Potential Uses of Waste Eggshell: A Review

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Abstract— India is the third largest producer of eggs in world subsequently producing tons of eggshell as a solid waste every year. Eggshells is mostly sent to landfill with high management cost and is considered hazardous waste according to UE regulations. However with an emerging interest in value extraction from waste eggshell several studies and experiments has been done. Waste eggshell shell has been successfully calcined and used as an active catalyst and found to be even more active than mineral CaO catalyst. The research work extends to application in Fertilizer, Gasification, Energy storage, Cement, Paper, Bio-diesel, Waste water Treatment, Food and Dietary Supplement. With such a broad spectrum of applications this review paper is an attempt to summarize all the key finding of research works, their outcomes, ambiguities in result and proposals, gaps in results and finding.

This review paper by scanning entire chemical and allied industry and their products globally takes opportunity to suggest possible application of eggshell where no application/published research work has been found like in soda ash, Methanol and calcium carbide.

Keywords: Eggshell waste, Waste Eggshell utilization

I. INTRODUCTION

India is the third-largest egg producer in the world after China and the USA and the fourth-largest chicken producer in the world after China, Brazil and the USA. In India, the per capita consumption of eggs has gone up from 30 eggs per annum to 68 eggs per annum. (Press Information Bureau 2018)

The total Egg production in the country during 2016-17 has been reported as 88.1 Billion showing a growth rate of 12.3% over 2013-14. Further, the productions have been estimated to be increased by 6.3% in 2016-17 over 2015-16. (Statista 2017)

About 96% of eggshell content is calcium carbonate, magnesium carbonate 1%, calcium phosphate 1% rest organic materials and water (Oliveira, Benelli and Amante 2013).

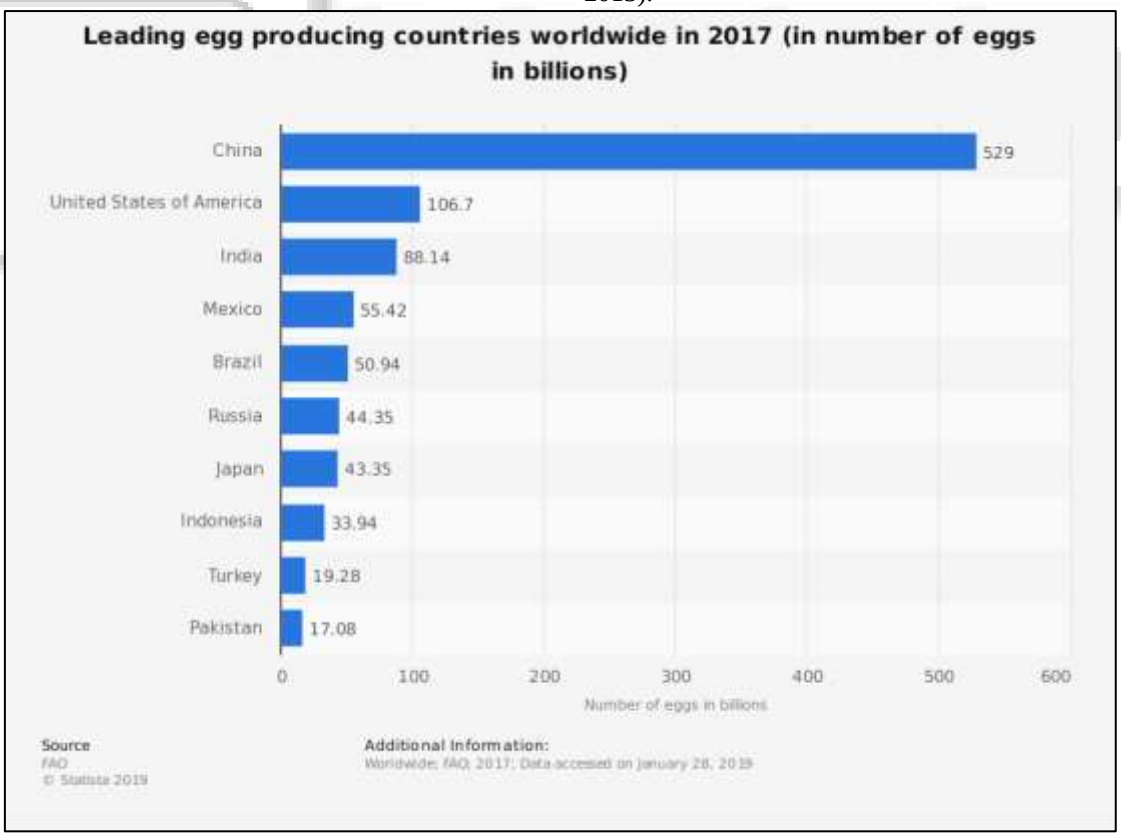


Fig. 1: Leading egg producing countries in 2017 (Statista 2017)

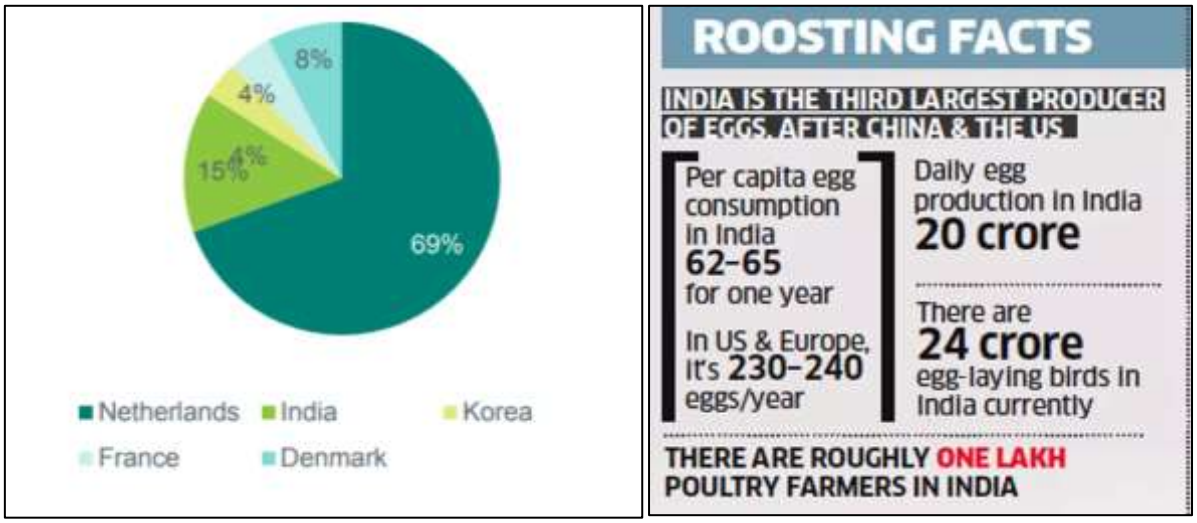


Fig. 2: Egg production in India (Statista 2017)

II. USES OF WASTE EGGSHELL

Researchers have found some uses of eggshell. A brief introduction to these finding are given below.

A. Calcination

Almost all of the eggshell application involved calcination of eggshell first to produce calcium oxide. Several authors in their independent studies reported the calcination variables as specific to their application.

Preparation of calcium oxide from chicken egg shells result showed that the optimum temperature for preparation of CaO from chicken eggshell is 900° C. Same result given independently by (Tangboriboon, Kunanuruksapong and Sirivat 2012) but he also reported purity of calcium oxide obtained to be 99.06%. (Mohadi, Anggraini, et al. 2016)

Characterization of CaO prepared from chicken eggshell using XRD, FT-IR spectrophotometer and BET analysis suggests that at 800° C only few $CaCO_3$ converted into CaO and hence 800° C is not a right choice for calcination (Mohadi, Anggraini, et al. 2016)

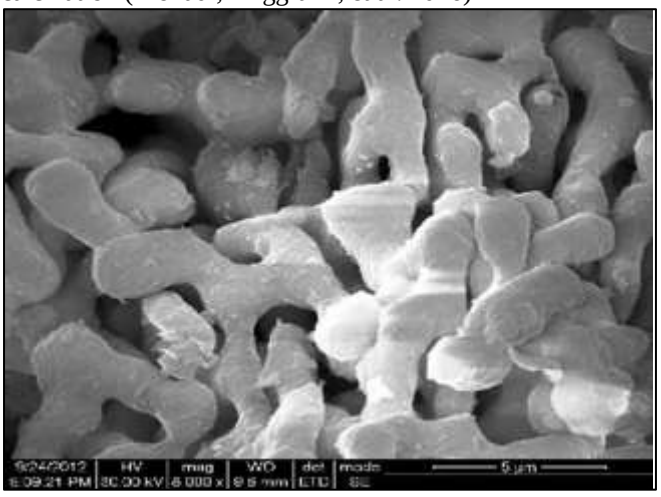


Fig. 3: SEM image of calcined egg shell at 900° C (Niju, et al. 2014)

Sample	Compound	2θ				
JCPDS data	CaO	32.2°	37.3°	58.3°	64.1°	67.3°
	$CaCO_3$	29.4°	39.4°	43.2°	47.4°	48.5°
	$Ca(OH)_2$	28.6°	34.1°	47.1°	50.8°	-
1000 °C	CaO	32.3°	37.5°	54 °	64.3°	67.5°
	$CaCO_3$	-	-	-	47.2°	48.4°
	$Ca(OH)_2$	28.8°	34.2°	47.2°	50.9°	-
900 °C	CaO	32.3°	37.4°	53.9°	64.2°	67.5°
	$CaCO_3$	29.1°	-	-	-	48.4°
	$Ca(OH)_2$	28.8°	34.1°	-	50.9°	-
800 °C	CaO	32.3°	37.5°	-	64.3°	67.3°
	$CaCO_3$	29.5°	39.5°	43.3°	47.3°	48.6°
	$Ca(OH)_2$	28.8°	34.2°	-	50.9°	-
700 °C	CaO	-	-	-	-	-
	$CaCO_3$	29.5°	39.5°	43.3°	47.3°	48.6°
	$Ca(OH)_2$	-	-	-	-	-
600 °C	CaO	-	-	-	-	-
	$CaCO_3$	29.7°	39.7°	43°	47.8°	48.8°
	$Ca(OH)_2$	-	-	-	-	-
Eggshell before calcinated	CaO	-	-	-	-	-
	$CaCO_3$	29.5°	39.5°	43.3°	47.6°	48.6°
	$Ca(OH)_2$	-	-	-	-	-

Table 1: XRD Data of CaO, $CaCO_3$ and $Ca(OH)_2$ from JCPDS and Metal Oxide CaO from Chicken Eggshell Calcinated at Various Temperatures (Mohadi, Anggraini, et al., Preparation Calcium Oxide From Chicken Eggshells 2016)

B. Utilization as Catalyst

Eggshells has been successfully utilized as catlysts for different purposes like biodiesel and gasification.

1) Biodiesel Synthesis

Transesterification of triglycerides with methanol using homogeneous catalyst has been used to synthesis of Biodiesel however it poses catalyst separation problem which requires intensive search for heterogeneous catalysts.

Calcined eggshells has been successfully used as heterogeneous catalyst for biodiesel production and conversion obtained was 94.52% whereas for commercial catalyst it was 67.67% (Niju, et al. 2014)

The optimum ratio of methanol to oil was 4:1, 2wt% catalyst, and a 65° C reaction temperature. The reaction temperature being set by the boiling temperature of methanol at 65° C (Wong and Ang 2018)

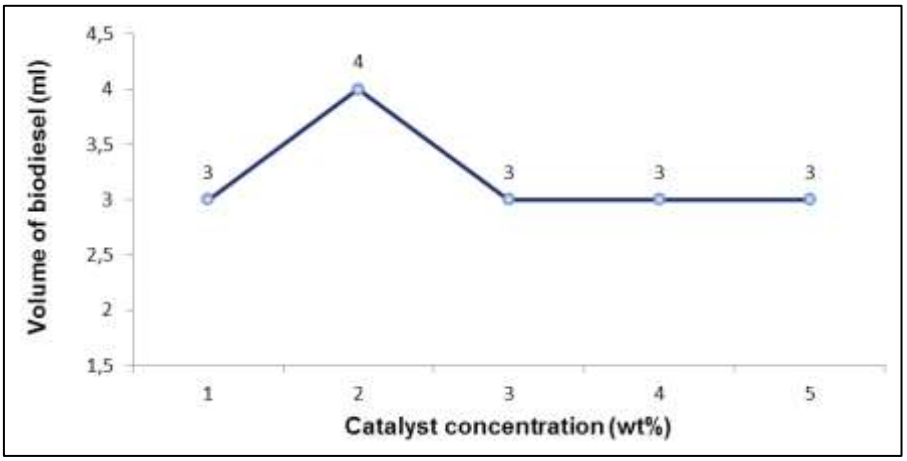


Fig. 4: the effect of different catalyst concentrations on the transesterification of used cooking oil. Conditions: reaction time of 2 hours, reaction temperature of 65°C, methanol to oil ratio 4:1. (Wong and Ang 2018)

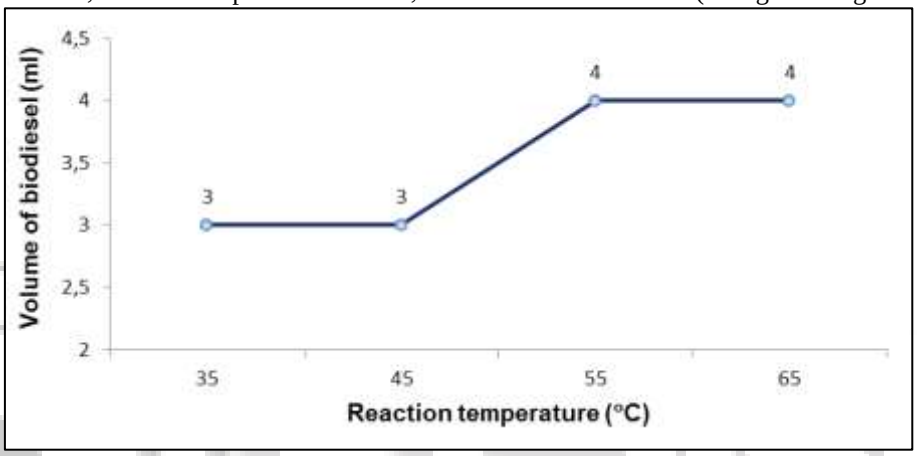


Fig. 5: The effect of different temperatures on the transesterification of used cooking oil. Conditions: reaction time of 2 hours, methanol to oil ratio 4:1, catalyst weight 2wt%. (Wong and Ang 2018)

The production of biodiesel from soybean oil using both mineral calcium oxide as well as calcined eggshell calcium oxide and then its XRF and XRD spectrometer analysis of both type of catalysts. The complete data on biodiesel yield against varying methanol/oil ratio, temperature for a fixed stirring rate at 450 rpm is below. (Ayodeji, et al. 2018)

XRF spectrometry of the conventional CaO, Uncalcined and calcined eggshell catalysts.

Compound	Chemical Composition of the Catalyst (wt%)		
	Conventional CaO	Uncalcined Eggshells	Calcined Eggshells
MgO	–	0.531	0.544
Al_2O_3	–	0.353	0.255
SiO_2	–	1.504	1.266
P_2O_5	–	0.283	0.316
SO_3	–	0.559	0.123
K_2O	–	0.118	0.103
CaO	99.230	96.131	97.080
Mn_2O_3	–	0.003	0.000
Fe_2O_3	0.110	0.017	0.013
SrO	0.120	0.305	0.263
ZrO_2	0.010	–	–
BaO	0.240	–	–
La_2O_3	0.130	–	–
CeO_2	0.040	–	–
Pr_2O_3	0.040	–	–
Yb_2O_3	0.050	–	–

Table 2: (Ayodeji, et al. 2018)

2) Hydrogen/Syngas Synthesis

Eggshells has been successfully utilized for Hydrogen/syngas synthesis. The reults of these research is summarized below.

Experimental design and biodiesel yields obtained from the transesterification process.

Rxn time (h)	Methanol/Oil (mole ratio)	Catalyst conc. (wt/wt%)	% Biodiesel yield (using CaO catalyst)	% Biodiesel yield (using eggshell catalyst)
2	8	3	88	85
3	14	4	94	91
1	8	4	86	83
1	11	3	82	81
2	11	4	83	82
2	8	5	84	84
2	14	5	87	85
3	11	5	82	80
3	8	4	91	89
2	11	4	82	81
1	14	4	89	86
1	11	5	80	80
2	14	3	89	87
2	11	4	83	82
3	11	3	84	81

Table 3: (Ayodeji, et al. 2018)

The calcined eggshell catalyst suppressed CO_2 production during gasification enhancing hydrogen production. (Taufiq-Yap, et al. 2013)

Comparison of the raw coal gasification without catalyst and found that eggshell catalytic gasification yielded greater syngas and reactivity. The gas release rates of eggshell catalyzed gasification at 800° C to be even greater than those of non-catalytic gasification at 900° Reported highest yield of H_2 is 1.24 mon/mol-C , 80% higher than raw coal gasification and CO yield was 0.34 mol/mol-C which is 31% higher than raw coal gasification. (Fan, et al. 2017)

C. Activated carbon

Activated carbon was createdfrom eggshell without removing the membrane. (Giraldo and Moreno-Piraján 2014)

Activated carbon was prepared from eggshell using sodium acetate and zinc at 400-600° C. The carbonized or activated products were characterized by SEM-EDS, TEM, FTIR and XRD. (Mopoung and Jitchaijaroenkul 2017)

1) CO-MO Catlyst Preparation

Prepared CO-MO catalyst using activated carbon prepared from eggshell by leaching it with HNO_3. This catalyst was shown effective for hydrogenation of methyl orange. (Ogunlaja, et al. 2010)

D. Wastewater Treatment

Complete removal of cd as well as 99% removal of cr from wastewater when treated for 10 min using calcined eggshells was found. (Heung Jai, et al. 2007)

Egg shell is found to be more effective for absorption of Cu (II) and Cd (II) from aqueous solution with maximum absorption capacity of 8.4 mg/g and 7.01 mg/g (Ziad Tark Abd Ali 2016)

Eggshell is used as an additive to precipitate divalent lead cations [Pb(II)] from aqueous solutions in the pH range of 2–5. Owing to high $CaCO_3$ content, eggshell precipitates Pb(II) to form lead carbonates, which will then settle on the surface of eggshell. (Vijayaraghavan and Joshi 2013)

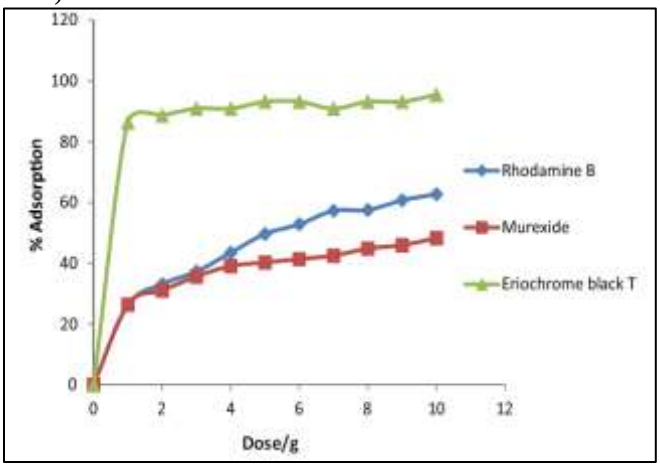
Investigation for removal of H_2S from water and found that for a 250 mg/l of pollutant the equilibrium loading for calcined eggshell was 2.1 mg/g. concluded that the chickens' eggshell are very useful green and economic adsorbents due to their availability and absence of any toxic and hazardous constituents' elements from all adsorbents the physically modified grounded chicken eggshell. Therefore, very suitable for the removal of H2S from wasters with the calcinate modified been the most suitable then the activate carbon modified adsorbent. (Habeeb, Yasin and Danhassan 2014)

The activate carbon prepared for adsorption of phenol from aqueous solution. Found the adsorbent to have high capacity and was easily available and it could remove copper from industrial effluents also. The adsorption process could remove phenol with capacity of 192mg/g and showed best fit for Langmuir model. (Giraldo and Moreno-Piraján 2014)

E. DYE Removal

Rhodamine B, Eriochrome black T and Murexide dyes are confirmed to be absorbed as the monolayer on calcined egg shell with maximum capacity of 2 mg/L (Borhade and Kale 2017)

Found uncalcined eggshell to remove 78% of methylene blue from dye solution. (Ngadi, Ee and Yusoff 2013)



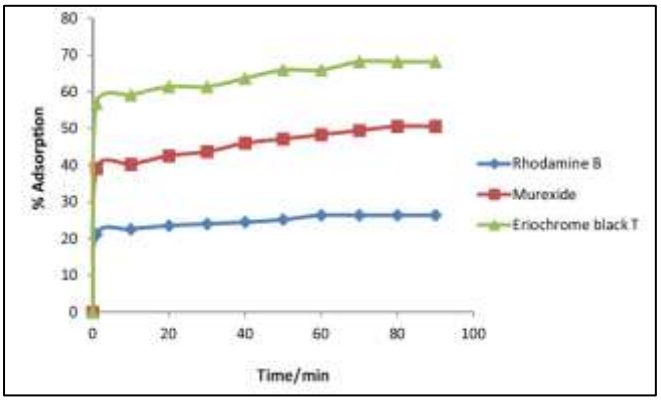


Fig. 6: Effect of absorption on dose and contact time (Borhade and Kale 2017)

F. Fertilizer

Use of eggshell powder as a fertilizer for Tomato plant and reported its application as fertilizer for blossom-end-root (BER) plants (Gaonkar and Chakraborty 2016)

Sr. No.	Sample	Parameter	Reading
1.	Without Eggshell Soil	pH	5
2.	10% Eggshell Soil	pH	87
3.	15% Eggshell Soil	pH	8.17

Table 4: Effect of eggshell on PH of soil

Day	Without eggshell	10% eggshell Powder	15% eggshell powder
7 Day	14.5	15	13
14 Day	15	17	16
21 Day	15	17	16

Table 5: Fertilizer percentage in soil

G. SUPPLEMENTS

Calcium tablets made from eggshell powder is equivalent against CIPCAL-500 calcium supplement tablets. Extraction of chicken eggshell is much easier than oyster shell which is source of CIPCAL-500 tablets. (Gaonkar and Chakraborty 2016)

Claims eggshell calcium to be the best natural source of calcium and it is about 90% absorbable. (Feed | Mendeley n.d.)

Egg shell powder can be used to prepare calcium fortified bread, pizza, corn flour, breaded chicken without change in flavor with minute change in texture. (Brun, et al. 2013)

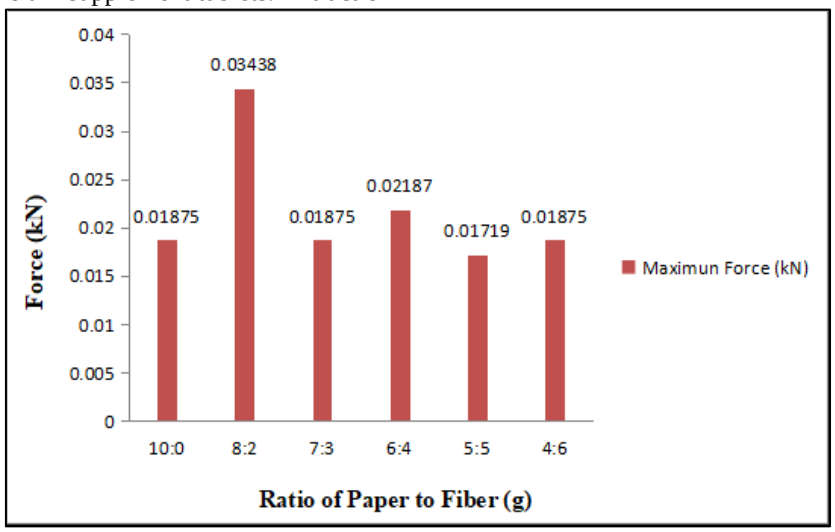
Cited the low level of toxic substances present in eggshell as the advantage over any other natural source of calcium. (Boron 2004)

Fig. 7: Chicken eggshell tablets (Gaonkar and Chakraborty 2016)

H. Paper

Eggshell was used as additional material to create recycled paper. Found 2:8 ratio of paper against pulp to be optimum giving maximum tensile strength as obtained by Universal Testing Machine. (Abdullah, et al. 2018)

He proposed to use this high tensile strength paper in food packing industries as substitute for polystyrene as the former is non-biodegradable and poses harmful effect to environment



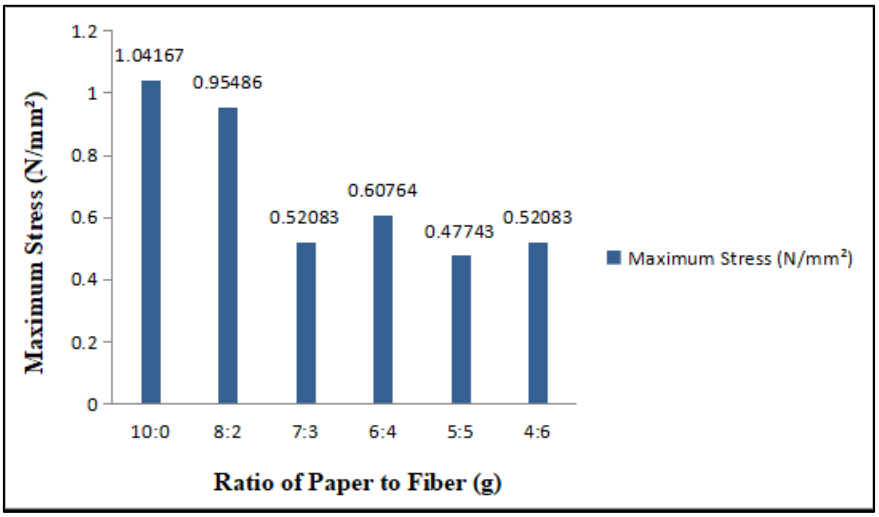


Fig. 8: Maximum force applied to composite paper of varying ratio (Abdullah, et al. 2018)

I. Cement

Production of cement using eggshells, calcium carbonate, sand, aluminium sulphate, ferrous sulphate and magnesium hydroxide. Further analysis of cement against ordinary Portland cement and discovered CaO to be much higher 5:1 in ordinary Portland cement to eggshell cement. The SiO_2 was 32% in eggshell cement as against 18.8% in Portland cement. (Bhardwaj n.d.)

Compound	Mass percentage of constituent's	
	Portland Cement	Eggshell Cement
CaO	63.4	38
SiO_2	18.8	32
Al_2O_3	5.0	16
Fe_2O_3	5.9	
SO_3	5.2	4.5
Na_2O	0.1	N/A
K_2O	0.3	
Insoluble substances	0.5	9.5

Table 6: Composition comparison of eggshell cement against Portland cement (Bhardwaj n.d.)

(Yerramala 2014) And (D. Gowsika 2014) independently investigated for compressive strength, Flexural strength, split tensile strength and absorptivity using varying composition of eggshell powder, saw dust, fly ash, and micro silica. (D. Gowsika 2014) Reported a sharp decrease in compressive strength when use of eggshell was more than 5%. Both of them independently concluded 5% uses of eggshell best for compressive strength.

Physical and chemical properties of cement using varying composition of the ingredients and emphasis was given on Micro silica uses (D. Gowsika 2014)

Study on more broad in terms of properties like absorption, sorptivity at varying composition and well-presented charts and figures in his research paper (Yerramala 2014)

J. Energy Storage

Study on electrochemical properties of calcined eggshell and fabricated a symmetrical super capacitor. They found that raw eggshell could be used as cathode and calcined eggshell at 900°C could be used as Anode. This marks eggshell usable as both anode and cathode. A discharge capacitance of 10 F/g was found with CaCO3 cathode but an excellent capacitance value of 47.5 F /g was shown by CaO anode. Both positive and negative regions the CaO electrode, exhibited 55 F/g at a current density of 0.15 A /g and even after 1000 cycles retention was nearly 100%. (Minakshi, et al. 2019)

III. Review Summary

Usage Type	Authors	Reviewers Findings/Comments
Coal Gasification	Shumin Fan, Li-Hua Xu, Tae-jin Kang, Hyung-Taek Kim (Fan, et al. 2017)	This paper seems to be complete with through discussion of kinetic models and constants. Currently India meets its hydrogen demand via recovery of refinery off gases which presents substantial higher cost of crude and recovery costs. Since India is abundant in coal reserves we may partially meet our demands by working upon the works of the Authors
Calcination	Risfidian Mohadi, Kiki Anggraini, Fahma Riyanti, Aldes Lesbani N.Tangboriboon, R. Kunaruksapong, A. Sirivat (Mohadi, Anggraini, et al.	Both the Authors got same calcination temperatures independently. Which proves authenticity of results

	2016) (Mohadi, Anggraini, et al. 2016)	
Cement Substitute	Amarnath Yerramala (D. Gowsika 2014) D. Gowsika, S.Sarankokila, K.Sargunan(Yerramala, 2014 (D. Gowsika 2014) Samarth Bhardwaj (Bhardwaj n.d.)	Cement is an energy intensive industry where about 99% of energy goes to crushing and grinding (Unit Operations, Mccabe Smith) However none of the Authors discussed this aspect when using egg shell as substitute. Papers presented therefore lacked discussions of energy requirements, crushing and preparation of egg shell as particles, grinder selection.
Paper Making	Mohammad Abdullah, Zaki Sukor, azmi Roslan (Abdullah, et al. 2018)	Though Authors have suggested egg shell paper as packing material but it lacked cost data and most industry will most likely adopt the alternates when it is economically feasible.
Biodiesel Production	S. Niju, K.M. Meera S. Begum, N. Anantharaman*** (Ayodeji, et al. 2018) Y.C. Wong, R.X. Ang**** (Wong and Ang 2018)	Cooking causes decomposition of oil and it is more severe during deep frying process. ***Didn't mention anything about oil treatment/purification before transesterification. There was ambiguity whether the conversion data was about oil or the triglycerides obtained from the oil All Authors focused on the discussions of conversion, calcination and transesterification temperature and their characterization analysis. However none of the Authors discussed the potential use of their product. Heating or calorific value or blend ratio.
Fertilizer Manufacture	Madhavi Gaonkar, A.P. Chakraborty (Gaonkar and Chakraborty 2016)	Authors prepared different samples of soil using varying composition of eggshells and evaluated the fertilizer %, PH, calcium in soil. They found the fertilizer useful for tomato plant but they didn't show any data on the germination, growth rates and yield of the plant with or without fertilizer application
Wastewater Treatment	Several Authors (Heung Jai, et al. 2007) (Habeeb, Yasin and Danhassan 2014)	Calcined eggshells can be used for heavy metals removal. It may also be used for H_2S and phenol removal from aqueous water
Energy Storage	Manickam Minakshi, Stephen Higley, Christian Baur, David R. G. Mitchell, Maximilian Fichtner (Minakshi, et al. 2019)	New type of uses of eggshell as electrode and capacitor. But eggshells have weak strength and batteries are subjected to impact and compressive stress in vehicles and other uses proper fabrication shall be required.
Catalyst Prepartion	Several Authors (Niju, et al. 2014)	Catalysts derived from eggshells showed more activity than mineral catalyst when applied to coal gasification and transesterification of triglycerides.

IV. SCOPE FOR FUTURE WORK

Soda Ash	Eggshell may be used as a substitute for mineral $CaCO_3$ in Solvay's Process. No work has yet been done in this prospective
Magnesium & Magnesium Compounds	Calcium oxide derived from eggshells may be used for lime treatment of brine to produce magnesium compounds
Calcium Carbide	Carbonation of calcium oxide derived from eggshell has potential for production of calcium carbide which then may be used as an intermediate for Acetylene (C_2H_2) production.
Methanol	Calcination of eggshell produces CO_2. Which may be then hydrogenated to form methanol. This if successful shall eliminate the CO_2 greenhouse release during the calcination processes.
CO_2 Sequestration	Some authors have reported methods of CO_2 capture via gypsum and gypsum may be derived by chemical treatment of calcium oxide derived from eggshells

V. CONCLUSION

Egg is used as an ingredient for both main course and fast food and subsequently producing tons of waste shells.

The increasing population growth, energy demand, reducing available land and forest area demands for use of "Reduce & Reuse" component of 3 R's methods of environmental management.

Successful research work on use of eggshell in various application has already been done like in calcination, cement, and catalyst however their commercial utilization and mass conversion is required. However these conversion work might not economical, this suggests further research work to unexplored fields as mentioned in this paper like soda ash, methanol, calcium carbide.

Thou, even if the conversion/value extraction proves to be uneconomical we need to coin the term to waste elimination and continue production/application as our endeavor to waste reduction and landfill requirement reduction.

This review summarizes the potential uses of eggshells in various applications. Calcined eggshells are now widely used as catalysts as they have even greater surface area as compared to mineral calcium oxide. The alternative uses of eggshell shall reduce the landfill and shall prove to be an environmental friendly step.

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