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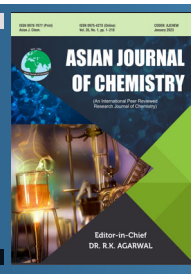

Structural Analysis of Calcium Oxide Derived from Waste Egg's Shells and their Application for Knoevenagel Condensation Reactions

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The present study demonstrates the simple and easy synthesis of calcium oxide (CaO) from waste egg's shells. Fine egg's shells powder were calcinated at 500, 700 and 800 ºC. The structural and morphological evaluation of calcium oxide derived from waste egg's shell were examined by using XRD, FT-IR, SEM, EDS and TEM techniques. The sintered calcium oxide was used as heterogeneous catalyst for the synthesis of 5-arylidene malononitrile and 5-arylidene barbituric acid derivatives by the condensation of various aromatic aldehyde and active methylene compounds *via* Knoevenagel condensations reaction under ultrasound irradiation method. The reaction completed within a very short reaction time with high yields of product. The present method describes easy synthesis of calcium oxide catalyst from the waste egg's shells, short reaction times, simple product isolation, excellent yield and reusability of the catalyst.

Keywords: Eggs shells, Calcium oxide, Heterogeneous catalyst, Ultrasound irradiation, Knoevenagel condensations.

INTRODUCTION

Eggs are one of the important cuisine ingredients used in daily life of human beings owing to its nutritional values. Eggs are very rich in containing proteins, amino acids, saturated and unsaturated fatty acids, vitamins and minerals [1-4]. Therefore, eggs have been recognized as a "essential and reference food" protein for humans. The use of egg and its derivative resulted in unwarranted quantity of waste residual shells or waste eggs shells, which are discarded into the environment and pose the environmental pollution. Calcium carbonate is one of the main ingredients of an egg shell that can be used in numerous applications including; pet food, a filler in animal feed, printing ink, glaze decoration, paper, tiles and also as a source of calcite or calcium oxide. A waste eggshell also contains magnesium, limestone, proteins, *etc*. [5-7].

Calcium carbonate contain in the discarded waste eggs shell easily transformed into calcium oxide by calcination at suitable temperature. The obtained calcium oxide from waste eggs shell after heat treatment or on calcination would be possibly used in many applications such as printing ink, fertilizer, cosmetics, pharmaceuticals, as starting materials for synthesis of calcium sulphate (gypsum), $CaAl_2O_4$, $CaTiO_3$, $CaSiO_3$, catalysis, adsorption [8-14]. Calcium oxide is used as a solid base catalyst for biodiesel production [15,16]. In many organic transformation such as aldol condensation [17], saponification [18], transesterification of sunflower oil [19], transesterification of palm oil [20], calcium oxide is used as a solid base heterogeneous catalysts. In addition to that calcium oxide is used in the removal of toxic metal such as chromium [21].

Knoevenagel condensation is a common type of organic reaction used for the synthesis of α,β-unsaturated carbonyl compounds by condensation of various aromatic aldehydes with active methylene compounds such as malononitrile, barbituric acid, Meldrum acids, *etc*. The product synthesized using Knoevenagel condensation reactions are extensively used as a starting materials in multistep organic syntheses, perfumes and polymers, pharmaceuticals [22,23], antibacterial, pesticides and antifungal products [24,25]. In addition to that, some of the compounds synthesized using Knoevenagel condensation reactions also shows the medicinal activities like kinase inhibitor [26], antiviral [27], antitubercular [28], anticancer [29] and antidibetic [30]. The Knoevenagel condensation

reaction is also carried out using various reported methods and catalysts such as Baker's yeast [31], CuO nanoparticle [32], carbon nanotube [33], ([TPPHSP]Br) ionic liquid [34], fly ash supported calcium oxide [35], anion-exchange resin [36], calcium hydroxide [37] Fe_3O_4@SiO_2-3N [38], Mn-MOF [39], PEG-400 [40], 2,2′-bipyridinium dihydrogen phosphate [41].

In recent years, ultrasound assisted organic synthesis has been widely used as a green alternative method to the existing conventional method [42,43]. The organic reactions carried out using ultrasonic irradiation method offer several advantages such as shorter reaction time, milder conditions, minimum toxic generation, high yields and good product purity [44-46]. In present work, we have successfully synthesized calcium oxide as a solid heterogeneous catalyst from waste egg's shell. The structural and morphological evaluation of synthesized calcium oxide was examined using XRD, FT-IR, SEM, EDS and TEM techniques. After surface characterization, the applicability of calcium oxide catalyst was examined in the synthesis of 5-arylidene malononitrile and barbiutric acid derivatives *via* Knoevenagel condensation reaction between various aromatic aldehydes and active methylene compounds under ultrasonic irradiation.

EXPERIMENTAL

The AR grade reagents were used throughout the work without further purification. The elemental composition investigated by energy dispersive X-Ray analysis (EDAX, Inca Oxford, attached to SEM). The crystallographic structures were identified by X-ray powder diffraction with Cu*K*α radiation (λ = 1.5405 Å) by Phillips-3710 X-ray diffractometer. Morphology and structure evaluated on JEOL-JSM-5600 N scanning electron microscope and on Philips-CM-200 transmission electron microscope. The IR spectra recorded with Perkin-Elmer infrared spectrophotometer. The melting points of synthesized compounds were recorded in open capillaries and are uncorrected. ^{1}H NMR spectra of the compounds were recorded using Varian Gemini spectrometer (500 MHz). Trimethyl silane was used as an internal standard for comparison of chemical shifts, which were reported in δ ppm.

Synthesis of calcium oxide (CaO) from waste eggs shells: The waste eggs shells were collected and washed with distilled water to eliminate the foreign impurities and then dried in an electric oven at 80 ºC for 24 h. The dried egg shells were ground to a fine powder using mortar and pestle. The fine powder so obtained was then sintered in a muffle furnace at different temperature *viz*. 500 ºC (M3), 700 ºC (M2) and 800 ºC (M1) for 2 h to eliminate any form of carbon and to achieve complete conversion of $CaCO_3$ to CaO. Finally, after calcination at 800 ºC, the white coloured fine powder of calcium oxide was obtained as shown in Fig. 1.

General procedure

Synthesis of 5-arylidene malononitrile derivatives (3a-h): In a typical reaction procedure an aromatic aldehydes (**1**) (5 mmol) and malononitrile (**2**) (5 mmol) were taken in 100 mL beaker and 10 mL ethanol and 200 mg of catalyst (CaO) was added in the mixture and then reaction mixture was exposed to ultrasound irradiation for 15-35 s. The reaction progress was examined using TLC [*n*-hexane + ethyl acetate (7:3)]. After completion of reaction, the product was isolated (**Scheme-I**). Initially, the solid product was filtered and recrystallized using hot water the pure solid product was obtained.

2-(Benzylidene)malononitrile (3a): m.p.: 81 ºC; IR (KBr, ν_{max}, cm^{-1}): 2221, 2920, 1586, 1487, 956, 673. ^{1}H NMR (500 MHz, DMSO-d_6) δ ppm: 7.24 (t, 1 H), 7.62 (d, 2 H), 7.78 (d, 2H), 8.02 (s, 1H).

2-(4-Chlorobenzylidene)malononitrile (3c): m.p. 160 ºC; IR (KBr, ν_{max}, cm^{-1}): 2220, 1574, 1482, 1361, 850; ^{1}H NMR

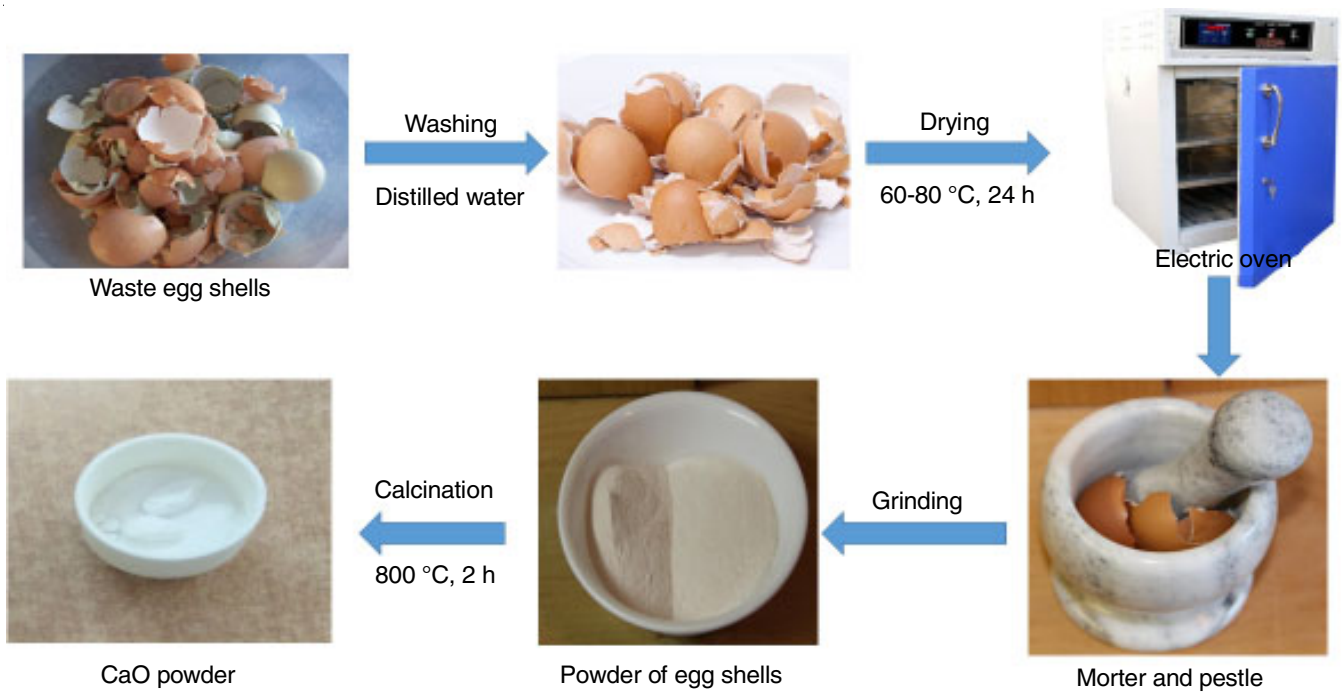
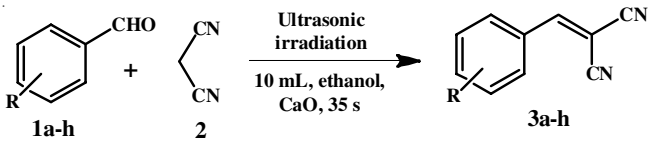


Fig. 1. Schematic diagram for synthesis of calcium oxide from waste egg's shells



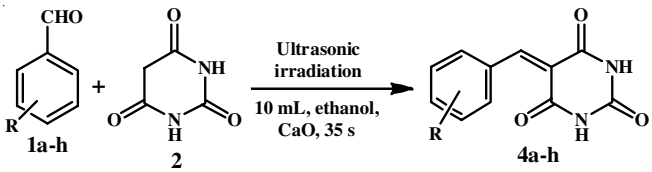
Scheme-I: Synthesis of 5-arylidene malononitrile derivatives catalyzed by calcium oxide

(500 MHz, DMSO-d_6) δ ppm: 7.45 (d, 2H), 7.78 (d, 2H), 7.86 (s, 1H).

2-(2-nitrobenzylidene)malononitrile (3e): m.p.138 ºC; IR (KBr, ν_{max}, cm^{-1}): 2240, 3051, 1530, 1332, 856. ^{1}H NMR (500 MHz, DMSO-d_6) δ ppm: 8.95 (s, 1H), 8.33 (d, 1H), 8.01 (t, 1H), 7.94 (d, 2H), 7.90 (t, 1H)

2-(4-(Dimethylamino)benzylidene)malanonitrile (3g): m.p. 180 ºC; IR (KBr, ν_{max}, cm^{-1}): 3283, 3157, 2136, 1997, 1417, 1164, 975, 748. ^{1}H NMR (500 MHz, DMSO-d_6) δ ppm: 3.1 (s, 6H, NCH_3), 6.85 (dd, 2H, Ar-H), 7.85 (dd, 2H, Ar-H), 8.9 (s, 1H).

Synthesis of 5-arylidene barbituric acid derivatives (4a-h): In a typical reaction procedure, an aromatic aldehydes (**1**) (5 mmol) and barbituric acid (**2**) (5 mmol) were taken in 100 mL beaker and 10 mL ethanol and 200 mg of catalyst (CaO) was added in the mixture and then reaction mixture was exposed to ultrasound irradiation in ultrasonicator bath for 1-2 min. The reaction progress was evaluated using TLC [ethyl acetate + *n*-hexane (3:7)]. After completion of reaction, the product was isolated (**Scheme-II**). Initially, the solid product was filtered and recrystallized using hot water, pure solid product was obtained.



Scheme-II: Synthesis of 5-arylidene barbituric acid derivatives catalyzed by calcium oxide

5-(4-Methoxybenzylidene)pyrimidine-2,4,6-(1*H*,3*H*,5*H*)trione (4b): m.p. 298 ºC; IR (KBr, ν_{max}, cm^{-1}): 3214, 2822, 1735, 1671, 1601, 1552, 1511, 1442, 1307, 1214. ^{1}H NMR (500 MHz, DMSO-d_6) δ ppm: 3.91 (s, 3H), 6.95 (d, 2H), 8.37 (s, 1H), 8.39 (d, 2H).

5-(4-Chlorobenzylidene)pyrimidine-2,4,6-(1*H*,3*H*,5*H*)-trione (4c): m.p.: 300 ºC; IR (KBr, ν_{max}, cm^{-1}): 3415, 3211, 2965, 1746, 1700, 1563. ^{1}H NMR (500 MHz, DMSO-d_6) δ ppm: 7.54 (d, 2H), 8.08 (d, 2H), 8.25 (s, 1H), 11.27 (s, 1H), 11.41 (s, 1H).

5-(4-Hydroxybenzylidene)pyrimidine-2,4,6-(1*H*,3*H*,5*H*)trione (4e): m.p.: 302 ºC; IR (KBr, ν_{max}, cm^{-1}): 3412, 3215, 2967, 1730, 1708, 1532. ^{1}H NMR (500 MHz, DMSO-d_6) δ ppm: 6.88 (d, 2H), 8.21 (s, 1H), 8.33 (d, 2H), 10.82 (s, 1H), 11.12 (s, 1H), 11.25 (s, 1H).

5-(4-Nitrobenzylidene)pyrimidine-2,4,6-(1*H*,3*H*,5*H*)-trione (4h): m.p.: 268 ºC; IR (KBr, ν_{max}, cm^{-1}): 3322, 3240, 3094, 1741, 1693, 1594, 1516. ^{1}H NMR (500 MHz, DMSO-d_6) δ ppm: 11.4 (s, 1H), 11.30 (s, 1H), 8.31 (s, 1H), 8.23 (d, 2H) 8.01 (d, 2H).

RESULTS AND DISCUSSION

XRD studies: The waste eggs shell powder calcinated at different temperatures were analyzed using X-ray diffraction technique and its XRD spectrum are shown in Fig. 2. It was observed that eggs shells powder calcinated at 500 and 700 ºC reveals the calcium carbonate as the major phase and calcium oxide as minor phase. The results showed that egg shells powder calcinated at 800 ºC (M1) resulted in the formation of nano sized calcium oxide (CaO) showed a single cubic phase with lattice constant a = 4.4500 Å. The average crystallite size of egg shell powders were estimated from the most intense peak of XRD using the Debye-Scherrer method. The observed average crystallite size of egg shell powders calcinated at 500 ºC (M1), 700 ºC (M2) and 800 ºC (M1) were 49.454, 42.494 and 12.549 nm, respectively. It is concluded that the egg's shells powder calcinated at < 800 ºC is the characteristic of $CaCO_3$ and contained $CaCO_3$ as the major phase and CaO as a minor phase. Powdered egg shells that have been calcined at 800 ºC exhibit diffraction patterns typical of CaO.

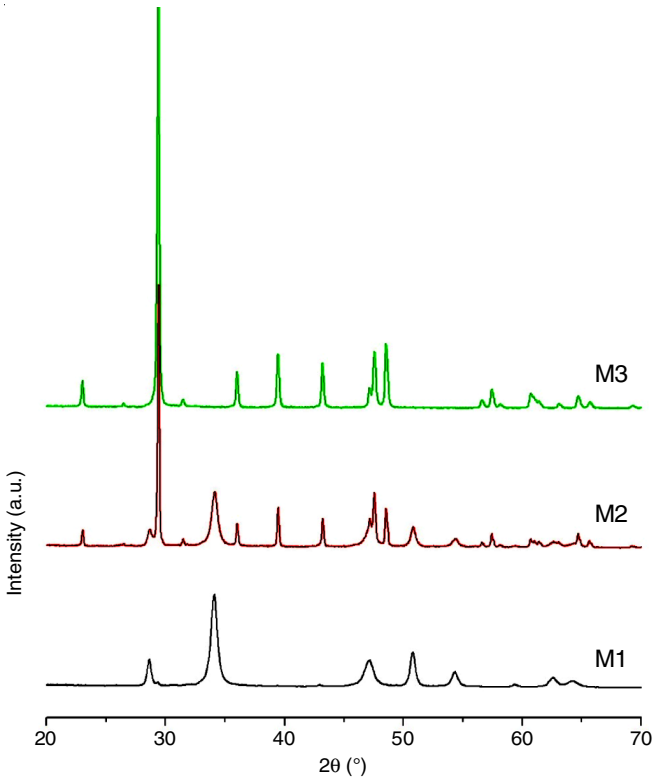


Fig. 2. XRD pattern of waste eggs shell powder calcinated at 800 ºC (M1), 700 ºC (M2) and 500 ºC (M3)

FT-IR studies: The FT-IR spectra of the egg shell powder calcinated at different temperatures are shown in Fig. 3. From these IR spectra of the waste eggs shell powder calcinated at 500 ºC (M3), 700 ºC (M2) and 800 ºC (M1) in each case a strong peak were observed in the 3700-3500 cm^{-1} range is attributed due to the presence of hydroxyl group that was adsorbed onto the surface of waste eggs shell calcinated samples and this

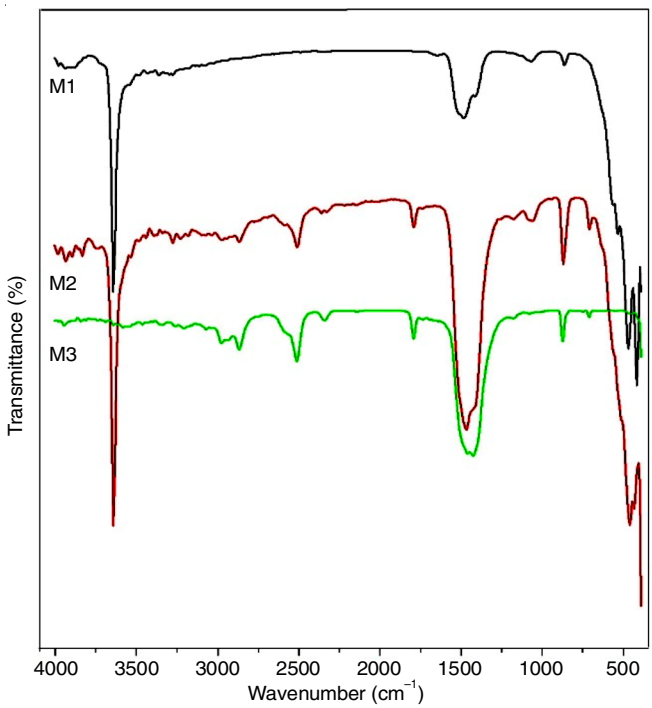


Fig. 3. FT-IR spectra of waste eggs shell powder calcinated at 800 °C (M1), 700 °C (M2) and 500 °C (M3)

supports to create the Brønsted acid sites [47]. Moreover, a strong peaks observed in 1600-1500 cm^{-1} and 1446-1410 cm^{-1} occured due to the deformation mode of the surface hydroxyl group and Ca-O bonding [47]. Furthermore, the strong bands were also observed at lower vibrational frequencies around at 500-400 cm^{-1}, which corresponds to the Ca-O bonds.

Elemental studies: The presence of calcium and its composition of waste eggs shell powder sintered at 800 ºC (M1), 700 ºC (M2) and 500 ºC (M3) were analyzed by energy dispersive spectroscopy. The eggs shell powder samples sintered at 800 ºC shows the presence of calcium and oxygen elements and absence of carbon element confirms the complete formation of calcium oxide (Fig. 4). The rest of the samples sintered at 700 ºC (M2) and 500 ºC (M3) show the presence of calcium, oxygen and carbon elements. The composition of an elements in weight and atomic percent of samples M1, M2 and M3 are shown in Table-1.

TABLE-1
COMPOSITION OF ELEMENTS IN WASTE EGGS SHELLS SAMPLES CALCINATED AT 500 °C (M-3), 700 °C (M-2) AND 800 °C (M-1)

Samples	Weight (%)			Atomic (%)		
	Ca	O	C	Ca	O	C
M1	35.16	64.84	0	17.80	82.20	0
M2	12.86	41.65	45.48	4.78	38.79	56.42
M3	14.80	45.32	39.88	5.66	43.43	50.91

Morphology studies: The morphological characteristics of the synthesized calcium oxide (CaO) from waste eggs at different calcined temperature were characterized by SEM. The SEM images in Fig. 5a shows the irregular shapes and particles were agglomerated no clear geometrical shapes were observed at 500 ºC. The SEM image (Fig. 5b) shows that slight different morphology of calcium oxide particles which are somehow looks like crystalline shapes annealed at 700 ºC. The highly crystalline and small needle and rod types of morphology was observed in the sintered sample at 800 ºC (M3, Fig. 5c). The

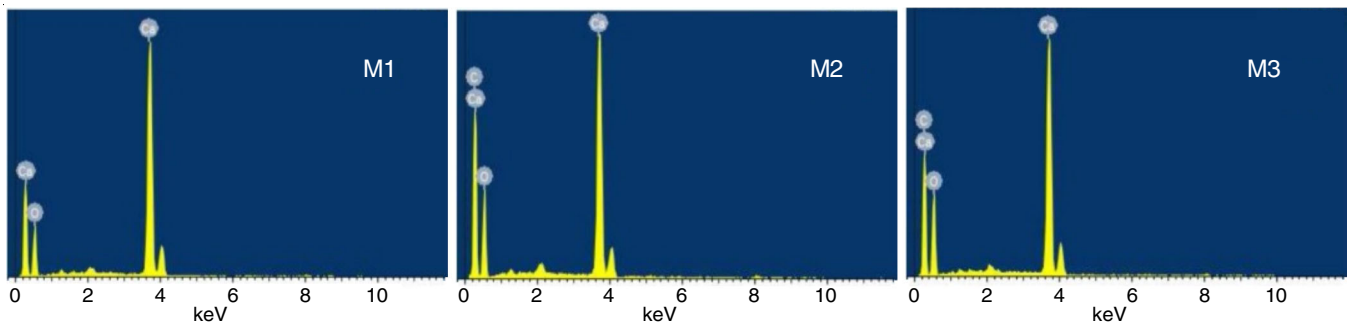


Fig. 4. EDS spectrum of waste eggs shell powder calcinated at 800 °C (M1), 700 °C (M2) and 500 °C (M3)

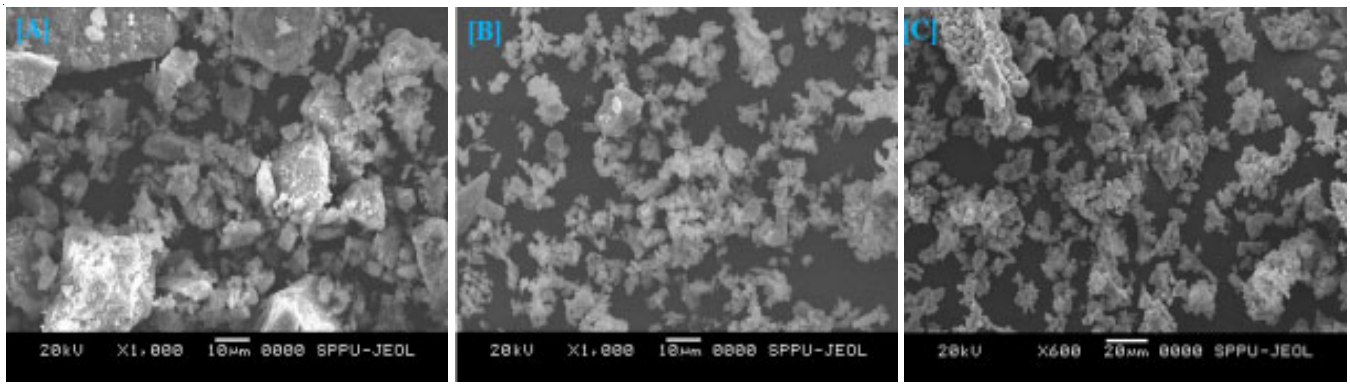


Fig. 5. SEM images of waste eggs shell powder calcinated at (A) 500 °C (M3), (B) 700 °C (M2) and (C) 800 °C (M1)

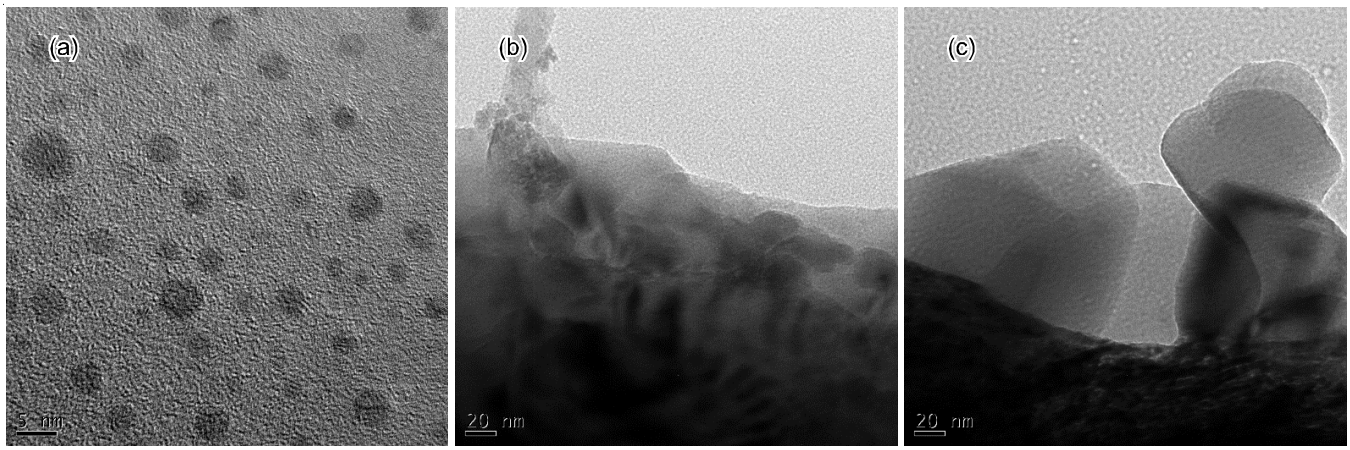


Fig. 6. TEM images of calcium oxide derived from waste egg's shell powder sintered at 800 °C

typical TEM images (Fig. 6) of eggs shell powder calcinated at 800 ºC shows that the particles were polydispersed and mostly spherical.

5-Arylidene malononitrile derivatives (3a-h)

Optimization of catalyst amount: The amount of catalyst was optimized by considering a model reaction of benzaldehyde (**1**) (5 mmol) and malononitrile (**2**) (5 mmol) in 10 mL ethanol and then the reaction mixture was exposed to ultrasound irradiation to yield 5-arylidene malononitrile. The catalyst was added in amounts of 0, 25, 50, 100 200 and 300 mg. The results indicated that 200 mg of catalyst amount gives excellent yield of product (Table-2), whereas in absence of catalyst, the reaction does not go to completion and the yield of product was very poor. Moreover, to examine the generality of this present method, a variety of different substituted aromatic aldehydes possessing an electron donating (CH_3, OCH_3, OH) and electron withdrawing groups (NO_2, Cl) offered good yields (93-98%) and reactions were completed within 1 min (Table-3).

TABLE-2
OPTIMIZATION OF CATALYST AMOUNT OF CALCIUM OXIDE FOR THE SYNTHESIS OF 5-ARYLIDENE MALANONITRILE UNDER ULTRASONIC IRRADIATION

Entry	Catalyst amount (mg)	Time (s)	Yield (%) of **3a**
1	0	200	50
2	25	180	72
3	50	120	75
4	100	80	78
5	200	30	98
6	300	65	85

[a]Isolated yield of the product.

Reusability studies: The recovery and re-applicability of the catalyst is important to check the capability of catalyst. The reusability of catalyst was examined using a model reaction between benzaldehyde and malononitrile in the presence of 200 mg of calcium oxide catalyst under ultrasound irradiation. The catalyst was separated, washed thoroughly with *n*-hexane, dried at 60 ºC and finally activated at 120 ºC for 1 h. The obtained catalyst was then reused for successive run under same conditions as shown in Fig. 7. It was observed that the catalyst was reused four times without any significant loss of its catalytic activity. Thus, calcium oxide catalyst is suitable and reusable even after four runs and the catalytic activity was almost as similar as the fresh one during the synthesis of various 5-arylidene malononitriles (96%, 95%, 93% and 93%, respectively).

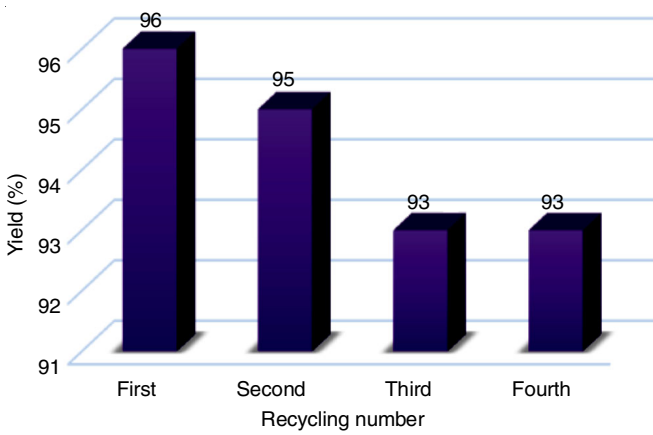


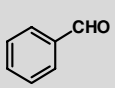
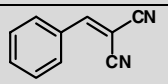
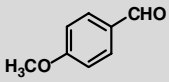
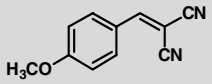
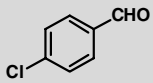
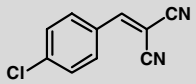
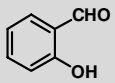
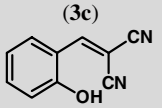
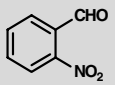
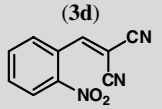
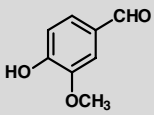
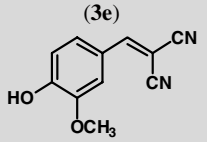
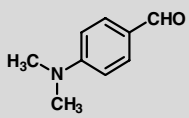
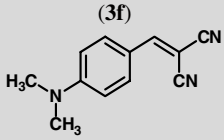
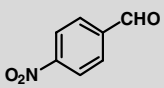
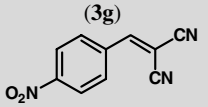
Fig. 7. Recovery and reusability of calcium oxide (**Scheme-I**)

5-Arylidene barbituric acid derivatives

To examine the generality of this present method, a variety of different substituted aromatic aldehydes possessing an electron donating (-CH_3, -OCH_3, -OH) and electron withdrawing groups (-NO_2, -Cl) offered good yields (92-98%) and the reactions were completed within 30 s (Table-4).

Reusability studies: The reusability of catalyst was examined by using a model reaction between benzaldehyde and barbituric acid in the manifest of 200 mg of calcium oxide catalyst under ultrasound irradiation for 25-35 s. The product was dissolved in hot water and the catalyst was filtered after the completion of reaction. The catalyst was then separated, washed thoroughly with *n*-hexane, dried at 60 ºC and activated at 120 ºC for 1 h before catalytic run. The catalyst was reused four times without loss of its catalytic activity as shown in Fig. 8. The catalyst is easy to separate by simple filtration and reused again. The most useful advantage of the catalyst is easy synthesis from waste eggs cells, inexpensive, non-toxic, easily recycled and reused.

TABLE-3
SYNTHESIS OF 5-ARYLIDENE MALONONITRILE DERIVATIVES CATALYZED BY CALCIUM OXIDE USING ULTRASOUND IRRADIATION METHOD

Entry	Substrate	Product	Time (s)	[a]Yield (%)	m.p. (°C) found	Ref.
1		 (**3a**)	15	98	81-83	[40]
2		 (**3b**)	30	93	112-114	[40]
3		 (**3c**)	25	96	160-162	[40]
4		 (**3d**)	3	97	98-99	[41]
5		 (**3e**)	20	98	138-140	[41]
6		 (**3f**)	25	93	136-137	[40]
7		 (**3g**)	35	96	178-180	[40]
8		 (**3h**)	15	98	158-160	[40]

Reaction condition: Benzaldehyde (5 mmol) and malononitrile (5 mmol), 200 mg calcium oxide catalyst, 10 mL ethanol; [a]Isolated yield.

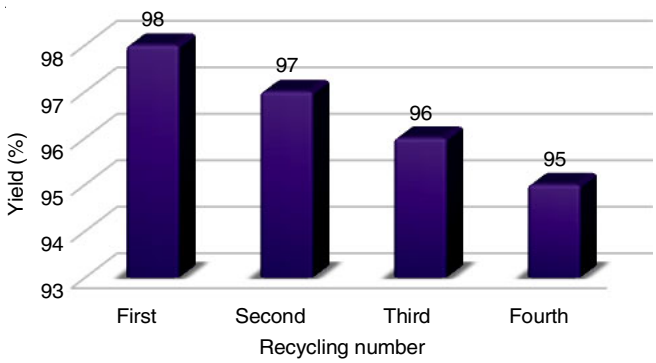


Fig. 8. Recovery and reusability of calcium oxide (**Scheme-II**)

Conclusion

In conclusion, calcium oxide (CaO) catalyst successfully synthesized from waste eggs shells in a simple manner. The X-ray diffraction pattern and FT-IR spectrum confirmed the successful formation of CaO. It was also clear that eggs shell powder calcinated at 800 ºC shows the average particle size of 12 nm probably provides high surface area. With increase in the calcination temperature, the surface morphology changes from irregular shape to highly crystalline and small needle and rod types of morphology as observed from SEM images. The typical TEM images of waste eggs shell powder calcinated at 800 ºC (M1) exhibited that the particles were polydispersed and mostly spherical. Finally, the synthesized calcium oxide was successfully applied as heterogeneous catalyst for the synthesis of 5-arylidene malononitrile and 5-arylidene barbituric acid derivatives by condensation of various aromatic aldehydes and active methylene compounds such as malononitrile and barbiutric acid using ultrasound irradiation method. The main advantageous of the present method under ultrasonic irradiation is that the products obtained within in a very short time 25-35 s only. The synthesized calcium oxide is non-toxic, inexpensive, shows excellent catalytic activity, easily recovered and recycled and acts as efficient heterogeneous catalyst.

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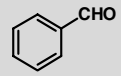
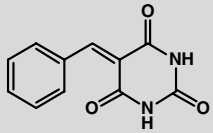
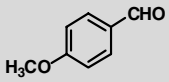
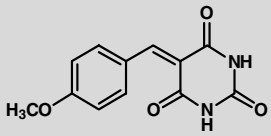
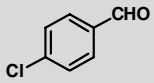
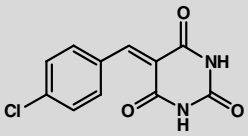
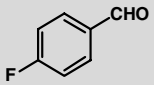
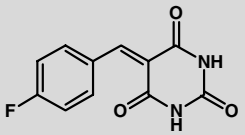
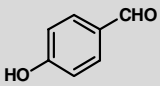
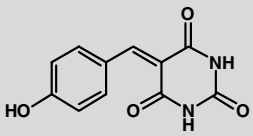
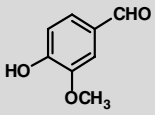
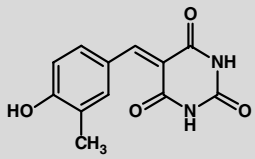
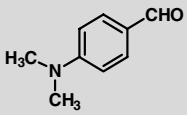
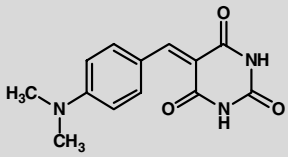
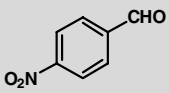
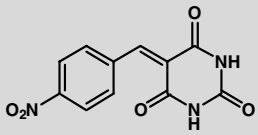
TABLE-4
SYNTHESIS OF 5-ARYLIDENE BARBITURIC ACID DERIVATIVES CATALYZED BY CALCIUM OXIDE USING ULTRASOUND IRRADIATION METHOD

Entry	Substrate	Product	Time (s)	[a]Yield (%)	m.p. (°C) found	Ref.
1		 (**4a**)	25	98	262	[34]
2		 (**4b**)	30	94	298	[34]
3		 (**4c**)	35	92	300	[34]
4		 (**4d**)	30	98	256	[34]
5		 (**4e**)	30	96	302	[34]
6		 (**4f**)	35	95	282	
7		 (**4g**)	35	92	314	
8		 (**4h**)	34	98	268	[34]

Reaction condition: Benzaldehyde (5 mmol) and barbituric acid (5 mmol), 200 mg calcium oxide catalyst, 10 mL ethanol; [a]Isolated yield.

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CONFLICT OF INTEREST

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

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