zaynab Muhammed Chellube, Joseph Clement Akan and Emmanuel Mshelia	38
6	Anti-Oxidant and Elemental Analysis of the Fruit of <i>Sarcocephalus Latifolius</i> (Smith Bruce) Isah Yinusa, Woli Ajoke Balkees and Hassan Adam Adeiza	43
7	Proximate, Mineral and Anti-Nutrient Composition of <i>Allium Sativum</i> (Garlic) Consumed in Uyo, Akwa Ibom State, Nigeria; E. I. Uwah and E. A. Moses	47
8	Phytoconstituents, Proximate Composition and Mineral Contents of Soybean (Glycine Max) Flour; Useh, M. U., ^{1*} Adebisi, A. B., ¹ and Dauda, M. S. ²	52
9	Equilibrium, Kinetics and Thermodynamic Properties of Methylene Blue Adsorption by Termite Mound; ¹ Anebi, P. O., ² Ugbe, F.A., ³ Ikudayisi, A.V.	57
10	Petrochemicals: Upstream to Plastic Industry Using Ideas, Methods, Developments and Procedures in Developing Countries.; Ogodu Aemurerhimen Dickson (Snr.) (Chartered Chemist)	63
11	Kinetics of the Photocatalytic Degradation of Chlorendic Acid (Flame Retardant) in Aqueous TiO ₂ Suspension; Boisa N., and Obunwo C.C.	78
12	Gas Chromatography-Mass Spectrometric Analysis of Methanolic Leaf Extract of <i>Blighia Sapida</i> ; Theresa I Edewor and Nimotalai O Kazeem	83
13	Characterization and Distributions of Aliphatic and Polyaromatic Hydrocarbons in Soils of Oil Sand Deposits Area of Ondo State, Nigeria; T.A. Adedosu ^{1*} , O.K. Adeniyi ² , O.H. Adedosu ³	89
14	Comparative Assessment of the Environmental Dynamics of Dissolved Organic Carbon (DOC), Nitrogen (DON), Phosphorus (DOP) and Characterization of DOM in Subtropical Wetlands of Some Northern Nigerian States.	98
15	Synthesis, Characterization and Kinetic Studies of Fe (ii) and Cu (ii) Complexes of Nicotinic Acid Hydrazide.; ¹ O.W. Salawu, ² M.S. Iorungwa and ³ M.U. Adaji.	109
16	Elemental Analysis and Cyanic Acid Contents of Selected Cassava Based Foods in Nigeria; Orishadipe Abayomi, Afolayan Michael, Odukomaia Doyinsola, Aguzue Onyinye & Bwai Macham David	115
17	Production of Methyl Ester (Biodiesel) Using Melon Seed Oil as a Raw Material ¹ Agbo, I.U. & ² Onyezeka, E.G.	119
18	Studies on Biosorption of Co ²⁺ , Cr ³⁺ and Cd ²⁺ from Aqueous Solutions by <i>Piliostigma Malabaricum</i> Seed Pod. Onwu, F. K and Amadi, O. K.	121
19	Corrosion Inhibition Properties of <i>Commiphora Africana</i> (A. Rich.) Engl. Gum Exudates on Mild Steel in Alkaline Medium.; ¹ Odoemelam, S.A., ² Onukwube, N.D. and ³ Eddy, N.O.	127
20	Ogodometrics: ISO Approval/Certifications/Product Design, Innovation Using Globally Harmonized International Reference Standards in Songhai Delta Amukpe Sapele as Targeted World Collaborating Centre.; Ogodu Aemurerhimen Dickson (Snr.) (chartered Chemist)	134
21	Chemical Constituents of Sandbox Tree (<i>hura Crepitans Linn.</i>) and Anti- Patotoxic Activity of the Leaves and Stem Bark Extracts; Ganiyat K. Oloyede ^{1*} , Oluwatosin A. Adaramoye ² and Mutairu B. Olatinwo ¹	161
22	Studies on the Molar Ratios of Optimal Interactions of Nr-Pvc Blend using Ir Spectroscopy ¹ Okoye, O. N. N., ² Eboatu, A.N	175
23	ANTIBACTERIAL AND ANTIOXIDANT ACTIVITY OF <i>JUSTICIA SPICIGERA</i> EXTRACTS: ACTIVITY ENHANCEMENT BY ADDITION OF METAL SALTS. Ohenhen O. N ¹ , Njoku P. C ² , Igara C. E ³ .	180

S/N	Topics	Page
24	Kinetics and Mechanism of the Oxidation of Potassium Trisoxalatoferate(iii) by Permanganate Ion in Aqueous Hydrochloric Acid Medium; ¹ B.O. Ogori, ² Y.N Lohdip and ³ J.N Egila	187
25	The Importance of Chemistry Practice and Research in the Oil and Gas Sector Ogodu Aemurerhimen Dickson (Snr.) (Chartered Chemist)(A2652) (Hnd, Pgd, M.sc., Ph.d (in- View), Aisl, C. Chem., Miccon, Mcsn, Mnes, Mspe)	193
26	The Effect of Extracts of Crinum Jugas on the Acute Toxicity of the Vernonia Amygdalina Root Poisoned Guinea Pigs; Ogbuanu Cyril C, Amujiogu Steve N, Nwagu Lauretta N, Chime Charlse C, and Arinze-Nwosu Uche L	200
27	Kinetics and Sorption Models of Unmodified and Modified Okra Stem Derived Cellulose for the Removal and Recovery of Hg and as Ions from Aqueous Environment; Obidiozor, C.J. and Okoro I.A	205
28	Synthesis, Characterization and Corrosion Inhibitory of Co (ii), Ni (ii), Cu (ii), And Zn (ii) Complexes Derived from Nicotinic Acid Hydrazide.; ¹ O.W. Salawu, ² R.A. Wuana, ³ J.T. Ashimom	215
29	Preparation and Characterization of Activated Carbon from <i>Hura Crepitans Linn</i> Seed Shells ¹ e. W. Nsi ¹ , A. E. Akpakpan ¹ And U. D. Akpabio	221
30	Phytochemical & Antimicrobial Studies of Crossopteryx Febrifuga Leaf Extract Against Methicillin-Resistant Staphylococcus Aureus Skin Infections; Nma N. Y.* ¹ , Mann A. ² , Ndamitso M. M. ² And Muhammad B. M. ³	227
31	Antibacterial and Antioxidant Activity of <i>Justicia Spicigera</i> Extracts: Activity Enhancement by Addition of Metal Salts.; Ohenhen O. N ¹ ., Njoku P. C ² ., Igara C. E ³ .	234
32	Comparison of Heavy Metals Pb, Cu, Ni, Zn And Al In Human Scalp Hair: A Case Study of Port-harcourt and Okigwe; Ngwu, Comfort M and Onwu, Francis K.	240
33	Geochemical Assessment of the Petroleum Generative Potential of Ekenkpon and Nkporo Shale Formations in the Calabar Flank South Eastern Nigeria; P. A Neji ¹ ; H A Neji ² ; Tnganje ³ ; O.O Oli ⁴ ; U J Ibok ⁵	249
34	Synthesis, Characterization and Biological Activity of New Mannich Base 1-((4-chlorophenyl)(2,5-dioxopyrrolidin-1-yl)methyl)urea; M.r. Abdullahi * ¹ , S. Rajeswari ²	262
35	Nanotechnology and Implementation in Natural Products Green Chemistry Processes; M.M. Adeyemi* ¹ , P.E. Omale ¹ ; O.A. Olawuyi ¹ A. Ngokat ¹ and A. Bhattacharya ²	267
36	Sorption Kinetics of Methylene Blue on Adsorbents Derived from <i>Delonix Regia</i> Seed Pods and <i>Vigna Subterranea</i> Fruit Hulls; L.K. Akinola ¹ ; Ali Ibrahim ² and Mohammed Mohammed ²	275
37	Quantum Chemical Studies on the Efficiency of 2-Hydroxyethylethylendiamine and Diethylenetriamine as Corrosion Inhibitors K. J. Uwakwe ¹ , P. C. Okafor ² ,	285
38	Comparative Studies of the Inhibitive Potentials of Red Peanut Skin Extract and <i>Lasianthera Africana P. Beauv.</i> (Nkanka Leaf) Mucilage on Mild Steel Corrosion in Sulphuric Acid Solution. ¹ A.O. James, ² Kalu Nnenna and ³ akaranta O.	291
39	<i>In Vitro</i> Antioxidant Activity of <i>Heinsia Crinata</i> Root Extracts.; *Ita, Basil N and Willie, Akaninyene H.	298
40	Evaluation of Medicinal Composition of <i>Cyathea Latebrosa</i> Leave Extract Ikpa, C.B.C ^{1*} ., Ikezu, U.J. M ² and Ugochukwu, M.I ³	304
41	Determination of Heavy Metals in <i>Telfeiria Occidentalis</i> (Pumpkin Leaves) Grown at Orimekpang – Ochor Village, Boki Local Government Area, Cross River State, Nigeria.; ¹ Ikezu, U. J., ² Udeozo, I.P. and ³ Odok, J.M.	309
42	Proximate Analysis and Production of Bioethanol from Post-Harvest Pineapple and Watermelon Waste Fruit G.U. Igelige, P. Ekwumemgbo, G.I, Ndukwe and J.I Achika	314
43	Evaluation of Chemical Composition of <i>Murraya Koenigii</i> (linn) Spreng Leaf ¹ C. E. Igara, ² D. A. Omoboyowa, ³ A. A. Ahuchaogu, ¹ N. U. Orji and ¹ M. K. Ndukwe	320
44	Studies on the Energy Properties and Fuel Potentials of Selected Indigenously Processed Plant-Based Bio-Resources used as Sources of Sustainable Flame in North Central Nigerian Communities J. S. Gushit* ¹ , J. T. Shimuan ¹ , M. O. Ajana ¹	324
45	Pollution Index of a Crude Oil Producing Community in Niger Delta: A Case Study of Ibeno Local Government Area of Akwa Ibom State, Nigeria; *Godwin Asukwo Ebong ¹ , Helen Solomon Etuk ¹ and Imeh Edet Essien ²	330
46	Synthesis and Biological Activities of Metal Complexes of Some Organic Bases Emwanta D. O. and Ngochindo R. I.	339

S/N	Topics	Page
47	Assessment of Rainwater Quality of Bichi Local Government Area of Kano State of Nigeria Emmanuel Bernard	343
48	Effect of Advance Organizers in the Teaching of Chemistry in Secondary Schools: Anambra State as A Case Study; Enekwechi E.e.	349
49	Removal of Contaminants from Industrial Wastewater using Photocatalyst; Ekwere, Ifiok Okon	358
50	Levels, Spatial Distribution and Ecological Risk Assessment of Cadmium and Lead in Surface Soil of Nsukka Industrial Cluster Areas, South-East Nigeria N. R. Ekere *, B.U. Ngang, J.N Ihdioha, P.O.Ukoha	361
51	Low Cost/Waste Catalyst for Fatty Acid Methyl Ester Production M. O. Ekeoma ^a , P. A. C. Okoye ^b , V.I.E., Ajiwe ^b	366
52	Coordination Chemistry in Oil Industry; ehirim, Appolinus I. And Ogwuegbu, Martin O.C.;	378
53	Using Index Models for Heavy Metal Pollution Estimation of Sediments from Bomu and Oginigba Rivers Marcus, A. C. and Edori, O. S.	389
54	Effect of Seasonal Water Fluctuation of a Water Body on Antioxidant Activity of Selected Plants of Lower Phylum(a Case Study of Nchestream) ¹ Duru M.K.C; ² Akubugwo E.I; ³ Chinyere G.C; ³ Amadi B.A; ⁴ Agomuo E.N; ⁵ Alisa, C. O; ⁶ Njoku V. O; and ⁴ amadi	395
55	Characterization of Violet Tree (<i>Securidaca Longepedunculata</i>) Root Powder ¹ Donatus, R.B. ¹ , ¹ Barminas, J.T., ¹ Maitera, O.N., ² Riki, .E.Y	403
56	Determination of the Saccharin Content in Some Ice Creams Consumed in Port Harcourt Dibofori-Orji Amalo Ndu & Didia Lucky Ejikeme	405
57	Evaluation of Chemical Composition of <i>Murraya Koenigii</i> (linn) Spreng Leaf ¹ C. E. Igara, ² D. A. Omoboyowa, ³ A. A. Ahuchaogu, ¹ N. U. Orji and ¹ M. K. Ndukwe	410
58	Phytochemical Studies and Aphrodisiac Activity of <i>Byrsocarpus Coccineus</i> Root Extract ¹ Balarabe M. M., ² Mann A., ² Ndamitso M. M. Adeyemi O. Y. H. And ³ Nma N. Y.	415
59	Gc-Ms Analysis of Leaf, Stem-Back and Root Extracts of <i>Alstonia Boonei</i> Babatunde O* ¹ ; Godwin R. E. ¹ ; Wahab N.O. ²	421
60	Isolation and Characterization of 2, 4 - Dihydroxycinnamic Acid from the Stem Bark of <i>Adina Microcephala</i> Delile; ^{1,2} R.G. Ayo, ² J.D. Habila, ² J. I. Achika and ² O.O. Akinwande	425
61	Groundwater Quality in Okitipupa Township of Ondo State, Nigeria; ^{*1} Ayedun, H. and Ediagbonya T. F	428
62	Preparation and Characterization of Activated Carbon from <i>Hura Crepitans Linn</i> Seed Shells E. W. NSI ¹ , and A. E. Akpakpan ^{2*} U. D. Akpabio ³	434
63	Characterization of Beach Water and its Effect on Nearby Groundwater Quality in Eti-Osa and Ibeju-Lekki Local Government Areas, of Lagos State, Nigeria.; V. E. *Agbazue and B. U. Ukazu	440
64	Comparative Studies of the Inhibitive Potentials of Red Peanut Skin Extract and <i>Lasianthera Africana P. Beauv.</i> (nkanka Leaf) Mucilage on Mild Steel Corrosion in Sulphuric Acid Solution. [^] A.O. James, [^] Kalu Nnennaand [^] akaranta O.	452
65	Assessment of Physicochemical Properties of Clay Samples Obtained From Ashaka, Potiskum and Tango (Nigeria) and a Commercial Bentonite Obtained from Kofar Ruwa Market.; Abdullahi S. L ¹ and A.A Audu ²	459
66	Extraction and Characterization of Magnesium Chloride from Different Brackish Water Sources in Rivers State; ¹ Konne, J.L., ² Ujile, A.A. and ¹ Ogolo, J.J.	465
67	Neutron Activation Analysis (NAA) and X-ray Fluorescence Analysis (EDXRF) Of <i>Corchorus Tridens</i> Linn ¹ Umar S. Gwarzo, ² Gimba, C.E., ³ Adeyemo, D.J. and ² Paul, E.D.	470
68	Synthesis and Evaluation of Substituted Benzimidazole Motifs for Preliminary Antimicrobial Drug Target Olayinka O. Ajani ^{1*} , Damilola V. Aderohunmu ¹ , Shade J. Olorunshola ² , Bamidele M. Durodola ¹ and Olatunde M. Ogunleye ¹	476
69	Heterogeneous Catalytic Efficiency of Silica Sulfuric Acid Toward the Synthesis of Substituted Pyrimidin-2(1 <i>h</i>)-one Derivatives. Olayinka O. Ajani [*] , Taiwo F. Owoeye, Ifedolapo O. Olanrewaju, Adebusayo E. Adedapo and Christiana O. Ajanaku	482
70	Phytochemical Screening and Pharmacognostic Evaluation of Leaves of <i>Enantia Chlorantha</i> Oliv. (OMO) Akpan, I. O [*] ., Ogali, R. E., Achugasim, O.	488

HETEROGENEOUS CATALYTIC EFFICIENCY OF SILICA SULFURIC ACID TOWARD THE SYNTHESIS OF SUBSTITUTED PYRIMIDIN-2(1H)-ONE DERIVATIVES

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ABSTRACT

Pyrimidine template is a highly privileged motif for the development of molecules of biological and pharmaceutical interest due to its prebiotic nature to life. This present study deals with the synthesis of pyrimidin-2(1H)-one derivative from chalcones by the action of silica supported sulfuric acid (SSA) or conventional refluxed in concentrated hydrochloric acid. The chemical structures were confirmed by analytical data and spectroscopic means such as UV, IR, mass spectra, ¹H and ¹³C NMR. SSA was found to be efficient method for the quantitative transformation to pyrimidine frame work. It can be re-used after simple washing with chloroform thereby rendering this procedure more economical.

Key Words: Spectroscopic means, chalcones, pyrimidine, 4-phenylbut-3-en-2-one.

INTRODUCTION

Over the years, tremendous amount of literature have been accumulated on pyrimidine heterocycle, owing to its widespread application in medicinal research and occurrence in many biological entities valuable to life¹. For instance, pyrimidine moiety is the core structure in biomolecules such as nucleic acids components (uracil, thymine and cytosine) and vitamin B1, and is an important constituent of numerous drug molecules in many therapeutic areas². The successful application of pyrimidine derivatives in many ways, their utility in applied chemistry and in more fundamental and theoretical studies has made the literature of the subject to be correspondingly vast. Many commercially available drugs are pyrimidine-based some of which are pyrimethamine **1**, trimethoprim **2**, ampicillin **3**, idoxuridine **4**, hexitidine **5** and phenobarbital **6** (Fig. 1)³. Diverse methods have been reported for the synthesis of substituted pyrimidines, the commonest being the reaction of 1,3-dielectrophiles with nitrogen donors such as urea⁴, thiourea⁵, guanidine⁶, amidine⁷, benzamidine⁸ and formamidine⁹. Thus, it is conceivable to develop a series of pyrimidinones using SSA catalyst technique and also compare it with the traditional method heating approach in conc. HCl.

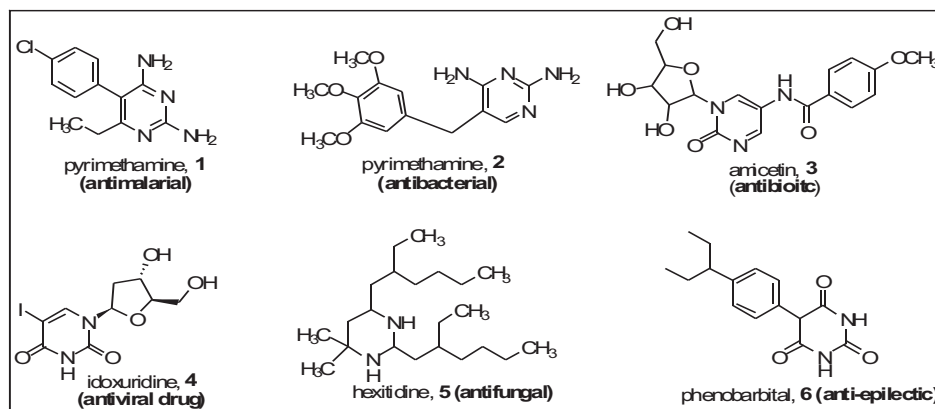


Fig. 1: Selected pyrimidine-based drugs commercially available in the market.

MATERIALS AND METHOD

General Condition: Melting points were determined in open capillary tubes on a Stuart melting point apparatus and were uncorrected. Infrared spectra were recorded on a Shimadzu Spectrometer. The Ultraviolet spectra were run on a Genesys Spectrometer using acetone solvent. ^1H and ^{13}C NMR were run on JEOL-JNM-GX-300 spectrometer at 300 MHz and 75 MHz respectively using DMSO-d_6 . Mass spectra were run on Finnigan MAT 312 machine. All compounds were routinely checked by TLC on silical gel G plates using $\text{CHCl}_3:\text{CH}_3\text{OH}$ (9:1, v/v) solvent system. The elemental analysis (C, H, N) of compounds were performed using a Carlo Erba-1108 elemental analyzer.

General procedure for chalcones (7a-g). To a solution of sodium hydroxide (2.5g) in water (20 mL), was added ethanol (10 mL) with continuous stirring until it cools down to room temperature. To this solution was added a mixture of appropriate ketone (14.15 mmol) and benzaldehyde (14.15 mmol or 28.30 mmol*) drop-wisely with continuous stirring at room temperature for 30 min. The resulting solution formed colored precipitate which was filtered by suction, washed and recrystallized from ethanol to afford **7a-g**. Where * = double molar equiv. of benzaldehyde.

General procedure for synthesis of pyrimidinone derivatives (8a-g)

Method I: A mixture of any of chalcones **7a-g** (10 mmol) and urea (1.30 g, 21 mmol) was ground in mortar and quantitatively transferred into a 250 mL round-bottomed flask containing ethanol (30 mL). Later, concentrated hydrochloric acid (10 mL) was drop-wisely added with continuous stirring and the reaction mixture was refluxed for appropriate time and reduced by evaporation to half of its original volume. It was then cooled to room temperature and neutralized with 30% sodium hydroxide and left in the freezer chest over night. The solid product obtained was recrystallized from ethanol to afford the corresponding pyrimidinone **8a-g** in moderate to good yields.

Method II: To a mixture of any of chalcones **7a-g** (10 mmol), urea (1.30g, 21 mmol) and ethanol (20 mL), was added a catalytic amount of SSA (100 mg, 0.26 mmol), and the reaction mixture was reflux for appropriate time. The SSA catalyst was extracted with chloroform (20 mL) and removed from the entire solution. The remaining solution was reduced to half its volume and cooled to room temperature. It was neutralized with 30% sodium hydroxide and left in the freezer chest over night. The solid product obtained was recrystallized from ethanol to afford the pyrimidinone **8a-g** in good to excellent yields.

4-Methyl-6-phenyl-5,6-dihydropyrimidin-2(1H)-one (8a): UV-VIS $\{\lambda_{\text{max}}(\text{Log})\}$: 325 (3.96), 274 (3.33), 244 (3.78), 226 (3.44), 202 (3.13). IR [cm^{-1} , KBr]: 3241 (N-H), 2928 (CH aliphatic), 1685 (C=O), 1612 (C=C), 1575 (C=N). ^1H NMR (300 Hz, δ ppm, DMSO-d_6): 8.0 (s, 1H, NH, D_2O exchangeable), 7.26-7.40 (m, 5H, Ar-H), 4.90 (t, 1H, CH_2 , $J=7.0$ Hz), 1.94 (s, 3H, CH_3), 1.91-1.66 (m, 2H, CH_2 , $J=7.0$ Hz). ^{13}C NMR (75 Hz, δ ppm, DMSO-d_6): 180.1 (C=O), 160.2, 143.5, 128.7, 128.5, 128.5, 126.9, 126.9, 126.7, 47.7, 40.0, 22.1 (CH_3).

4-(4-Ethylphenyl)-6-phenyl-5,6-dihydropyrimidin-2(1H)-one (8b): UV-VIS $\{\lambda_{\text{max}}(\text{Log})\}$: 310 (3.68), 265 (3.86), 230 (3.97), 215 (3.77). IR [cm^{-1} , KBr]: 3133 (N-H), 1685 (C=O), 1570 (C=N). ^1H NMR (300 Hz, δ ppm, DMSO-d_6): 8.0 (s, 1H, NH, D_2O exchangeable), 7.27-7.40 (m, 7H, $2\times$ Ar-H), 7.78 (d, 2H, Ar-H, $J=7.5$ Hz), 4.90 (t, 1H, CH_2 , $J=7.0$ Hz), 1.91-1.66 (m, 2H, CH_2 , $J=7.0$ Hz), 2.60 (q, 2H, CH_2 , $J=8.0$ Hz), 1.25 (t, 3H, CH_3 , $J=8.0$ Hz). ^{13}C NMR (75 Hz, δ ppm, DMSO-d_6): 164.6 (C=O), 160.1, 146.7, 143.5, 137.8, 128.5, 128.5, 127.8, 127.8, 127.0, 127.0, 126.9, 126.9, 126.7, 47.3 (CH), 42.7 (CH_2), 28.2 (CH_2), 14.5 (CH_3).

4-Phenyl-3,4,4a,5,6,7-hexahydro-2H-cyclopenta[d]pyrimidin-2-one (8c): UV-VIS $\{\lambda_{\text{max}}(\text{Log})\}$: 328 (4.12), 274 (3.39), 247 (3.41), 208 (4.02). IR [cm^{-1} , KBr]: 3295 (NH), 2928 (CH aliphatic), 1690 (C=O), 1600 (C=C), 1565 (C=N). ^1H NMR (300 Hz, δ ppm, DMSO-d_6): 8.01 (s, 1H, NH, D_2O exchangeable), 7.25-7.41 (m, 5H, Ar-H), 4.92 (d, 1H, CH), 2.67-2.84 (m, 5H, Cp-H), 1.22-1.41 (m, 4H, $2\times\text{CH}_2$, $J=7.1$ Hz). ^{13}C NMR (75 Hz, δ ppm,

DMSO- d_6): 208.4 (C=O), 150.0, 146.1, 142.9, 135.0, 135.0, 128.1, 128.1, 115.0, 115.0, 39.1(CH₂), 23.8(CH₂), 20.4(CH₂). MS m/z 214 [M^+ , 12.5%], 137 [M^+ - Ph, 75%], 109 [M^+ - Ph - CO, 100%].

7-Benzylidene-4-phenyl-3,4,4a,5,6,7-hexahydro-2H-cyclopenta[d]pyrimidin-2-one (8d): UV-VIS $\{\lambda_{\max}(\text{Log})\}$: 330 (3.98), 208 (4.14). IR [cm^{-1} , KBr]: 3387 (NH), 1685 (C=O), 1612 (C=C), 1575 (C=N). ¹H NMR (300 Hz, δ ppm, DMSO- d_6): 8.0 (s, 1H, NH, D₂O exchangeable), 7.27-7.60 (m, 10H, 2 \times Ar-H), 6.34 (s, 1H, CH), 4.91 (d, 1H, CH, J = 7.0 Hz), 2.69 (t, 1H, CH, J = 7.0 Hz), 1.22-2.02 (m, 4H, 2 \times CH₂, J = 7.1 Hz). ¹³C NMR (75 Hz, δ ppm, DMSO- d_6): 163.0 (C=O), 160.1, 141.5, 137.1, 135.2, 130.8, 128.6, 128.6, 128.5 (four times), 128.1, 128.1, 127.9, 125.9, 49.9, 45.3, 33.6 (CH₂), 31.3 (CH₂).

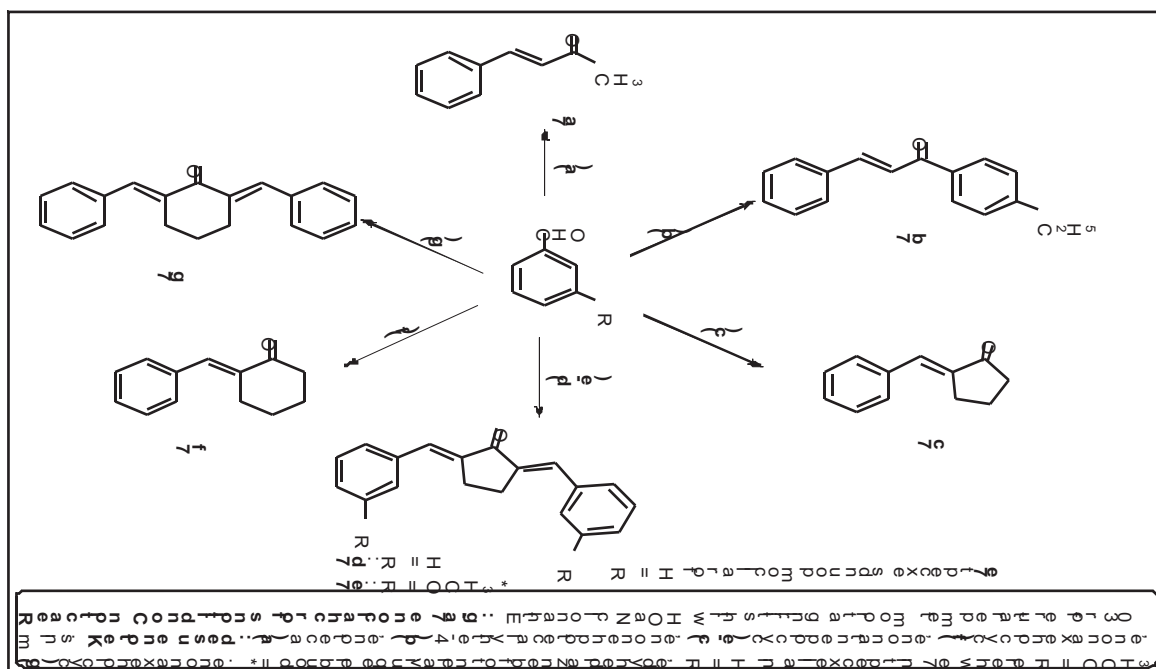
7-(3-Methoxybenzylidene)-4-(3-methoxyphenyl)-3,4,4a,5,6,7-hexahydro-2H-cyclopent- a[d]pyrimidin-2-one (8e): UV-VIS $\{\lambda_{\max}(\text{Log})\}$: 366 (3.98), 345 (3.77), 210 (4.14). IR [cm^{-1} , KBr]: 3387 (NH), 1685 (C=O), 1612 (C=C), 1575 (C=N). ¹H NMR (300 Hz, δ ppm, DMSO- d_6): 8.0 (s, 1H, NH, D₂O exchangeable), 6.82-7.59 (m, 8H, 2 \times Ar-H), 6.35 (s, 1H, CH), 4.90 (d, 1H, CH, J = 7.0 Hz), 3.84 (s, 6H, 2 \times CH₃, J = 7.0 Hz), 1.81-2.32 (m, 5H, Cp-H, J = 7.1 Hz).

4-Phenyl-4,4a,5,6,7,8-hexahydroquinazolin-2(3H)-one (8f): UV-VIS $\{\lambda_{\max}(\text{Log})\}$: 375 (3.69), 344 (3.87), 210 (4.02). IR [cm^{-1} , KBr]: 3387 (NH), 1685 (C=O), 1600 (C=C), 1573 (C=N). ¹H NMR (300 Hz, δ ppm, DMSO- d_6): 8.0 (s, 1H, NH, D₂O exchangeable), 7.27-7.41 (m, 5H, Ar-H), 4.91 (d, 1H, CH, J = 7.0 Hz), 2.19 (q, 1H, CH, J = 7.0 Hz), 1.19-1.41 (m, 8H, 4 \times CH₂, J = 7.1 Hz). ¹³C NMR (75 Hz, δ ppm, DMSO- d_6): 164.7 (C=O), 160.1, 137.1, 128.5, 128.5, 128.1, 128.1, 125.9, 49.8, 41.9, 33.8, 27.0, 24.8, 24.2.

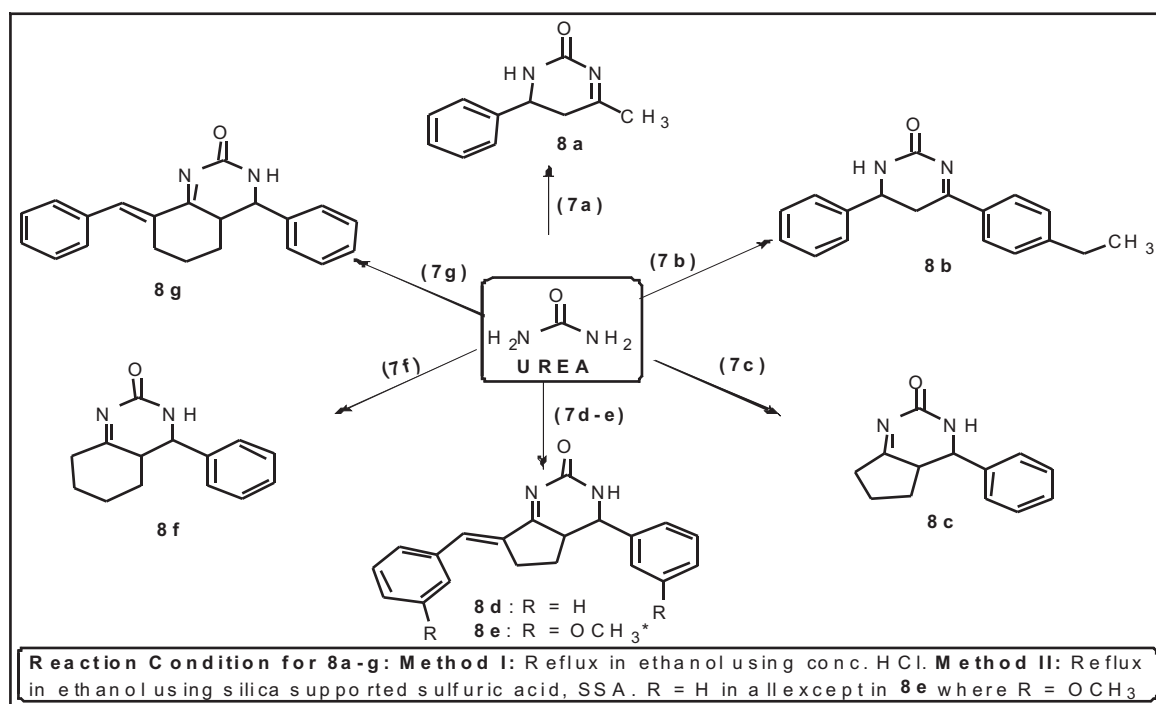
RESULTS AND DISCUSSION

In continuation of our recent works concerned with the synthesis of a variety of heterocyclic systems for biological evaluation¹⁰, we report here on the facile synthesis of substituted pyrimidine motifs under the influence of silica sulfuric acid (SSA) as heterogeneous catalyst. The synthetic route of the precursor, α -unsaturated carbonyls, otherwise known as chalcones, was as illustrated in Scheme 1 wherein α -unsaturated carbonyls (**7a-g**) were synthesized via condensation of benzaldehyde with ketones in basic medium. Compounds **7a-g** were formed in good yields via a continuous stirring and agitation at room temperature. They were subsequently reacted with urea under two different conditions which include the common technique in the presence of concentrated HCl (*Method I*) and the proposed technique using solid support catalyst, Silica Sulfuric Acid (SSA) (*Method II*), to afford pyrimidin-2(1H)-one derivatives (**8a-g**).

As a case study, the condensation of an equimolar mixture of benzaldehyde and acetone afforded 4-phenylbut-3-en-2-one, **7a**. The reactive intermediate chalcone **7a** was subsequently treated with urea in ethanol in the presence of either concentrated hydrochloric acid (*Method I*) or Silica Sulfuric Acid, SSA (*Method II*) under reflux at 140 °C to afford 4-methyl-6-phenyl-5,6-dihydropyrimidin-2(1H)-one, **8a**. This procedure was repeated for the chemical transformation of other chalcones to their corresponding pyrimidinone derivatives **8b-g** (Scheme 2). In *Method I*, upon completion (TLC), the reaction was worked up to afford **8a** in low yield 44% after refluxing for 8 h. However, in *Method II*, where conc. HCl was replaced with solid support catalyst SSA, the reaction time did not only reduce drastically to 3 h but also led to the formation of the product **8a** at a higher yield, 77% (Fig. 2). The SSA catalyst was recovered with chloroform (20 mL).



Scheme 1: The pathway for the synthesis of α,β -unsaturated carbonyl, chalcones, **7a-g**.



Scheme 2: The pathway for the synthesis of substituted pyrimidin-2(1*H*)-ones, **8a-g**.

The resulting filtrate was reduced to half its volume and cooled. It was neutralized with ammonium hydroxide and filtered by suction to afford **8a**. In a nutshell, it was observed that SSA did not only emerge as an efficient catalyst in this study but also afforded the pyrimidinone products **8a-g** in higher yields (75-95%) within smaller reaction time (3-4 h) compared with concentrated hydrochloric acid which gave smaller yields (45-71%) at higher reaction time of 8-9 h (Fig. 2). The physico-chemical parameters and analytical data were as shown in Table 1 and were consistent with the proposed structures of **8a-g**.

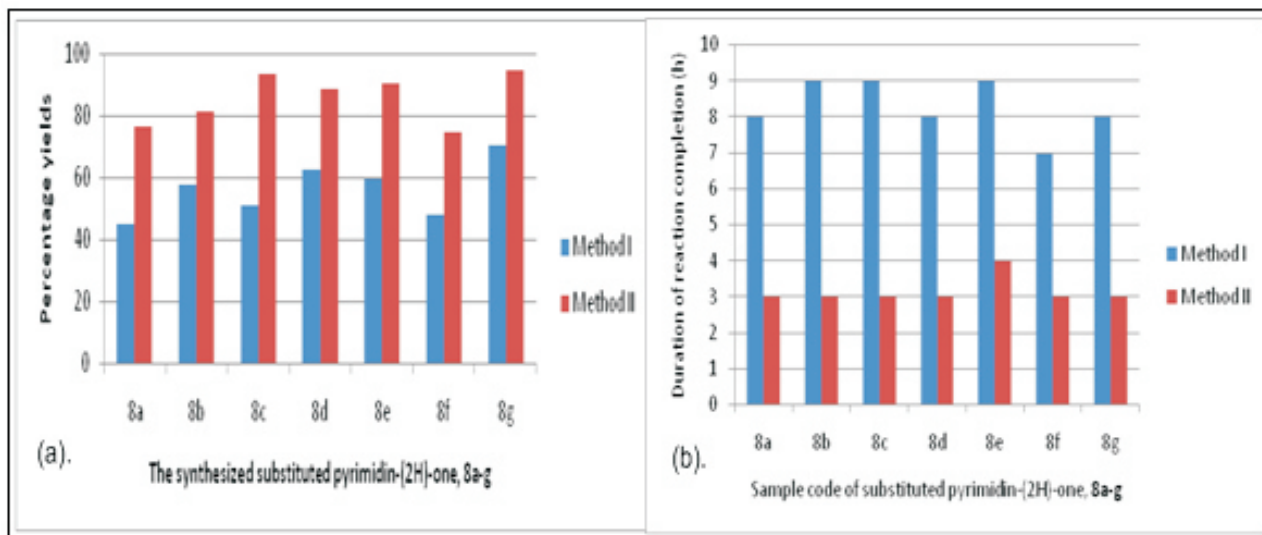


Fig. 2: The comparative study of efficiency of tradition method using conc. HCl (Method I) to that of new approach using SSA (Method II) (a) considering % yields factor. (b). considering reaction time (h) factor.

Table 1: Physico-chemical properties of synthesized pyrimidin-2(1H)-ones (8a-g)

p	Molecular Formula	Mol. Wt.	M.P. (°C)	R _f ^a	Colour	Elemental Analysis %Calcd. (%Found)		
						CH	N	
	C ₁₁ H ₁₂ N ₂ O	188	124-127	0.77	Yellow	70.21(70.73)	6.38(6.09)	14.89(15.01)
	C ₁₈ H ₁₈ N ₂ O	278	211-213	0.56	white	77.70(77.31)	6.47(6.72)	10.07(9.85)
	C ₁₃ H ₁₄ N ₂ O	214	184-186	0.85	Green	72.90(72.71)	5.04(5.52)	13.08(13.53)
	C ₂₀ H ₁₈ N ₂ O	302	227-229	0.49	green	79.47(79.95)	5.98(5.82)	9.27(9.45)
	C ₂₂ H ₂₂ N ₂ O ₃	362	240-242	0.69	green	72.93(73.04)	6.08(5.97)	7.73(7.55)
	C ₁₄ H ₁₆ N ₂ O	228	198-200	0.76	Orange	73.68(73.81)	7.02(7.16)	12.28(12.33)
	C ₂₁ H ₂₀ N ₂ O	316	>320	0.63	black	79.75(79.11)	6.33(6.72)	8.86(8.43)

^aCHCl₃:CH₃OH (9:1, v/v), Mol. Wt. = Molecular Weight, M.P. = Melting Point

CONCLUSION

The pyrimidine derivatives were successfully achieved by synthetic modification of the various chalcone precursors via Silica sulfuric acid (SSA) heterogeneous catalytic approach. SSA was found to be a mild, efficient and reusable solid catalyst for the reaction of α ,-unsaturated carbonyl with urea to furnish the corresponding pyrimidinone derivatives in good to excellent yield. The interesting behaviour of SSA lies in the fact that it can be re-used after simple washing with chloroform thereby rendering this procedure more economical compared with concentrated HCl method.

ACKNOWLEDGEMENT

OOA acknowledges The World Academy of Sciences for the sponsorship of this project under the TWAS Research Grant for Individual Programme (Grant No: 14-069 RG/CHE/AF/AC_1; UNESCO FR: 324028564).

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