Synthesis of Dimethyl Octahydroquinazolinones by using Electrochemically Prepared Copper Oxide Nanoparticles

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Present work involves the synthesis of substituted dimethyl octahydroquinazolinones by using electrochemically prepared copper oxide nanoparticles as catalyst in microwave. Copper oxide nanoparticles were prepared in appreciable yield by using electrochemical reduction method of synthesis in the presence of tetrabutylphosphonium bromide for capping. The prepared nanoparticles were analyzed by UV, FTIR, TGA, X-ray diffraction and electron microscopic studies. The nanoparticles of copper oxide then were successfully used as a catalyst in multicomponent reaction between diketones, substituted aldehydes and urea/thiourea to synthesize substituted dimethyl octahydroquinazolinones.

Keywords: Electrochemical reduction, Copper oxide nanoparticles, Tetrabutylphosphonium bromide, Octahydroquinazolinones.

INTRODUCTION

A rapid research increase in the usefulness of nanomaterial (1 to 100 nm) in the various fields is due to their particle size. When particle size is decreased, it alters the properties of material to larger extent when compared with properties of bulk material with bigger sized particles. The properties which are mainly altered and beneficial for their use in various areas are size of particle, distribution of size and morphological properties. Because of nanosize surface reactivity of nanomaterial is high and that property increases its importance in different fields such as medicine, energy and nutrition [1].

Most commonly used metals for the preparation of nanoparticles are platinum, gold and silver. The nanoparticles prepared from such noble metals are having beneficial effects on the health [2]. Various types of nanomaterial like nanostructures, nanowires, nanorods and nanoparticles are being synthesized from the metals and they find their usefulness in catalysis [3], preparation of polymers [4], diagnosis of diseases and treatment [5], sensing technology [6] and to label optoelectronic recorded media [7]. Though the nanoparticles are very useful in many fields the main challenge is to prepare stable

nanoparticles. Wide varieties of methods have been successfully used to prepare and stabilize the nanoparticles of metal. The chemical, photochemical and electrochemical methods are the most common for the preparation. The properties of nanomaterial are greatly affected by the method used in their preparation. During the preparation various processes takes place like adsorption of agent used for stabilization, interaction between ion of metal and reducing agent used and rate of interaction. These processes largely alter physicochemical and morphological properties of nanoparticles.

Preparation and application of nanoparticles are getting significant scientific and industrial importance because of their unique and improved properties [8,9], which are estimated by size, composition and structure. Various methods have been proposed for the preparation of copper oxide nanoparticles having different shapes and sizes are thermal oxidation [10], sonochemical [11], combustion [12] and quick-precipitation [13,14].

Recently, dimethyl octahydroquinazolinone is attractive nucleus for synthesis because it shows variety of activities such as antimicrobial [15,16], calcium antagonist activity [17] and analgesic and anti-inflammatory agents [18]. In literature, it

has been reported that derivatives of octahydroquinazolinone can be synthesized by use of trimethylsilylchloride [19], Nafion-H_2O [20], vanadium oxide sulphate [21], conc. sulphuric acid [22], conc. hydrochloric acid [16], ionic liquid [23] and silica sulfuric acid [24] as catalyst. Most of these are associated with some disadvantages like more time for reaction, low yield, purity of product, critical conditions for reaction, side reactions leading to impurities and requirement of organic solvents in large amount. Hence method that is environmental friendly and offering high yield is a present need.

The present work provides synthesis and characterization of electrochemically prepared copper oxide nanoparticles using ultraviolet (UV), fourier transform infrared (FTIR), thermogravimetry (TGA), X-ray diffraction (XRD), scanning electron microscopy-energy dispersive spectroscopy (SEM-SDS) and transmission electron microscopy selected area electron diffraction (TEM-SAED) studies along with their application as catalyst in the synthesis of various substituted dimethyl octahydroquinazolinones using microwave.

EXPERIMENTAL

Electrochemical synthesis: For the preparation of nanoparticles of copper oxide, tetrabutylphosphonium bromide was (TBPB) dissolved in water to get 0.01 N solution. The said solution was placed in a 50 mL electrolysis cell vessel. A 1 cm × 1 cm sheet of copper was used as a one of the electrode. Another 1 cm × 1 cm sheet of platinum acts as an inert electrode. The two electrode sheets were dipped in to vessel containing 0.01 N solution of TBPB. In this synthesis process, TBPB acts as a supporting electrolyte and stabilizing agent which does not allow the formed nanoparticles to grow further. In this electrochemical process for synthesis of nanoparticles oxidation of copper metal takes place and converted to copper ions. The copper ions thus formed move towards the cathode and forms oxides of copper in nanosize in the solution of electrolyte as well as at the region of interface between surface of cathode and solution of electrolyte. The process of electrolysis was carried out at different current densities. The current densities applied in mA/cm^2 were 5, 10, 15 and 20 and the duration of electrolysis was 2 h. During this process of electrolysis, the colour of solution changes. Firstly colourless solution turns to light blue followed by dirty green and finally precipitate of dark brown colour was formed. The process of electrolysis was terminated after the period of 2 h. The solution contained brown coloured precipitate was then collected in a bottle. Allowed to settle the solid particles. The solid thus obtained was separated by decantation. The separated solid washed with water for 3 to 4 times to ensure complete removal of excess capping agent.

Analysis of copper oxide nanoparticles: Copper oxide nanoparticles were analyzed with the help of UV-visbile spectrophotometer (UV 1800 Shimadzu), fourier transform infrared spectrometer (FTIR Affinity 1 Shimadzu), thermogravimetric analysis (TGA 50 Thermoanalyzer Shimadzu), high end X-ray diffractometer, scanning electron microscopy-energy dispersive spectroscopy (SEM-EDS) and transmission electron microscopy selected area electron diffraction (TEM-SAED) techniques.

Synthesis of substituted dimethyl octahydroquinazolinones: Urea/thiourea (1.5 mmol), dimedone (1 mmol), substituted aromatic aldehydes (1 mmol) and copper oxide nanoparticles (40 mg) were mixed together. The mixture was then irradiated with microwave radiations at 300 W using CEM discovery microwave synthesis system. The completion of the reaction was monitored by TLC. The mobile phase used was ethyl acetate and benzene. The total period of microwave irradiation varies from reaction to reaction. After completion of reaction the solid reaction mixture was transferred to beaker containing ethyl acetate. The organic compound gets dissolved in ethyl acetate and solid catalyst was filtered off. The ethyl acetate solution was washed thoroughly with water to remove inorganic material and dried by use of anhydrous sodium sulphate. Ethyl acetate was allowed to evaporate and solid residue obtained. The solid residue containing title compound was purified by recrystallization using methanol.

RESULTS AND DISCUSSION

Electrochemical synthesis: Copper oxide nanoparticles were prepared in appreciable yields by electrochemical reduction technique using tetrabutylphosphonium bromide as capping agent. The capping agents are organic surface active agents and used to bind selectively on the specific plane of crystal of nanomaterial. Tetrabutylphosphonium bromide is a surfactant, which play important role and prevent rapid aggregation and flocculation of the particles. Surface active agents due to capping of surface of nanoparticles control growth rate thus affect morphological properties of nanoparticles and orient it in crystal formation. The rate of attachment and diffusion of surface active agent on the surface control the growth rate of nanoparticles. After 15 min of electrolysis growth in the size of nanoparticles was accessed by UV-visible spectral studies.

UV-visible studies: Copper oxide nanoparticles were subjected for UV-visible spectroscopic analysis by scanning in double beam spectrophotometer in the range of 200-1100 nm. From the scanning, it was observed that wavelength maxima occurred in the UV region for all the samples. Figs. 1 and 2 show the UV-visible spectra of copper oxide nanoparticles prepared in water with the help of tetrabutylphosphonium bromide as a capping agent recorded at current densities of 5 and 10 mA/cm^2, respectively. The said analysis revealed that at current density of 10 mA/ cm^2 wavelength maxima is lower than that of 5 mA/ cm^2. It confirmed that with increase in current density particle size decreases and shows blue shift. Absorbance of sample was recorded after 15 min of electrolysis and was found higher than that after 2 h of electrolysis. From these observations, it can be confirmed that particle size at lower time of electrolysis is smaller than that of higher. With increase in electrolysis time particle size was found increasing.

FT-IR studies: The electrochemically prepared copper oxide nanoparticles by use of tetrabutylphosphonium bromide for capping were analyxed for their capping efficiency of ligand by means of FTIR spectroscopy. The FTIR spectra of as prepared copper oxide nanoparticles using tetrabutylphosphonium bromide (Fig. 3) shows peaks at 2970, 2864, 2832, 3462 and 3111 cm^{-1}
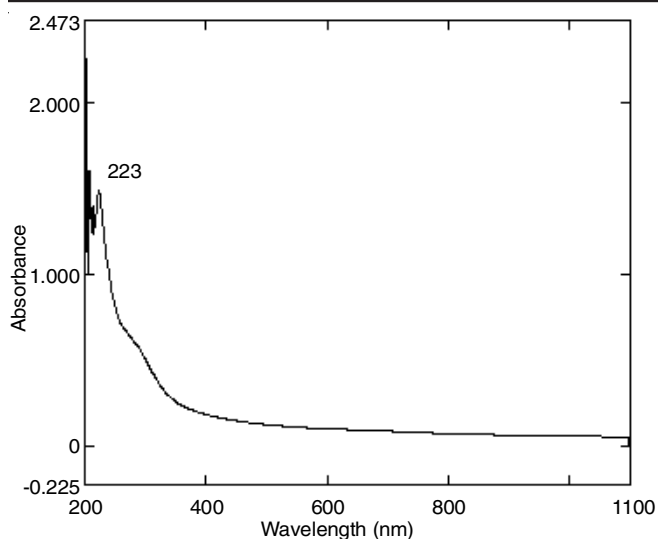
Fig. 1.   UV-visible spectrum of copper oxide nanoparticles at 5 mA/cm² capped with TBPB in water
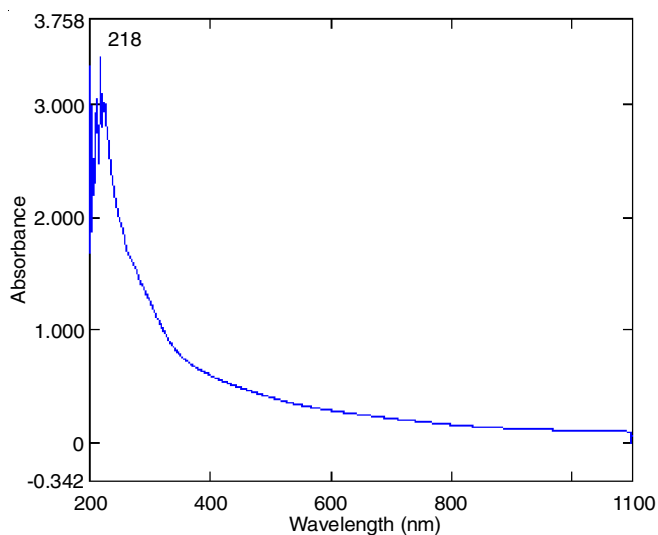

Fig. 2.   UV-visible spectrum of copper oxide nanoparticles at 10 mA/cm² capped with TBPB in water
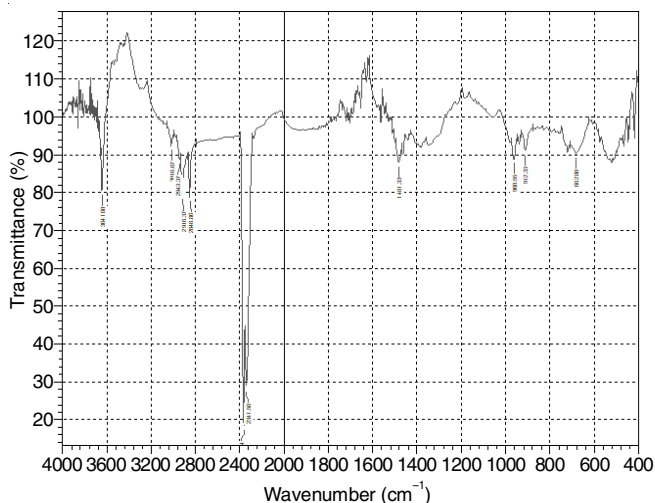

Fig. 3. FTIR spectrum of as prepared copper oxide nanoparticles

indicating C-H stretching vibrations, 2440, 2331 cm⁻¹ P-C stretching vibrations and 1450-1280 cm⁻¹ P-C and C-H bending vibrations, respectively. It revealed the presence of tetrabutyl-phosphonium bromide at nanoparticles surface. The peaks observed at 3640 and 3483 cm⁻¹ indicate the presence of water due to hygroscopic nature of the ligand. The FTIR spectrum of calcinated copper oxide nanoparticles (Fig. 4) at 500 ℃ in muffle furnace shows peaks corresponding to ligand are disappeared or became weak indicating removal of ligand in calcination process. The broad band at 661 cm⁻¹ indicates presence of Cu-O bond.
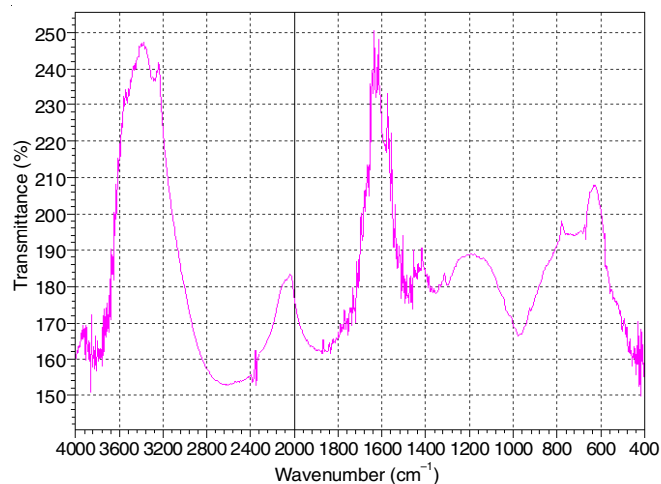
Fig. 4. FTIR spectrum of calcinated copper oxide nanoparticles

Thermal studies: In case of TGA curve of copper oxide nanoparticles (Fig. 5) above 200 ℃ weight of material was constant as it indicates that over 200 ℃ tetrabutylphosphonium bromide which was used as capping agent while preparation of nanoparticles of copper oxide is not present with it.
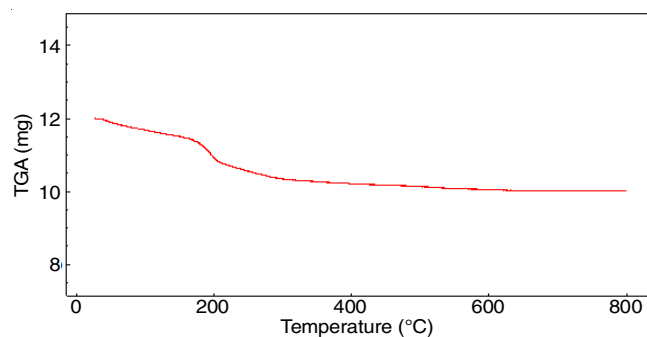
Fig. 5. TGA curve of as prepared copper oxide nanoparticles

XRD studies: The XRD pattern of synthesized copper oxide nanoparticles were obtained after calcination at 500 ℃ for the period of 2 h (Fig. 6). The diffraction peaks observed reveals the monoclinic system of copper oxide nanoparticles. The ten characteristic peaks having lattice parameter a = 4.679, b = 3.431, c = 5.136 at β = 99.262 with 2θ = 32.889, 35.990, 39.177, 49.182, 54.113, 58.795, 62.094, 68.627, 72.968 and 75.731 corresponding to Miller indices of ($\bar{1}$10), (002), (200), ($\bar{1}$12), ($\bar{2}$02), (020), (202), ($\bar{1}$13), (221), ($\bar{2}$22) were observed
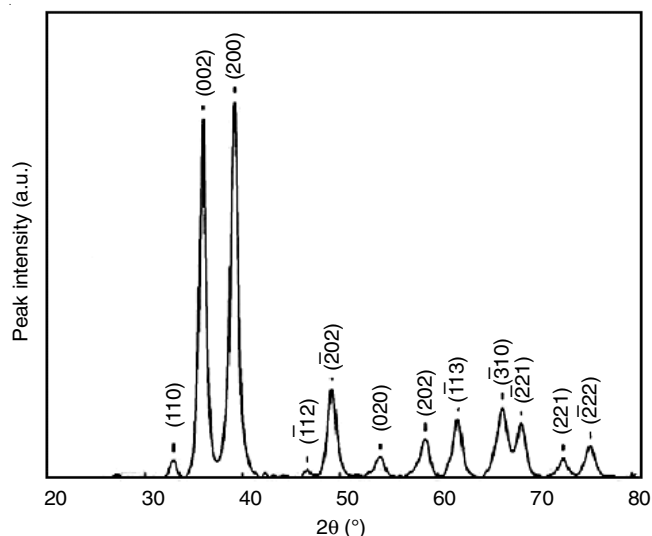
Fig. 6. XRD pattern of copper oxide nanoparticles

which exactly coincide with the JCPDS no. 80-0076 of copper oxide. The average size of particles was calculated with the help of Debye-Scherrer's equation which states that $D = k\lambda/\beta\cos\theta$. In this equation k stands for shape constant, λ is wavelength, θ for angle of diffraction whereas β indicates full width half maxima. The average size of copper oxide nanoparticles calculated on the basis of XRD data was 8.17 nm.

SEM-EDS studies: In the present work, copper oxide nanoparticles were analyzed by scanning electron microscopy at various resolutions. The SEM image (Fig. 7) shows morphological features of copper oxide nanoparticles. The SEM image indicates presence of uniform, regular and non-spherical particles. It also revealed the formation of crystalline particles in nano size along with aggregates oriented randomly. It is being observed that calcination results in increased crystal size of particles. The copper oxide nanoparticles were analyzed qualitatively and quantitatively by EDS as shown in Figs. 8 and 9. The spectrum of EDS revealed that copper and oxygen present in composition are shown in Table-1.
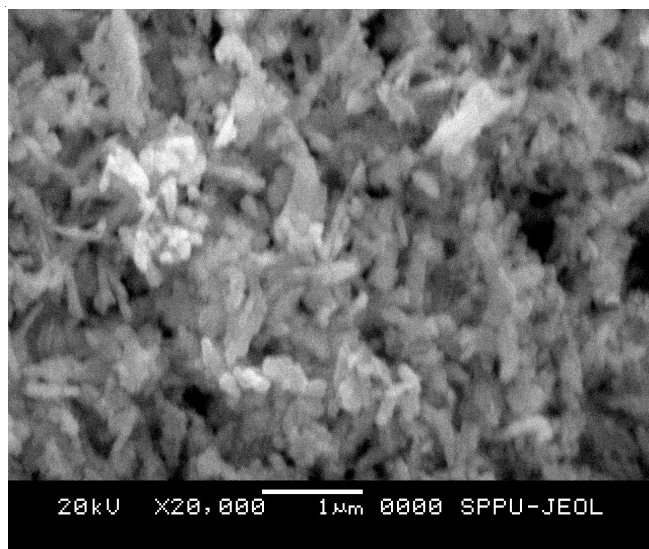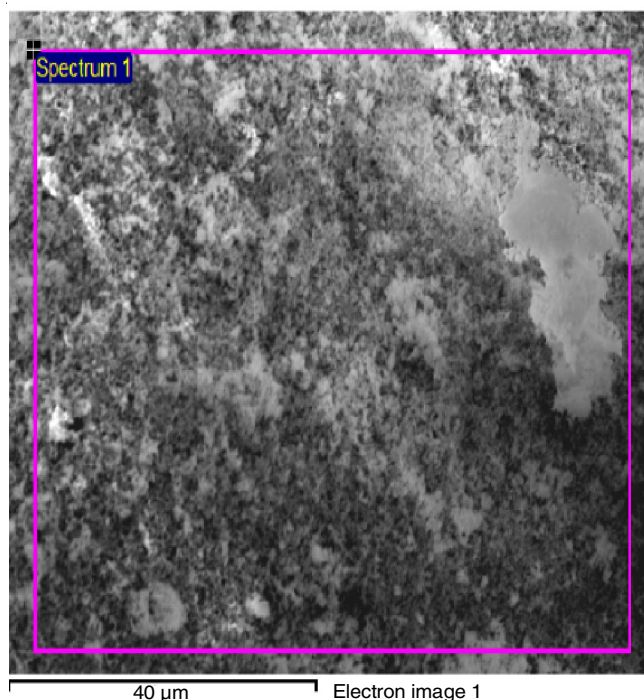
Fig. 8. Electron image of copper oxide nanoparticles
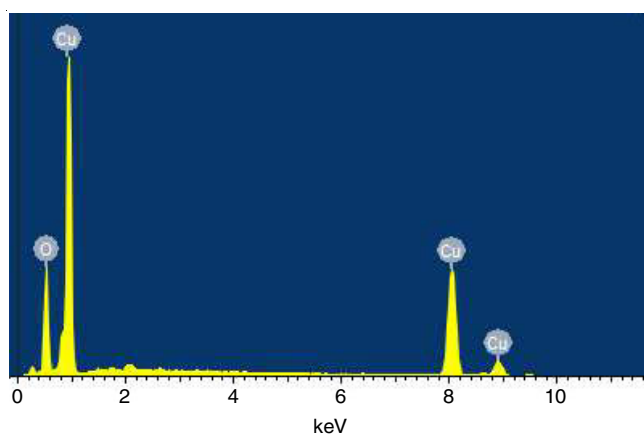

Fig. 9. EDS spectrum of copper oxide nanoparticles

TABLE-1
EDS DATA OF COPPER OXIDE NANOPARTICLES

Element	Weight (%)	Atomic (%)
O K	44.80	75.38
Cu K	58.10	24.69
Total	102.89	

TEM-SAED analysis: The different shades are the result of different densities shown by TEM images (Fig. 10). The TEM image shows the particle size of 19.53, 10.74, 9.75, 9.82, 9.92, 13.79 and 11.38 nm with average of 12.13 nm having maximum particles with size range from 9-10 nm which is in well agreement with XRD analysis. The random distribution of metal nanoparticle aggregates was clearly observed in the image. The electron diffraction pattern of selected area of nanoparticle that is reflected in Fig. 11 verifies the crystalline nature due to absence of diffused holes normally associated with the presence of amorphous phases.
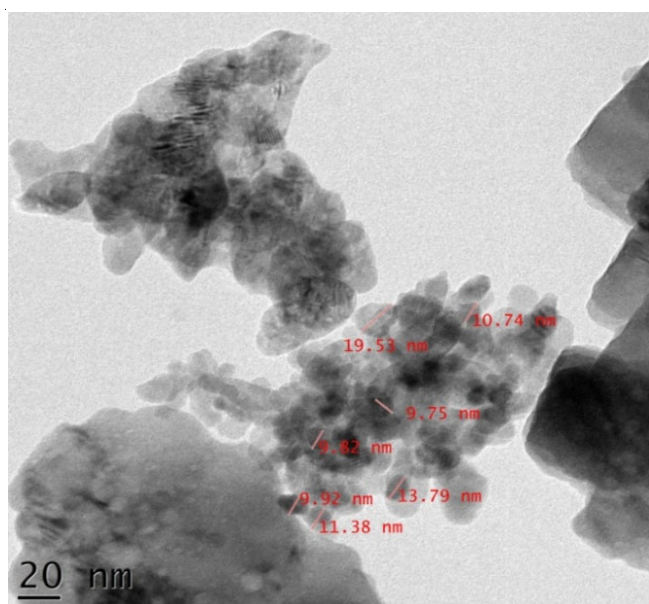

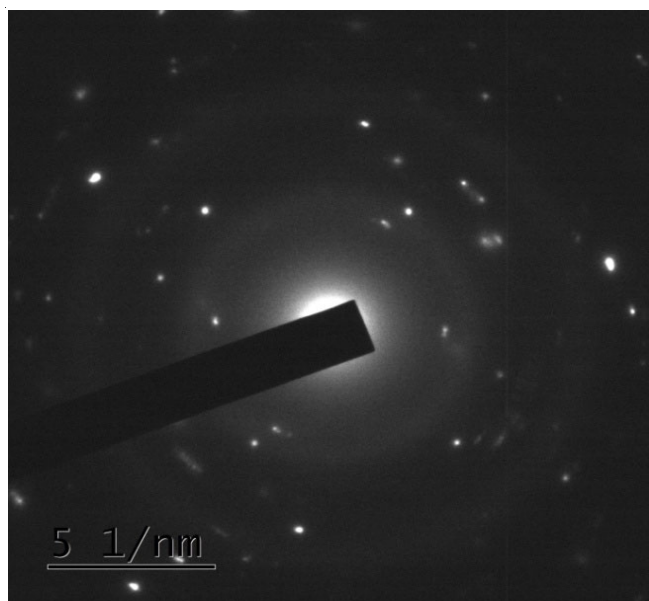
Fig. 7. SEM image of calcinated copper oxide nanoparticles

Fig. 10. TEM image of calcinated copper oxide nanoparticles
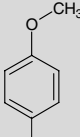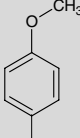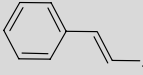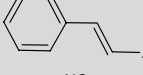

Fig. 11. SAED pattern of calcinated copper oxide nanoparticles
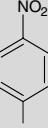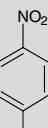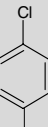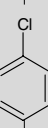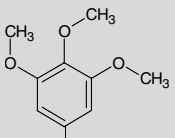
Synthesis of substituted dimethyl octahydroquinazo-linones: Various substituted dimethyloctahydroquinazolinones have been synthesized as per the reaction are shown in **Scheme -I**. The thin layer chromatography was used to monitor progress of reaction. The melting point of the title compounds was uncorrected. The open capillary method was employed to record the melting point. The found melting points were compared with reported data [25] as shown in Table-2. The structure of the title compounds was confirmed by IR and ^{1}H NMR studies. The presence of stretching bands for C=O, N-H and C-H as well as C=S in particular compounds have been observed (Table-3). The proton NMR spectra of compounds **1**, **3**, **11**, **17** and **21**, which are not being found reported in the said literature and confirmed their structure (Table-4).
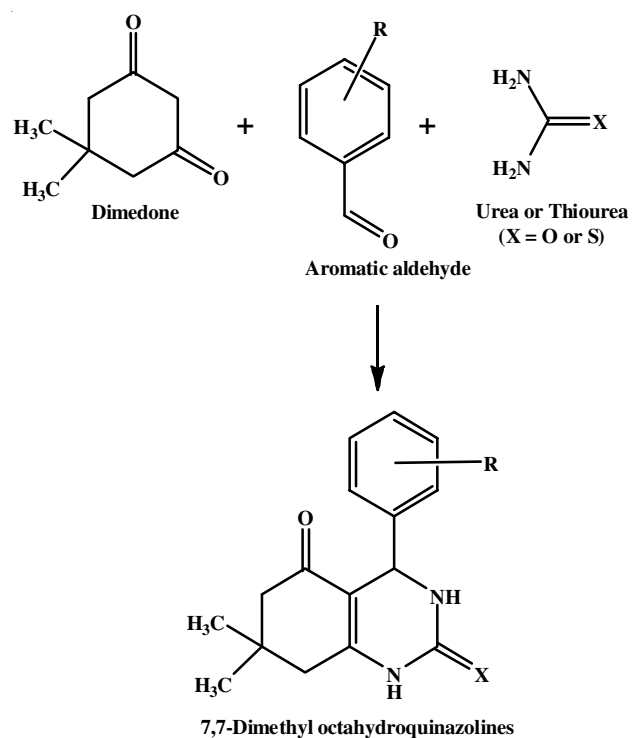
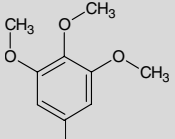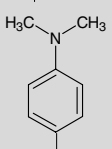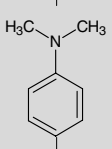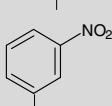**Scheme-I:** Synthesis of various substituted dimethyl octahydroquinazo-linones using copper oxide nanoparticles

TABLE-2
PHYSICO-CHEMICAL DATA OF SYNTHESIZED SUBSTITUTED DIMETHYL OCTAHYDROQUINAZOLINONES (1-22)

Compound	R	X	m.f.	m.p. (°C) Found	m.p. (°C) Reported	Yield (%)	R_f value
1	(phenyl)	S	$C_{16}H_{18}N_2OS$	284-286	286-288	92	0.6
2	(phenyl)	O	$C_{16}H_{18}N_2O_2$	291-293	292-295	95	0.72
3	(2-hydroxyphenyl)	S	$C_{16}H_{18}N_2O_2S$	194-195	New	87	0.45
4	(2-hydroxyphenyl)	O	$C_{16}H_{18}N_2O_3$	188-189	New	89	0.63

No.	Structure	X	Formula	m.p.	lit.	Yield	R_f
5		S	$C_{17}H_{20}N_2O_2S$	276-279	278-280	91	0.65
6		O	$C_{17}H_{20}N_2O_3$	273-276	274-276	93	0.52
7		S	$C_{18}H_{20}N_2OS$	171-174	172-174	84	0.58
8		O	$C_{18}H_{20}N_2O_2$	151-154	150-152	89	0.59
9		S	$C_{16}H_{17}N_3O_3S$	286-289	288-290	88	0.45
10		O	$C_{16}H_{17}N_3O_4$	> 300	302-304	94	0.66
11	H	S	$C_{10}H_{14}N_2OS$	196-198	New	95	0.70
12	H	O	$C_{10}H_{14}N_2O_2$	188-190	New	95	0.53
13		S	$C_{16}H_{17}N_2OSCl$	285-287	288-290	68	0.5
14		O	$C_{16}H_{17}N_2O_2Cl$	> 300	318-320	78	0.60
15		S	$C_{19}H_{24}N_2O_4S$	133-136	134-136	90	0.63
16		O	$C_{19}H_{24}N_2O_5$	138-141	140-142	84	0.49
17		S	$C_{18}H_{23}N_3OS$	212-214	New	93	0.64
18		O	$C_{18}H_{23}N_3O_2$	228-230	New	93	0.51
19		S	$C_{16}H_{17}N_3O_3S$	280-283	281-283	95	0.60
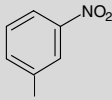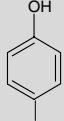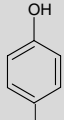
20		O	$C_{16}H_{17}N_3O_4$	296-298	297-299	98	0.60
21		S	$C_{16}H_{18}N_2O_2S$	202-204	New	89	0.72
22		O	$C_{16}H_{18}N_2O_3$	230-231	New	88	0.53

TABLE-3
INFRARED SPECTRAL DATA OF SYNTHESIZED SUBSTITUTED DIMETHYL OCTAHYDROQUINAZOLINONES (1-22)

Compound	IR value (cm^{-1})
1	3433.64, 3357.46 (N-H), 2965.02 (C-H), 1712.48, (C=O), 1556.27 (C=S).
2	3489.56, 3426.89 (N-H), 2965.02 (C-H), 1615.12, 1596 (C=O).
3	3451.96, 3169.44 (N-H), 2954.41 (C-H), 1636.3 (C=O), 1587.13 (C=S), 1375 (C-OH).
4	3460.63, 3162.69 (N-H), 2948.63 (C-H), 1708.62, 1640.16 (C=O), 1305.57 (C-OH)
5	3315.03, 3057.58 (N-H), 2958.27 (C-H), 1659.45, 1601.59 (C=O), 1504.2 (C=S)
6	3447.13, 3309.25 (N-H), 2954.41 (C-H), 1667.16, 1598.7 (C=O).
7	3447.13, 3286.11 (N-H), 2951.52 (C-H), 1648.84 (C=O), 1550.49 (C=S).
8	3433.64, 3387.35 (N-H), 2965.02 (C-H), 1646.91, 1548.56 (C=O).
9	3301.54, 3196.43 (N-H), 2958.27 (C-H), 1655.59 (C=O), 1509.99 (C=S).
10	3196.43 (N-H), 2965.02 (C-H), 1666.2 (C=O).
11	3416.96, 3395.12 (N-H), 2956.34 (C-H), 1612.2 (C=O), 1576.2 (C=S)
12	3453.88, 3308.29 (N-H), 2962.13 (C-H), 16.59.45, 1620.68 (C=O).
13	3315.03 (N-H), 2955.38 (C-H), 1655.59 (C=O), 1583.27 (C=S).
14	3347.82 (N-H), 2951.52 (C-H), 1701.87, 1587.13 (C=O).
15	3460.63, 3007.44 (N-H), 2953.45 (C-H), 1588.09 (C=O), 1510.95 (C=S)
16	3460.63, 3421.1 (N-H), 2944.77 (C-H), 1587.13, 1507.1 (C=O)
17	3426.92, 3374.82 (N-H), 2970.8 (C-H), 1715.37, 1665.59 (C=O), 1509.99 (C=S)
18	3434.6, 3316.96 (N-H), 2971.17 (C-H), 1718.33, 1652.7 (C=O)
19	3447.13, 3309.15 (N-H), 2954.41 (C-H), 1667.16, 1598.7 (C=O).
20	3433.64, 3315.03 (N-H), 2962.13 (C-H), 1648.84, 1593.88 (C=O)
21	3288.04 (N-H), 2956.34 (C-H), 1662.34, 1614.13 (C=0), 1508.06 (C=S)
22	3352.64 (N-H), 2964.05 (C-H), 1665.23, 1609.31 (C=0)

TABLE-4
NMR SPECTRAL DATA OF COMPOUND 1, 3, 11, 17 AND 21

Compound	Chemical shift (δ ppm)
1	1.1 (s, 6H, Me), 2.2-2.9 (M, 4H, CH_2), 4.7 (s, 1H, CH), 7 - 7.4 (m, 5H, Ar), 9.4 (s, 2H, NH).
3	1.1 (s, 6H, Me), 2.1-2.6 (M, 4H, CH_2), 4.8 (s, 1H, CH), 5.4 (s, 1H, OH), 7-7.4 (m, 4H, Ar), 9.5 (s, 2H, NH).
11	1.1 (s, 6H, Me), 2.1-2.5 (M, 4H, CH_2), 4.2 (s, 2H, CH_2), 8.1 (s, 2H, NH).
17	1.2 (s, 6H, Me), 2.1-2.9 (M, 4H, CH_2), 3.1 (s, 6H, NCH_3), 4.8 (s, 1H, CH), 7-7.7 (m, 4H, Ar), 8.7 (s, 2H, NH).
21	1.2 (s, 6H, Me), 2.1-2.6 (M, 4H, CH_2), 4.6 (s, 1H, CH), 4.8 (s, 1H, OH), 7 - 7.8 (m, 4H, Ar), 8.9 (s, 2H, NH).

Conclusion

The electrochemical method was sucessfuly employed to prepare copper oxide nanoparticles using tetrabutylphosphonium bromide as capping agent with appriciable yields. The as prepared copper oxide nanoparticles heated at 500 ºC to remove capping agent and removal is confirmed by FTIR studies. The characterization techiques confirmed the size, size distribution along with shape of the nanoparticles. The calcinated copper oxide nanoparticles were used sucessfully as a catalyst in multicomponent reaction between diketones, substituted aldehydes and urea/thiourea to synthesize dimethyl octahydroquinazolinones.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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