REVIEW

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A Low-Cost, Well-Designed Catalytic System Derived from Household Waste "Egg Shell": Applications in Organic Transformations

Manashjyoti Konwar[1] · **Mitali Chetia**[1] · **Diganta Sarma**[1]

Abstract

A waste feedstock-derived economical basic alternative catalyst is described in this review. Eggshell is one of the household wastes created in tons of weight daily. Therefore, in order to reduce the environmental pollution-related problems, its use in heterogeneous catalysis can be attributed as a great contribution for the chemical and material science society to carry out several known reactions and for the much-needed energy alternative biodiesel production as low-cost catalytic system. Keeping green chemistry in mind, industrial use of these catalysts may also reduce the use of other traditionally used high-cost chemical catalytic systems.

Keywords Egg shell powder · Heterogeneous catalysis · Green chemistry · Transesterification · Waste-derived catalyst

1 Introduction

1.1 Green Heterogeneous Catalysis and Egg Shell Powder (ESP)

Due to the greater toxicity problem and cost-effectiveness of organic chemicals, it is very necessary to reuse and recycle all the chemicals and catalysts as much as possible for the better and greener future of the environment. Keeping these points in mind, we have to plan our research work in a green manner so that there should be less harmful effects of chemicals on the environment [1]. To undergo a reaction, there should be some kind of catalyst that is responsible for dramatically enhancing the rate of a reaction without altering the thermodynamic equilibrium of a particular reaction [2–5]. Therefore, the important factor is to adapt the organic transformation

✉ Diganta Sarma
   dsarma22@gmail.com; dsarma22@dibru.ac.in

1   Department of Chemistry, Dibrugarh University, Dibrugarh, Assam 786004, India

with less pollution effects along with a reduction of energy and raw material consumption.

There are two types of catalysts: homogeneous and heterogeneous; however in this review, we will focus on heterogeneous catalysis and we are going to briefly discuss the use of egg shell powder [6–8] or calcined egg shell [9, 10]. Homogeneous catalysis are well recognized in synthetic organic chemistry, and have some disadvantages such as difficult separation of the soluble complexes from the reaction mixture, non-recyclability, possibility of breakdown of the complexes, decreasing catalytic activity, etc. [11, 12]. On the other hand, heterogeneous catalysts are easy and simple to recover from the reaction mixture, which furnish the practical importance in both research sectors as well as in the industrial sector, e.g., appropriate in flow reactors [13, 14]. Heterogeneous catalysts are less selective than the homogeneous ones due to the presence of a large number of active sites present on it [15]. In recent years, the development of green heterogeneous catalytic systems has attained great significance for its implication in the chemical processes, which may cause benign environment consequences with high selectivity of the desired molecules, great yield, and lesser amount of side products [16, 17].

With the rapid progress of the industrial sector day by day, there will be a possibility of an energy crisis in near future. Because of this energy crisis, improvements in renewable energies such as wind power, solar energy, and bio-derived solvent extraction have been focused on worldwide [18–20]. Therefore, the development of green and ecologically safe energy techniques for a better and sustainable future is always a great initiative with enormous significance [21]. Nowadays, economic and environmental apprehensions encourage researchers and scientists in the application of heterogeneous catalyst to carry out diverse organic transformations, which make the transformation clean, environmentally benign, and with a high product yield [22–24]. Thus, heterogeneous catalysis has a great impact on the field of synthetic organic chemistry due to its immense recyclability, reusability, and it attributes all these requirements for the synthetic organic chemistry to overcome the problems faced by the researchers [25–30].

1.2 Chemistry of ESP

Egg shell is an important natural calcium feedstock in the form of calcium carbonate along with little percentage of calcium phosphate [31–33]. Literature reveals that egg shell contains approximately 95% $CaCO_3$, 2% $Ca_3(PO_4)_2$, 2% $MgCO_3$ and 1% of organic substances mostly of albuminous character. In egg shell, the content of calcium is 28.2–41.2% and the content of phosphorus is 0.102% [34–36]. Due to the basic nature of ESP, [37–39] we thought it could be widely used in synthetic organic processes where an external base is necessary for a reaction to proceed. Accordingly, we carried out C–C (Suzuki–Miyaura cross-coupling reactions) and C-hetero atom (peptide coupling, click chemistry) bond formation reactions using ESP as a base alternative as well as solid support (discussed in the later part of this review). SEM-EDAX and TEM-EDAX analysis of the ESP are shown below [40] (Fig. 1):
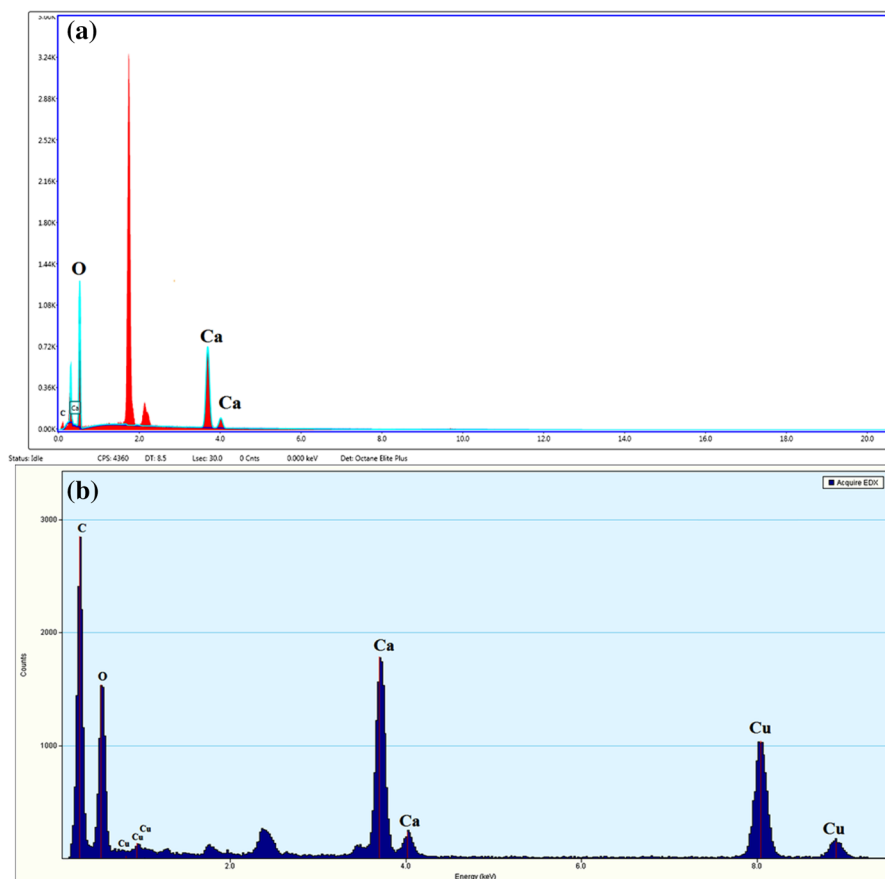
Fig. 1 **a** SEM-EDAX of ESP. **b** TEM-EDAX of ESP [40]

1.3 Traditionally Used Heterogeneous Catalyst vs. Egg Shell-Based Heterogeneous Catalyst

The principal features for environmentally acceptable processes are based on green heterogeneous catalysts since they are easy to handle and inexpensive in nature.

Literature reports reveal the development of many heterogeneous catalysts to carry out different organic transformations in recent decades. Heterogeneous catalysts are based on impregnation of transition metal on the solid support such as silica [41, 42], clay [43–53], surfactant/clay composites [54, 55], metal exchange clay composite [56, 57], etc.

The use of silica-supported reagents in one-pot multi-component constructions of heterocycles has received considerable importance in organic synthesis since these kinds of catalysts are easy to prepare, inexpensive, and have reusability power due to insolubility in volatile organic compounds (VOCs). Many silica-based reagents such as $FeCl_3$–SiO_2 [58], $HClO_4$–SiO_2 [59–61], Fe_3O_4@$nSiO_2$–NH_2–RhNPs@$mSiO_2$

nanocatalyst [62], Brønsted acids supported on silica [63], TfOH-SiO$_2$ [64] sulfuric acid on nano silica [65], were developed to carry out different organic transformations. These catalysts are very effective and inexpensive along with recyclability power.

Similarly, clay-supported heterogeneous catalysts also find significant applications and the processes using clay-modified catalysts are relatively greener. Since clays are readily available, very cheap, and non-toxic in nature, they have many advantages over other catalysts. A good number of papers are available on the use of Montmorillonite K10 [66–78], Montmorillonite KSF [79–85], kaolinite [86–88], bentonite [89, 90], etc., as solid supports for the preparation of heterogeneous catalyst.

Some other heterogeneous catalysts were prepared by using zeolite [91–97], solid acid [98, 99], nano-structured catalyst [100, 101], metal organic framework (MOF) [102–106], chitosan [107–114], hydrotalcite [115–127], titania [128, 129], alumina [130–134], cellulose [135–141], carbon/charcoal [142–150], polymer and nano-based composite [151, 152], tungstate [153, 154], molecular sieve [155–159], etc., for various organic transformations such as C–H activation [160] and C–C and C–N bond formations [161–163].

Comparing egg shell powder with other above-mentioned solid supports, we found that egg shell powder also acts as a solid support with superior selectivity, since calcium carbonate is insoluble in almost all of the organic solvents including water. By using ESP as a solid support, it is easy to recover the catalyst from the reaction mixture and reuse it. Several reports are available in the literature describing the use of egg shell as a solid support for different applications such as in synthesis of hydrogen/syngas, bioactive compound, and in waste-water treatment, etc., but to the best of our knowledge, no reports are available in the literature describing the ESP promoted organic transformations that will be discussed briefly in a later part of this review.

1.4 Importance of Biodiesel and Its Production Through Heterogeneous Catalyst vs. Egg Shell-Modified Catalyst

Due to the limited stock of non-renewable energy sources, researchers have been paying much more attention to energy alternatives since the last decade to meet energy demands. Therefore, to fulfill the increasingly high energy demand in everyday life, the research field has been directed towards the development of alternative and environmental friendly fuel with less pollution effects [164, 165].

Biodiesel, a renewable energy source, is composed of long-chain fatty acid alkyl ester. It has similar physical properties to that of petroleum diesel, with some advantages like being biodegradable, renewable, lower toxicity, and low emission of toxic chemicals. Foremost technique for the synthesis of biodiesel is done by transesterification of vegetable oil or animal fat in the presence of methanol by using low-cost catalysts (Scheme 1) [166–169]. The usual transesterification reaction for biodiesel production is carried out in the presence of a strong base and it is homogeneous in nature with certain disadvantages like equipment corrosion, formation of unwanted
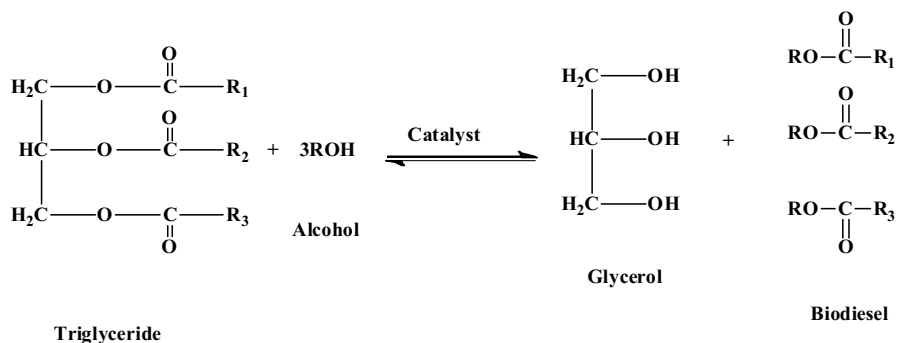
Scheme 1 Biodiesel synthesis by transesterification of triglycerides

by-products, being hard to separate, difficult to recycle, high temperature, along with certain environment-related problems. Therefore, the development of solid catalysts for biodiesel production always has a great importance and recently gained attention due to their ease of separation, lack of corrosion, and less toxicity problems.

Use of waste material-derived solid catalyst in biodiesel production has a great impact on the environment as well as on the cost-effectiveness of the catalyst. One of these kinds of waste solid catalysts was derived from eggshell, which is a household waste in daily life with a high percentage of calcium components. These kinds of eggshell-derived solid catalyst were used as heterogeneous catalysts for biodiesel production in batch reactors. Various research groups [166–170] derived such low-cost catalysts for biodiesel production, which is discussed in detail in the later part of this review.

Use of eggshell:

A. As heterogeneous catalysts in synthetic organic chemistry:

There are two ways through which eggshell can act as a heterogeneous catalyst: (i) through calcinations of the eggshell, and (ii) eggshell without calcinations. It is very hard to compare the efficiency between them since both contain calcium as the major source while the difference is that upon calcinations traces of organic matters are removed from the eggshell. Upon calcination, eggshell, which mainly contains $CaCO_3$, is converted to CaO and it is free from organic content. Therefore, both these catalysts are equally efficient towards different organic transformations and their applications are listed below in detail.

a. Use of CaO (from calcined egg shell) as catalyst for organic transformations:

The synthesis of Schiff base is always an important reaction in synthetic organic chemistry by the simple condensation of an aldehyde and an amine. Patil and cow-orkers [9] synthesized functionalized Schiff bases by using the natural catalyst (calcined egg shell or CES) under solvent-free conditions in which CES acts as a dehydrating agent (Scheme 2).
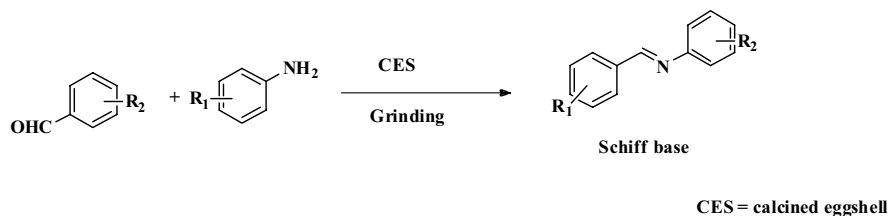
Scheme 2 Synthesis of Schiff base by using Calcined Egg Shell (CES)
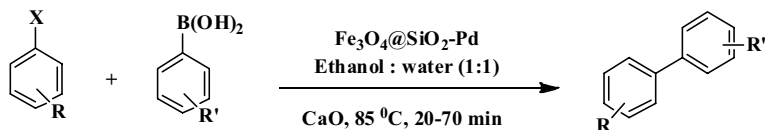
Similarly, Knoevenagel condensation reaction is another base catalyzed reaction in which condensation of an aldehyde and active methylene compound takes place. CES mainly contains calcium carbonate as the major component, which is basic in nature, and due to this, the condensation of the above compounds takes place. Patil et al. [171] also extended the work on CES, showing its application on the Knoevenagel condensation in aqueous medium as a heterogeneous catalyst as well as a base alternative (Scheme 3).

Khazaei et al. [172] carried out Suzuki–Miyaura cross-coupling reaction in the presence of nano-Fe_3O_4@SiO_2 supported Pd(0), which is a magnetically recoverable nanocatalyst. Here, the silica was synthesized from rice husk and considered as a natural support for the stabilization of magnetic palladium nanoparticles. Due to the combination of palladium source and egg shell, Suzuki–Miyaura reactions easily proceeded giving the desired biaryl moieties in the presence of calcium oxide with the binary mixture of water and ethanol as solvent (1:1) at 85 °C. The synthesized catalyst was well characterized by different spectroscopic analyses such as UV–Vis, FT-IR, X-ray diffraction (XRD), scanning electron microscope (SEM), transmission electron microscope (TEM) and energy-dispersive X-ray spectroscopy (EDX), etc. (Scheme 4).

Waghadhare and coworkers [173] also synthesized a magnetically separable cobalt-iron nano-catalyst based on egg shell as a heterogeneous catalyst as well as a base alternative for Knoevenagel condensation reaction (Scheme 5).
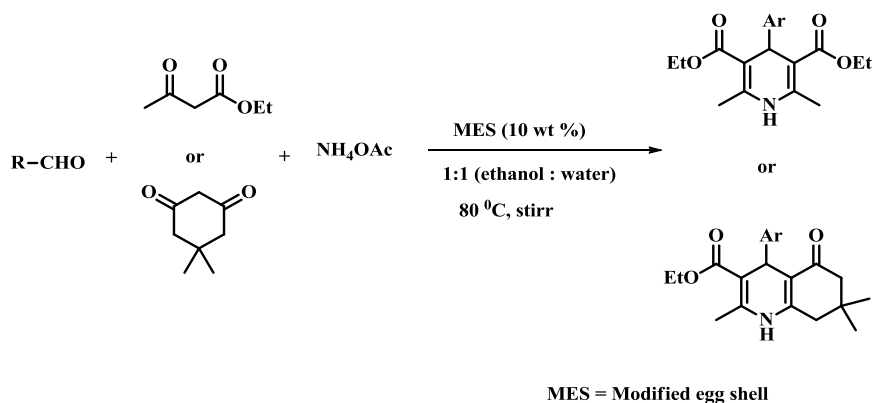

R' = -CN, -COOEt
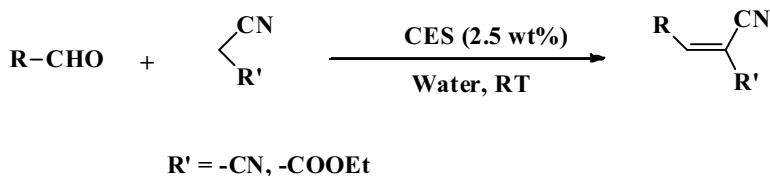
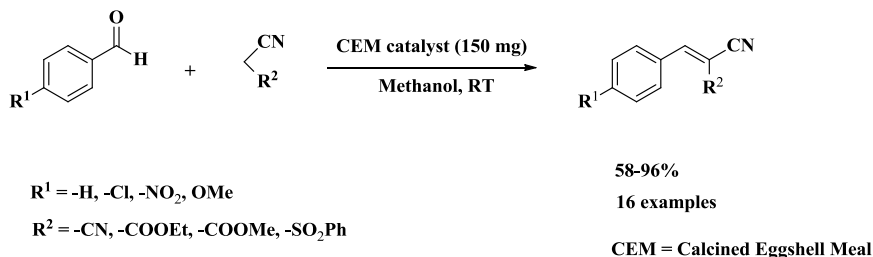**Scheme 3** Knoevenagel condensation reaction in presence of CES as catalyst

Scheme 4 Suzuki-Miyaura reaction in presence of Pd-eggshell based catalyst
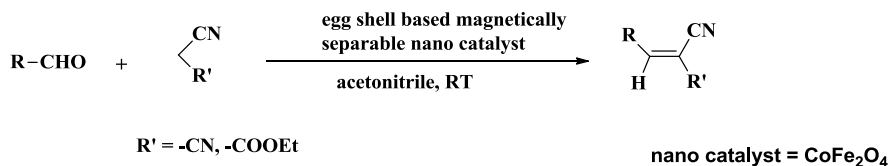
Scheme 5 Knoevenagel condensation reaction inpresence of egg shell based magnetically separable nano catalyst
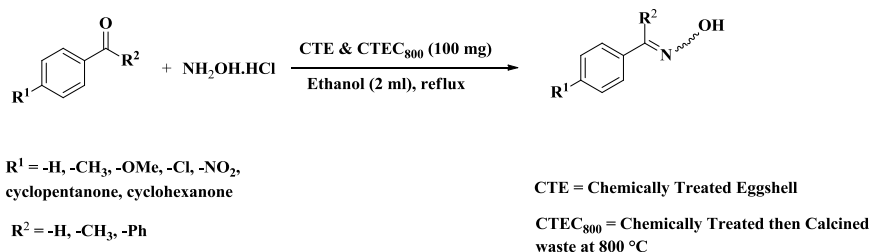
Scheme 6 Modified eggshell (MES) catalyzed synthesis of 1,4-dihydropyridine and polyhydroquinoline

Scheme 7 Eggshell meal (CEM) catalyzed Knoevenagel condensation reaction

Morbale et al. [174] synthesized 1,4-dihydropyridine and polyhydroquinoline through a green and efficient catalytic system by the simple summarizing of aldehydes, dimedone, or ethylacetoacetate and ammonium acetate using an alternative heterogeneous basic catalyst "modified eggshells" (MES) in a water:ethanol system at 80 °C (Scheme 6).

Riadi et al. [175] also carried out the Knoevenagel reaction in methanol at room temperature using calcined eggshell meal (CEM), which was considered as an alternative green solid support as heterogeneous media since it was easy to separate and reuse. Due to these advantages, egg shell can be considered as a well-established solid support for organic transformations (Scheme 7).

R^1 = -H, -CH$_3$, -OMe, -Cl, -NO$_2$,
cyclopentanone, cyclohexanone

R^2 = -H, -CH$_3$, -Ph

CTE = Chemically Treated Eggshell

CTEC$_{800}$ = Chemically Treated then Calcined
waste at 800 °C

Scheme 8 Synthesis of oxime derivatives by the use of chemically treated eggshell

Taleb et al. [176] synthesized oxime derivatives by simple condensation of aldehyde and hydroxylamine hydrochloride by the use of chemically treated eggshell as a heterogeneous catalyst in ethanol at reflux conditions. The catalytic activity of this newly developed heterogeneous catalyst can be used up to seven times (Scheme 8).

Gao et al. [177] synthesized dimethyl carbonate (DMC) using waste eggshell as a heterogeneous catalyst by the treatment of propylene carbonate and methanol at 25 °C and 1 atm pressure. Calcined eggshell was found to be a highly active base, reusable solid heterogeneous catalyst for organic transformations instead of using traditional solid supports, and characterized by using various analytical techniques such as TGA, XRD, nitrogen physisorption, and energy-dispersive X-ray spectroscopy (EDS) (Scheme 9).

b.   Use of CaCO$_3$ (from uncalcined egg shell) as catalyst for organic transformations:

M. Khazaei et al. [178] developed reusable and efficient Pd nanoparticles using barberry fruit extract through in situ greener reduction method and then supported over eggshell. Here, barberry fruit extract has a dual role of reducing agent as well as stabilizing agent for palladium nanoparticles. These newly developed Pd nanoparticles were applied for ligand-free ipso-hydroxylation of phenylboronic acid and
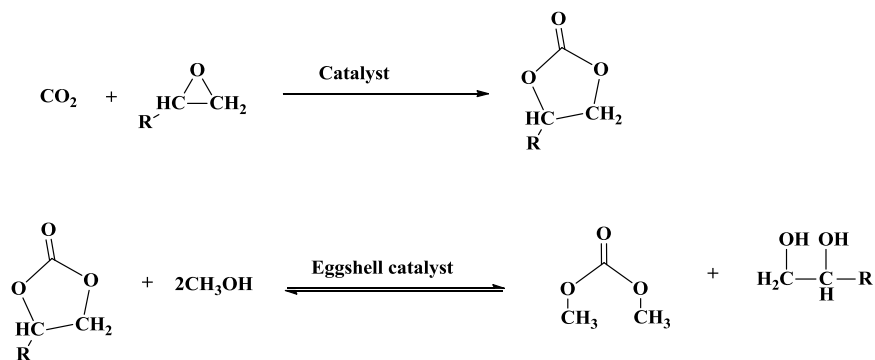
**Scheme 9** Synthesis of dimethyl carbonate (DMC) by using waste eggshell
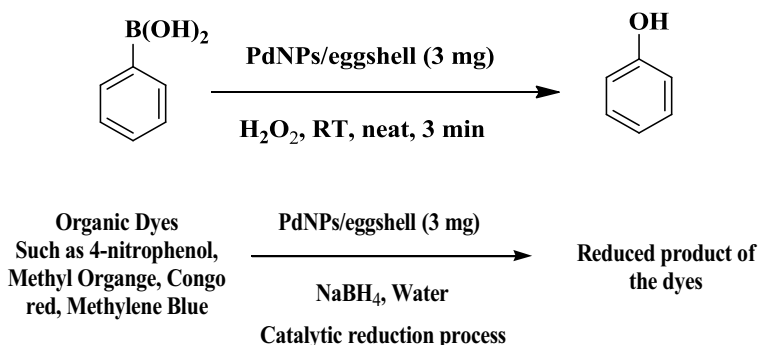
Scheme 10 Ligand-free ipso-hydroxylation and catalytic reduction of organic dyes by using eggshell supported PdNPs

catalytic reduction of different organic dyes, such as 4-nitrophenol, methylene blue, Congo red, and methyl orange by using $NaBH_4$ and aqueous media at room temperature (Scheme 10).

Montilla et al. [179] developed a feasible way to produce lactulose from lactose by employing egg shell as a catalyst (which accelerates the formation of lactulose) through ultra filtrate, which is an alternative for consumption of these industrial wastes. Here, the reaction was stirred and refluxed at 98 °C in glycerol for 60 min using 6 mg/ml of egg shell catalyst loading giving the optimal production of lactulose (Scheme 11).

Youseftabar-Miri et al. [180] carried out the organic transformations for the synthesis of pyrano[3,2-c]quinoline derivatives using eggshell as a heterogeneous catalyst at 60 °C and ethanol as a solvent and described that the eggshell catalyst can be recovered and reused for several times without losing its activity (Scheme 12).

Mallampati et al. [181] also reported a simple method for synthesis of nanoparticle in which metal cations are reduced by eggshell membrane to metal atoms. Eggshell membrane (ESM) stabilized these nanoparticles and acts as a supporting material for the nanoparticles. These newly developed membrane-supported nanoparticles were employed as a heterogeneous catalyst for the nitro reduction and synthesis of propargylamine and these nanoparticles were used for the testing of their catalytic efficiency (Scheme 13).
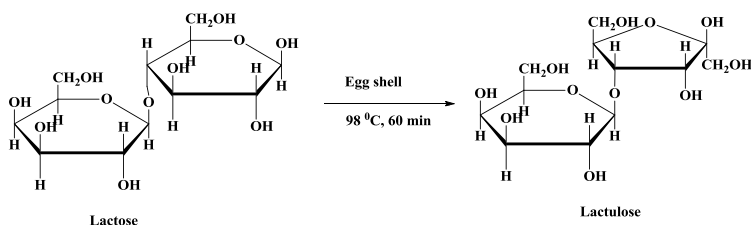
**Scheme 11** Eggshell catalyzed lactulose synthesis from lactose
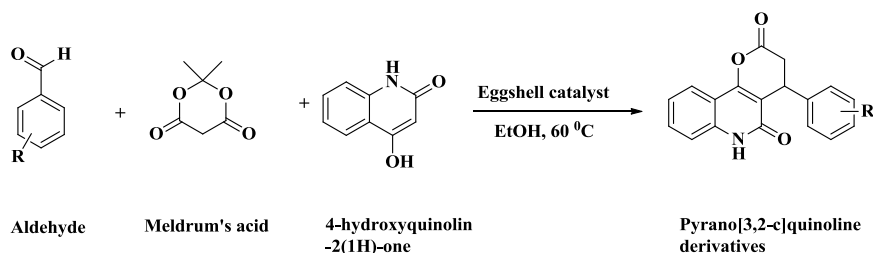
Scheme 12 Eggshell catalyzed synthesis of pyrano[3,2-c]quinoline derivatives
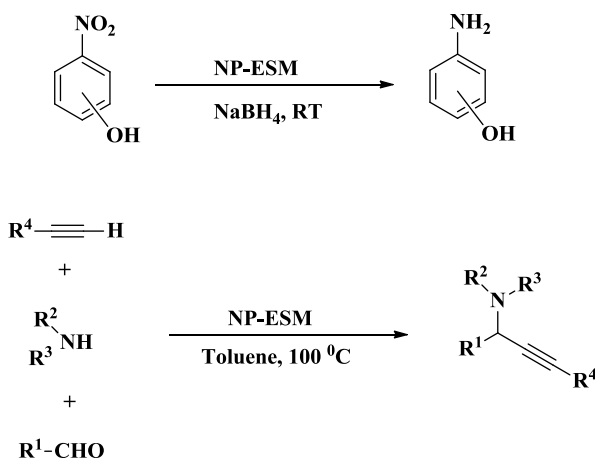
Scheme 13 Eggshell membrane (ESM) supported nanoparticles as a catalyst for nitro reduction and synthesis of propargylamine derivatives

Our group extended the scope of eggshell heterogeneous catalyst and successfully carried out several reactions such as click or azide-alkyne cycloaddition, peptide coupling reaction, and palladium catalyzed Suzuki–Miyaura reaction using ESP [40, 182, 183]. The role of eggshell in these reactions is a base alternative/reducing agent. Moreover, eggshell is insoluble in water; therefore it also acts as a solid support for these reactions. In case of peptide synthesis, ESP neutralizes the amino acid methyl ester hydrochloride, a coupling partner of the peptide synthesis. Again, in Suzuki–Miyaura reaction, ESP acts as an internal base as well as a solid support for incorporation of in situ-generated palladium nanoparticles. The Pd nanoparticles, stabilized by several components of eggshell powder, were characterized by SEM, SEM-EDAX, TEM, TEM-EDAX, and XRD analysis [40] (Fig. 2 and Scheme 14).

Kuhn et al. [184] developed Pd–Ag/Al$_2$O$_3$ egg-shell catalysts with different Pd/Ag mole ratios and tested for selective hydrogenation of acetylene under tail-end conditions in H$_2$ atmosphere. Here, the total number of Pd surface atoms has
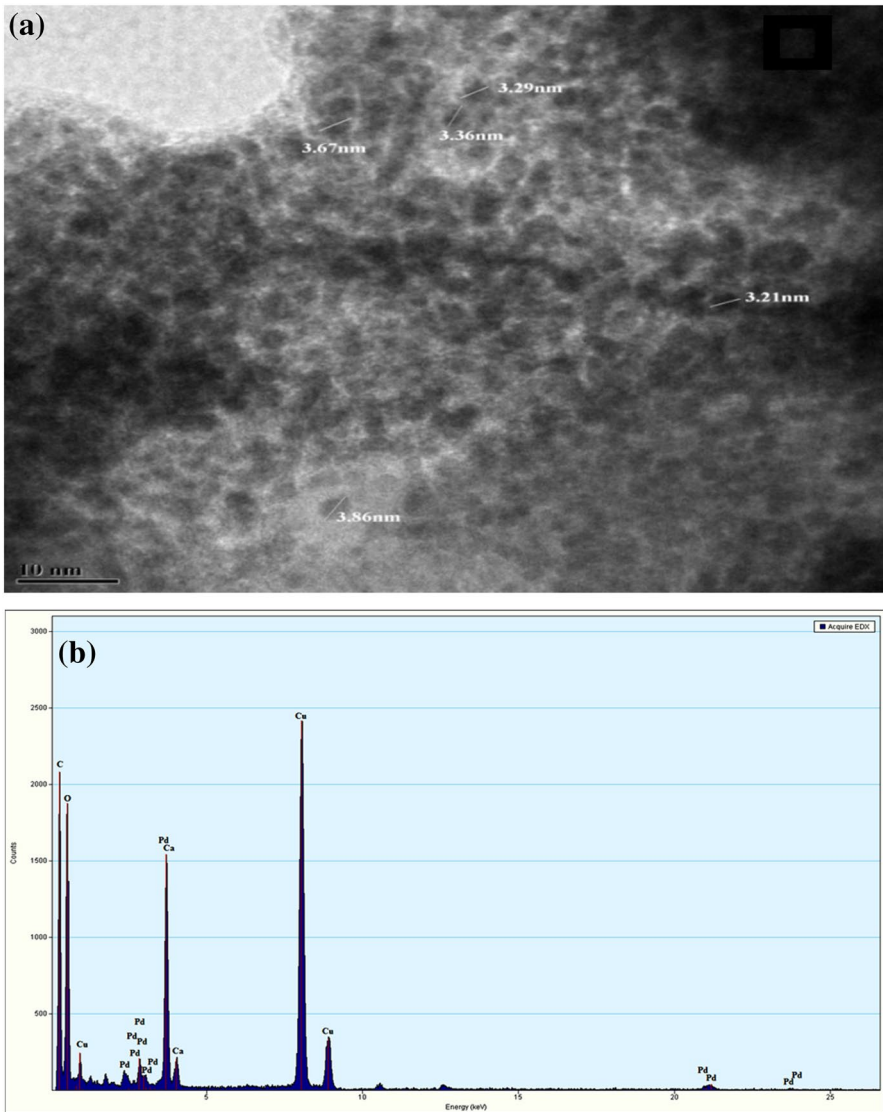
Fig. 2 **a** TEM image of Pd-ESP through in situ generation of palladium nanoparticles. **b** TEM-EDAX of Pd-ESP composite

been reduced and isolation of large Pd ensembles takes place, and it is confirmed from characterization data (Scheme 15).

Shao et al. [185] have developed a simple and efficient approach in which eggshell $Pd/SiO_2–Al_2O_3$ catalyst proceeds through CO reduction deposition of $PdCl_2$ at room temperature and aqueous ethanolic solution. Compared to a traditionally used catalyst, this catalytic system has appreciably higher activity in the partial hydrogenation of phenyl acetylene and the formation of ethylbenzene is

Click reaction
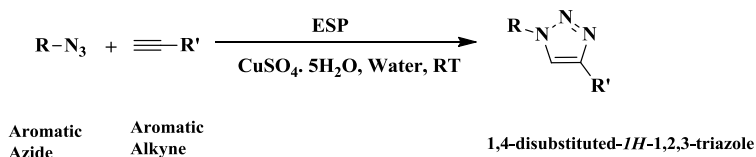
Peptide coupling
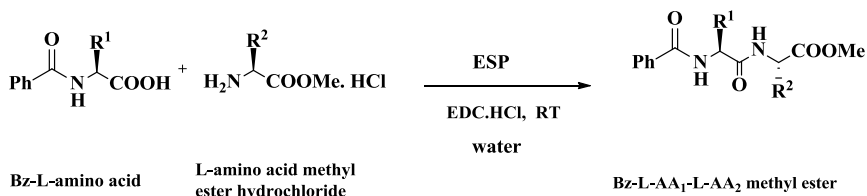
Suzuki-Miyaura reaction through insitu generated palladium nanoparticles
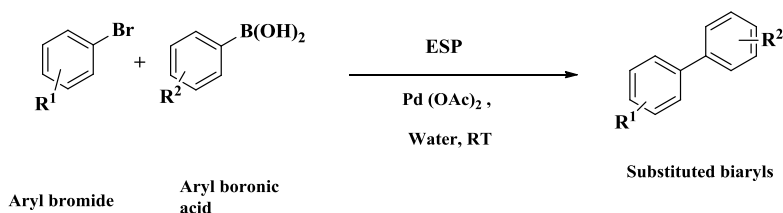
Scheme 14 Eggshell powder (ESP) catalyzed Click or azide-alkyne cycloaddition, peptide coupling reaction and Suzuki-miyaura reaction
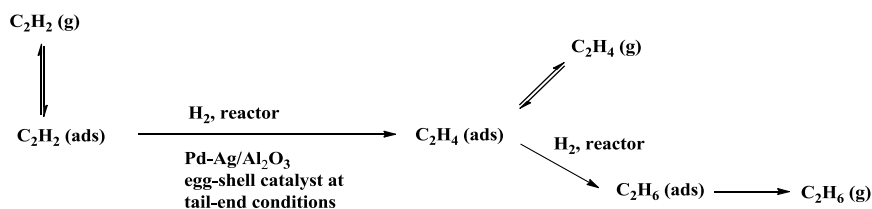
Scheme 15 Selective hydrogenation of acetylene in presence of Pd-Ag/Al$_2$O$_3$ egg-shell catalyst

suppressed due to uniform active metal distribution over the surface of eggshell (Scheme 16).

Wen et al. [186] also worked on selective hydrogenation of pyrolysis gasoline by using eggshell Ni/Al$_2$O$_3$ catalyst in a micro-flow reactor. Here, Wen and coworkers derived this catalyst from LDHs precursor with advanced catalytic hydrogenation
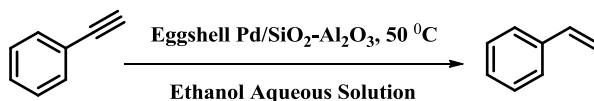
Scheme 16 Partial hydrogeneation of Phenyl acetylene in presence of eggshell Pd/SiO$_2$-Al$_2$O$_3$ catalyst
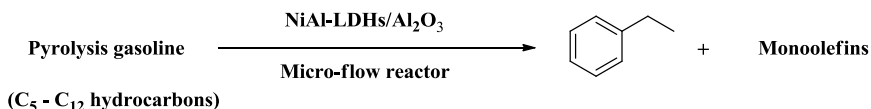
Scheme 17 Selective hydrogenation of pyrolysis gasoline by using eggshell Ni/Al$_2$O$_3$ catalyst

Scheme 18 Selective hydrogenation of styrene in presence of eggshell-Pd supported catalyst over polymeric composite materials

efficiency, mainly due to the deposition of Ni metal on Al$_2$O$_3$ as a solid support with stronger interaction between the nickel metal and Al$_2$O$_3$ support (Scheme 17).

Badano et al. [187] developed a supported Pd catalyst over two new polymeric composite materials UTAI (made from triethylene glycol dimethacrylate (TEG), benzoyl peroxide (BPO) and diurethane dimethacrylate (UDMA) and m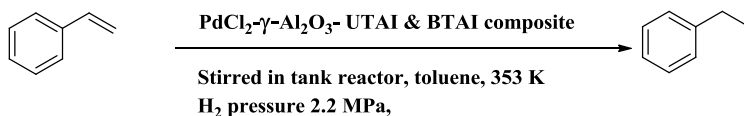ixed with alumina in monomer ratio of 1:1) and BTAI [made from TEG, BPO and bisphenol A glycerolate dimethacrylate (BGMA) and mixed with alumina in monomer ratio of 1:1] which was found to be extremely active for the selective hydrogenation of styrene stirred in tank reactor at 353 K and H$_2$ pressure in toluene as a solvent. With smaller diffusive restriction of the catalyst than other supported Pd catalysts made the catalyst more efficient for hydrogenation reaction (Scheme 18).

Khajavi et al. [188] developed an eggshell-based Pt-metal organic framework catalyst that can be applied in the selective hydrogenations of olefin mixtures to 1-octene, benzonitrile to benzylamine, and linoleic acid to oleic and stearic acid (Scheme 19).

Gao et al. [189] also developed TiO$_2$-modified porous hollow silica nanoparticles supported Pd-based eggshell nano-catalysts for selective hydrogenation of acetylene. Here, modified TiO$_2$, as support, promotes the selectivity for hydrogenation of acetylene, which showed better performance in the reaction to reduce the temperature and time to 300 °C and 1 h instead of 500 °C and 3 h in TiO$_2$ alone (Scheme 20).

Richter et al. [190] carried out nitric oxide selective catalytic reduction by using novel eggshell MnO$_x$/NaY composite over ammonia, which was operated at
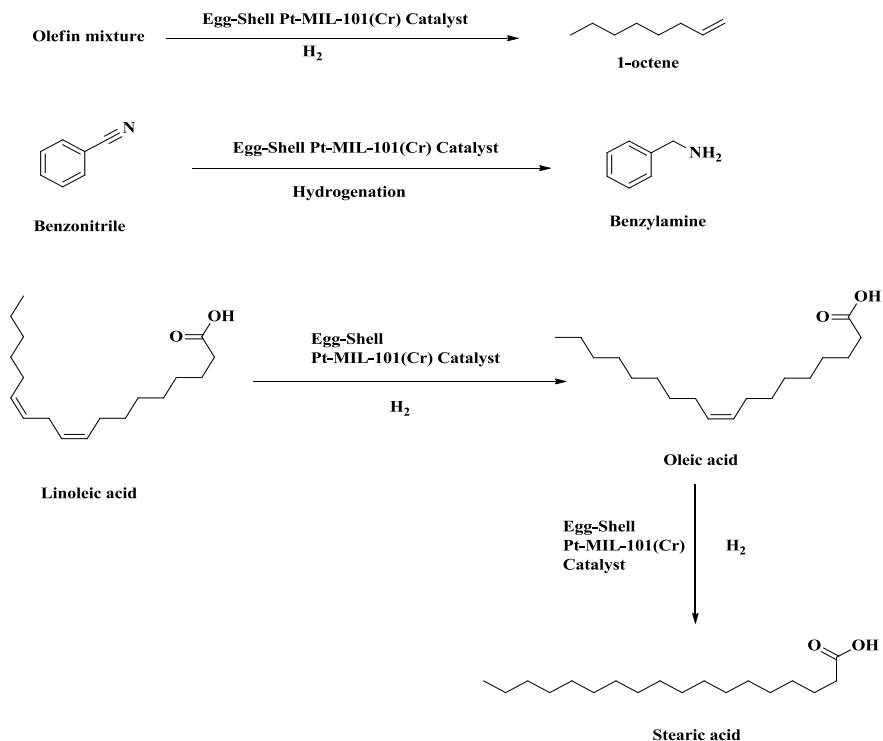
Scheme 19 Selective hydrogenation of olefin mixtures, benzonitrile and linoleic acid in presence of egg-shell-based Pt-metal organic framework catalyst
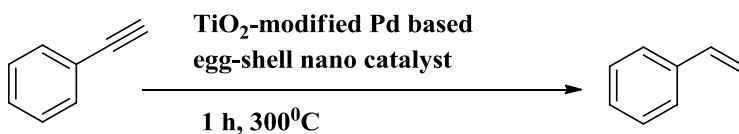
Scheme 20 Selective hydrogenation of acetylene in presence of TiO$_2$-modified Pd based eggshell nano catalyst
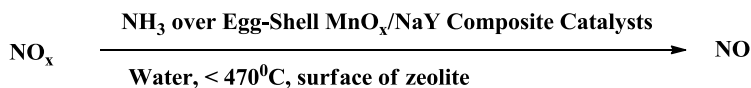
Scheme 21 Nitric oxide selective reduction in presence of eggshell MnO$_x$/NaY composite

temperatures lower than 470 K in water as a solvent giving around 80–100% NO conversion (Scheme 21).

$$NH_3 \xrightarrow[\text{Decomposition, 650 }^0\text{C, water}]{\textbf{Fe-Ni/}\boldsymbol{\gamma}\textbf{-Al}_2\textbf{O}_3 \textbf{ Egg-Shell Catalyst}} N_2$$

Scheme 22 Ammonia decomposition reaction in presence of eggshell based catalyst

Coke plant wastewater $\xrightarrow[\textbf{Bubbling bed reactor,}]{\textbf{Ru/eggshell catalyst, 250 }^0\textbf{C, 4.8 MPa}}$ **Removal of harmful chemicalsfrom water to reduce water pollution**

(contains high NH$_3$ content, cyanide and thiocyante, toxic organic compounds ike phenols,   monoand polycyclic nitrogen-containing aromatics, oxygen and sulfur containing   heterocyclic compounds and polynuclear aromatic hydrocarbons)

Scheme 23 Wet air oxidation of waste water of coke plant by using eggshell/ruthenium-based catalyst

Silva et al. [191] synthesized an efficient and novel Fe–Ni/γ–Al$_2$O$_3$ eggshell catalyst for the decomposition of ammonia to H$_2$ at 650 °C and water as a solvent (Scheme 22). Here, the eggshell catalyst shows superior activity towards ammonia decomposition and can reduce the ammonia trace concentration to equilibrium level.

Yang et al. [192] described wet air oxidation of waste water of coke-plant over eggshell/ruthenium-based catalyst for high chemical oxygen demand (COD) and ammonia/ammonium compounds (NH$_3$–N) exclusion at temperature (250 °C), which is relatively efficient co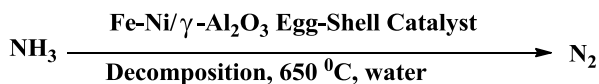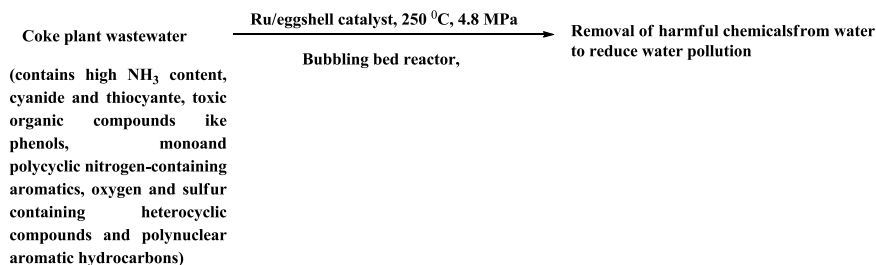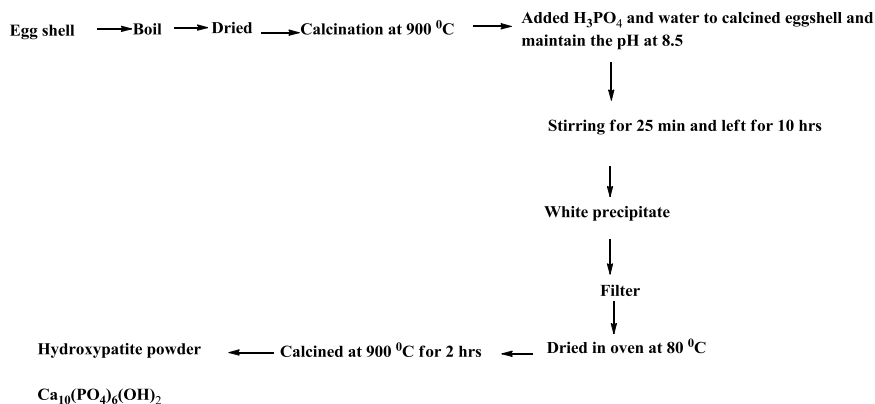mpared to other ruthenium-based catalysts with the same amount of loading. The reason for the high activity of eggshell catalyst towards treatment of waste water of coke-plant can be ascribed to the higher density of active Ru sites in the eggshell layer compared to other uniform catalysts with the same amount of Ru loading (Scheme 23).

Khandelwal et al. [193] developed a method for the synthesis of hydroxyapatite (HA) with the chemical formula Ca$_{10}$(PO$_4$)$_6$(OH)$_2$, which is the major chemical constituent of human bone tissue (70%). This is the reason why it has been widely engaged in the dental and non-load bearing implantations, to cope up with the bone response as a bioactive material. Here, wet chemical method was employed to synthesize HA powder using eggshells and phosphoric acid (H$_3$PO$_4$). HA powder was characterized by XRD, SEM, EDX FTIR, and TGA-DTA analysis (Scheme 24).

B.   As heterogeneous catalysts for biodiesel production

Buasri et al. [165] described an eggshell waste-derived catalyst through calcination of eggshell, which converts eggshell calcium carbonate to calcium oxide at 600–900 °C for 4 h. For catalyst testing, it was applied in transesterification of palm oil in the presence of methanol for the synthesis of biodiesel for 1–5 h, at 50–70 °C with 10–30 wt% catalyst loading (Table 1, entry 1).

Egg shell $\longrightarrow$ Boil $\longrightarrow$ Dried $\longrightarrow$ Calcination at 900 ^{0}C $\longrightarrow$ Added H$_3$PO$_4$ and water to calcined eggshell and maintain the pH at 8.5

$\downarrow$

Stirring for 25 min and left for 10 hrs

$\downarrow$

White precipitate

$\downarrow$

Filter

$\downarrow$

Hydroxypatite powder $\longleftarrow$ Calcined at 900 ^{0}C for 2 hrs $\longleftarrow$ Dried in oven at 80 ^{0}C

Ca$_{10}$(PO$_4$)$_6$(OH)$_2$

Scheme 24 Hydroxyapatite synthesis by the use of eggshell catalyst

Wei et al. [166] also described an eggshell-based solid catalyst by calcination of egg shell at 200–1000 °C for 2 h in a muffle furnace under static air. The catalytic activity was confirmed by applying it for the biodiesel synthesis from soybean oil and methanol in a batch reactor under 65 °C and vigorous stirring with 3 wt% of catalyst loading (Table 1, entry 2).

Yin et al. [194] also developed an eggshell catalyst for the biodiesel synthesis from soybean oil deodorizer distillate (SODD). Here, eggshells were calcined in a muffle furnace up to 900 °C to get the CaO particles. This catalyst was then employed for the transesterification of pre-esterified SODD with methanol in varying catalyst loading, time, and temperatures for the synthesis of biodiesel (Table 1, entry 3).

Navajas et al. [195] derived a catalyst based on eggshell that was calcined in a muffle furnace at 900 °C for 2 h. This catalyst was applied in used cooking oil (UCO) in a batch-type reactor along with previously mixed 2% sulfuric acid–methanol solution at 60 °C for 3 h to obtain the desired biodiesel (Table 1, entry 4).

Jazie et al. [196] developed an egg shell eco-friendly catalyst for transesterification of rapeseed oil for the biodiesel production in which eggshell was calcined in a muffle furnace up to a temperature of 1000 °C for 2 h under static air. This eco-friendly catalyst was characterized by XRD, N$_2$ adsorption–desorption, BET surface area, and FTIR analysis. This catalyst was applied for biodiesel production by the treatment of methanol along with rapeseed oil, which was previously heated at 378 K for 1 h in N$_2$-atmosphere to remove water and other volatile organic impurities and cooled (Table 1, entry 5).

Boro et al. [197] synthesized eggshell-derived CaO/Li-doped catalyst which was calcined at 800 °C for 2 h and was used for biodiesel production from nonedible oil feedstock with 94% of conversion with 5% catalyst along and 2% of lithium loading. This derived catalyst was characterized by using XRD, FTIR, and BET surface area

Table 1 Various egg shell-based catalysts used for biodiesel production

Entry	Reactant	Reaction condition	Product	References
1	Palm oil + methanol	10–30 wt% of eggshell catalyst (calcined at 900 °C) for 1–5 h at 50–70 °C	Biodiesel	[165]
2	Soybean oil + methanol	3 wt% of eggshell-based solid catalyst (calcined at 1000 °C for 2 h) in batch reactor at 65 °C	Biodiesel	[166]
3	Soybean oil Deodorizer Distillate + methanol	Eggshell-based catalyst (calcined in muffle furnace at 900 °C to get CaO particle) in reactor at 70 °C	Biodiesel	[194]
4	Used cooking oil (UCO) + methanol	Eggshell-based catalyst (calcined in muffle furnace at 900 °C for 2 h) and 2% H_2SO_4 in Batch Reactor, 60 °C for 3 h	Biodiesel	[195]
5	Rapeseed oil + methanol	Eggshell-based catalyst (calcined in muffle furnace at 1000 °C for 2 h) at 378 K for 1 h using N_2 atmosphere	Biodiesel	[196]
6	Nonedible oil Feedstock + methanol	5% wt of eggshell catalyst (CaO-derived Li-doped catalyst calcined at 800 °C for 2 h) at 60 °C for 3 h	Biodiesel	[197]
7	Palm oil + methanol	3–10 wt% of eggshell-derived catalyst (calcined at 800 °C for 4 h) at 60 °C for 1–3 h	Biodiesel	[198]
8	Palm olein oil + methanol	10 wt % eggshell-derived CaO catalyst (calcined at 800 °C for 4 h) at 60 °C	Biodiesel	[199]
9	Waste frying oil + methanol	Eggshell catalyst (calcined in muffle furnace at 900 °C for 2.5 h) at 65 °C in reactive distillation system or CaO catalyst (prepared through calcinations-hydration-dehydration technique of egg shells at 65 °C for 1 h	Biodiesel	[170, 200–202]

analysis. This catalyst was used for esterification of *Mesua ferrea* Linn (Nahor oil) with concentrated H_2SO_4 and methanol for 3 h at 60 °C (Table 1, entry 6).

Chen et al. [198] described calcium oxide derived from eggshell and explored the effectiveness of the catalyst for the biodiesel synthesis through transesterification of palm oil. Here, the ostrich eggshells were calcined at 800 °C for 4 h with a heating rate of 10 °C/min. The catalyst was used in the reaction along with palm oil and methanol at 60 °C for 1–3 h with varying amount of ultra power and low amount of catalyst loading (3–10 wt%) (Table 1, entry 7).

Viriya-empikul et al. [199] described a catalyst derived from eggshell $CaCO_3$, which was transformed to CaO by calcinations in the presence of air at 800 °C for 4 h for the biodiesel production from palm olein oil. This catalyst was characterized and confirmed by using XRD, TG/DTA, SEM, EDXRF, N_2 adsorption, and BET surface area analysis and applied in palm olein oil and methanol at 60 °C with 10 wt% of catalyst (Table 1, entry 8).

Niju et al. [170, 200–202] described biodiesel production using waste egg shells as heterogeneous catalyst derived from eggshell through calcination in a muffle furnace at 900 °C for 2.5 h under static air in order to form active CaO catalyst which was characterized by using XRD, SEM, EDAX, and BET surface area analyzer. Afterwards, this newly developed calcined catalyst was applied in biodiesel synthesis from waste frying oil (WFO) with methanol at 65 °C in a reactive distillation (RD) system. The general method for the production of biodiesel was also employed by using the same catalyst from WFO giving around 95% of conversion with 3 wt% of the catalyst in methanol at 65 °C for 3 h. Similarly, Niju and coworkers also derived highly active CaO catalyst through calcination-hydration-dehydration technique of egg shells. It was used for biodiesel production from WFO with methanol for 1 h at 65 °C (Table 1, entry 9).

2 Conclusions

The use of eggshell, a household waste material, -derived feedstock reduces the probable pollution-related problems and can be used as a better renewable catalyst/ product for maximum use of feedstock. Again, the production of biodiesel using eggshell as catalyst/solid support has gained much interest in the field of chemistry and chemical engineering for a better and sustainable future. The industrialization of this kind of waste feedstock for synthesis of the above-mentioned reactions may also get high significance and thus it is expected that eggshell will definitely play an important role in biodiesel production and heterogeneous catalysis in the near future.

Compliance with ethical standards

Conflict of interest The authors declare no competing financial interests.

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