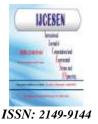


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Research Article

Possibility of Using Eggshell in Industry

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Eggshell Biowaste calcium sustainability The amount of eggshells (chicken, duck, ostrich, turkey, goose, and quail eggs) that go to waste every year around the world is frightening. Eggshells are foods with high nutritional value that should not go to waste. Chicken egg shells, which contain significant amounts of calcium and minerals, end up in landfills without further processing. As a by-product, it creates an important problem for producers both economically and ecologically. The purpose of this review is to summarize the results of various research articles on eggshell and to provide a reference for subsequent studies in this field and to expand the usage areas of eggshell. Numerous studies have shown that eggshells for nutritional and non-nutritional purposes not only reduces waste but also provides several economic benefits. A total of 83 sources were used in the review. In the compilation study, the studies on saving the eggshell from the waste position and transforming it into a material with high added value are summarized. It is a reference in studies to be conducted on alternative uses of eggshell.

1. Introduction

In proportion to the high consumption of eggs in the world, a huge amount of waste eggshell is generated. Eggshell has the opportunity to be reused in various fields due to its valuable organic and inorganic components, and studies on this subject have increased in recent years [1]. Eggshells reached a worldwide production of 7.8 million tons in 2018 [2]. While approximately 10% of eggshells can be used in the feed industry in Turkey, the remaining 90% goes to waste. This means that 120,000 tons of eggshells are thrown away unused annually [3]. It is important to evaluate waste materials in the production of some useful products and also to follow the 3R sustainability concept (reduce, reuse, and recycle) [4]. The main components of eggshell are carbonates, sulfates and phosphates of calcium and magnesium, and organic matter. The density of the egg shell is about 2.53 g/cm3. The main components of eggshell are calcium carbonate (94%) (380 mg Ca per gram), organic matter (4%), calcium phosphate 1% and magnesium carbonate (1%) [1;5] Ajala et al. [6], eggshell characterization Ca: 2300.33 \pm 3.80, Mg; 850.00 \pm 1.24, Na; 33.83 \pm 0.72, N; 17.06 ± 1.04 , Fe; 1.4 ± 0.03 , Zn; 0.99 ± 0.04 and Cu; as 0.063 ± 0.01 mg/L, 0.95 ± 0.89 moisture,

 45.29 ± 0.06 , ash; 1.40 ± 0.25 , crude protein; 0.37 ± 0.06 , lipid; 4.38 ± 0.32 , crude fiber; 47.63 ± 0.32 (%), carbohydrate and caloric value it was found to be 811 ± 12.71 (cal/g). The shell makes up 9-12 percent of the total weight of an egg and there are about 8,000 microscopic pores in the eggshell [7; 8]. Unlike commercial activated carbon, the pore size of eggshell is about 10 times higher. The measured pH values of eggshell and commercial activated carbon are 9.49 and 5.76, respectively [9].

Among the toxic gases released into the atmosphere during the decomposition process of the eggshell are ammonia (NH3) and hydrogen sulfide (H2S). Proper management of these wastes can reduce the risk of microbiological problems and the costs of their disposal into the environment and provide a better source of CaCO3 than limestone mined from nonrenewable sources [10].

After removing the membranes from egg shells, drying, grinding, and sieving, it is possible to obtain calcium carbonate. The purity of the obtained calcium carbonate is about 93.7% [11]. Ayawanna et al. [12] reported that glass-ceramic implants produced from eggshells have good chemical stability in the biological environment (in vitro). Biomaterials are materials used in the human body to perform the functions of living tissues that have

lost their function [13]. Egg shells are biodegradable and biocompatible. When used in biomedical materials, it is possible to obtain fillers or biologically neutral implants for bones and implants. Shells can be a valuable source of hydroxyapatite, especially in biomedical applications [14;15]. Yönetken et al. [16] produced composite material using a powder metallurgy technique with titanium, boron carbide, and eggshell powders, which is one of the indispensable elements of biomaterials, and demonstrated the alternative use of eggshell. Yönetken et al. [17] reported that the composite obtained by adding eggshell powder to Ti-Titaniumbased intermetallic compounds would be suitable for cutting or scratching tools used in the soil industry. Bilici et al. [18] and Yönetken et al. [19] reported that the hardness and ultrasonic wave speeds of the material produced increased by using certain amounts of eggshell powder in ceramic-metal composites. Jannet et al. [20] reported that an aluminum alloy (AA2024) composite containing 7% eggshell powder has properties that make it suitable for high tensile and compression applications in the automobile and aerospace industries. Akgürbüz and Uvsal [21] determined that the composites obtained by using eggshell, sewage sludge, and meerschaum provided sufficient mechanical properties (tensile strength, modulus of elasticity in tensile, bending strength, modulus of elasticity in bending, elongation at break and impact resistance). Skórczewska et al. [15] successfully produced poly(vinyl chloride) composites with significantly improved thermal stability by using waste eggshell as a natural filler in poly(vinyl chloride) composites [15].

In recent years, the human population and anthropogenic activities have increased and the need to develop an efficient gas sensor has arisen [22]. Ammonia (NH3) is a poisonous gas and its threshold concentration is 25 ppm [23]. Therefore, the development of an NH3 gas sensor is of great importance due to its various applications in many industries such as food, automotive, chemical, and pharmaceutical [22]. A simple, cost-effective, and novel chemical sensor for ammonia (NH3) gas detection has been developed from polyaniline (PANI)/eggshell (ES) composites. Norsham et al. [22] found that PANI/ES biocomposites exhibit excellent selectivity towards NH3 gas even in the presence of other gases such as acetone, ethanol, and hexane.

The pores in the eggshell indicate that they can also be used as effective adsorbents. In the last decade, eggshell has been used in wastewater treatment to remove organic and inorganic hazardous substances [5]. Heavy metals slow down metabolic reactions and are toxic to living organisms. Lee et al. [24] reported that calcined eggshell had the highest P adsorption in an aqueous solution. Researchers have stated that eggshell can be used as a P-binding absorbent and P-adsorbed calcined eggshells can be used as a potential P fertilizer, thus allowing P to circulate from water to soil [24]. One of the compounds used to absorb heavy metals in water is activated carbon. Activated carbon is the most widely used adsorbent and is used in drinking water treatment, solvent recovery, decolorization, metal mining processes, and various domestic applications. Activated carbon has disadvantages such as regeneration costs and less neutralization of acids [25; 26]. In eggshells, CaCO3 is a polar adsorbent that can be used not only as an absorber for heavy metals but also for neutralizing acids in water [27; 27]. The adsorption process takes place in the pores or in certain parts of the particles [29]. Eggshell can be developed in nanoporous form using sodium hypochlorite and hydrochloric acid [30]. Setiawan et al.[30] stated that duck egg shell had a better ability to absorb metal and neutralize acid than chicken and quail egg shells to reduce heavy metal and acidity in water. They also reported that the nanoporous eggshell had a better ability to absorb metals and neutralize acids compared to activated carbon. Ajala et al. [6] revealed the presence of the correct ratio of carbon and oxygen in the eggshell as a good alternative adsorbent for the removal of heavy metals and dyes from aqueous solutions.

Kamgang-Syapnjeu et al. [31] used eggshell in the production of a new ceramic membrane for the removal of pathogenic microorganisms in water and reported that the obtained ceramic membrane had the ability to retain 90% of E. coli bacteria and could be used for a preliminary water treatment. Ok et al. [32] reported that eggshell wastes can be used as an alternative to CaCO3 for the immobilization of heavy metals in polluted soils.

Recently, in removing heavy metal ions from industrial wastes, conventional metal ion removal methods have become ineffective in removing metal ions below 1 mg/kg concentration. Therefore, there was a need for other methods that could be effective at low concentrations [33]. It has been determined that eggshells have a biosorption capacity two times higher than activated carbon in the absorption of Cr (III) ions from aqueous solutions [34]. According to Kim et al. [9], eggshells are an effective bioadsorbent in removing heavy metals such as Pb2+, Cd2+, and Cr3+ from water. Muhaisen et al. [35] reported that cadmium was significantly adsorbed by eggshells. According to Somaia et al. [36], copper (II) ions in aqueous solutions can be removed from eggshell nanoparticles significantly more quickly than from untreated eggshell. Fluoride is a dangerous pollutant that must be removed during water treatment to provide water suitable for consumption [37]. Lee et al. [24] reported that eggshells are highly efficient adsorbents that can replace bone charcoal and activated alumina for fluoride removal in water as adsorbent, and that their adsorption capacity can be improved by heat treatment. The presence of various functional groups and high carbon content in eggshell make it a potential material for adsorbent to remove heavy metals and dyes in wastewater treatment [6]. Eggshell is a low-cost adsorbent to remove cobalt (CO+2) and lithium (Li+) ions from polluted soils [38].

Biodiesel is an environmentally friendly fuel and a good alternative to fossil fuels. Wei et al. [39] found that eggshell could be used as a 98% efficient solid catalyst in biodiesel synthesis, Chavan et al. [40], on the other hand, 90% efficiency in biodiesel production from Jatropha curcas oil using calcium oxide prepared from eggshell as a catalyst, Navajas et al. [41], on the other hand, obtained 100% yield from edible oil. Cho and Seo [42] reported that calcined quail egg shells showed better catalytic activity than chicken egg shells. Çetin [43], Rashid et al. [44] reported that eggshells, which are used as heterogeneous catalysts, reach high yields in the green fuel production reaction, are environmentally friendly, and reduce catalyst costs. Ayoola et al. [45] revealed that duck eggshells can lead to the production of high quantity and quality CaO catalysts. The pores in the eggshell have made it suitable for the use of heterogeneous catalysts in biodiesel production [6].

Stainless steel is one of the most widely used metals in many industries because it forms a protective film on its surface that prevents the metal from reacting with corrosive environments such as sulfuric acid [46]. Sanni et al. [46] revealed that eggshell powder can be used as a corrosion inhibitor on stainless steel. Ultraviolet (UV) light has harmful effects on synthetic polymers and biopolymers such as leather, hair, and wood [47]. These materials absorb UV, triggering photolytic and photooxidative reactions, causing photodegradation and permanent loss of beneficial physical, mechanical, and optical properties [48]. While many materials used today contain UV-protective additives, they are not always effective and durable, can be expensive, and their natural extraction or synthetic production can be harmful to the environment. Lippens et al. [48] used untreated egg shells to provide UV protection to polystyrene and nylon and compared them with TiO2 UV protective additive. The eggshell used in the study provided effective and durable protection to nylon and polystyrene. Nylon protected with eggshell showed slower photo-oxidative rates

compared to nylon protected with embedded TiO2 particles [48].

Due to the high CaCO3 content in eggshells, Binici et al. [49] found that the radiation permeability of composites with eggshell added significantly decreased. These composites could be used as radiation-scavenging flooring and partition walls in areas with radioactive devices.

Nath et al. [50] determined that the antibacterial activity of pure CaO obtained from egg shells at 900 °C against E. coli was quite effective in the preparation of antibacterial composites. The mechanism behind the antibacterial activity is due to the generation of superoxide (O2-) radicals from the CaO surface, reacting with hydrogen and oxygen to produce ROS (reactive oxygen species) in the system. ROS damaged the bacterial cell colony through several mechanisms [51; 50].

Cement production requires a significant amount of energy and produces a significant amount of CO2 [52; 53]. It was observed that the use of calcined eggshell powder at low replacement rates (5%) increased the compressive strength of cement-based materials [53; 54]. Teara et al. [55] reported that 35% fly ash and eggshell and cement substitute in concrete production improved the compressive strength and durability of concrete compared to conventional concrete, Hamada et al. [56], the eggshell powder could be used to improve concrete properties and reduce cement production, Bhartiya and Dubey [57] stated that 10% substituted egg instead of cement, Adogla et al. [58] reported that 30% of eggshell powder significantly improved the density, compressive strength and durability properties of laterite bricks. Since the eggshell has a specific gravity that varies between 0.85 and 2.66, which is lighter than cement, it is lighter than normal concrete and can be used in structures that require lightweight concrete. The different effects of eggshell powder on cement and concrete are due to surface morphology, CaO content and particle size [57].

In their study, Razzaq and Yousif [59] added eggshell powder at the rate of 3, 5, 7, 10 and 15% by weight of asphalt. As a consequence of the testing, the flash point improved with increased additive ratio despite a large loss in penetration value and a significant rise in viscosity. Temperature has strengthened the resistance to persistent deformation. It has been reported that the addition of eggshell to asphalt can be used as an asphalt binder material because it reduces high temperature deformation and rutting damage of asphalt pavement.

The stabilization of the soil is done to improve certain properties of the soil (plasticity index, gradation, etc.) by adding expensive additives such as lime, cement, and bitumen. Since the chemical composition of the eggshell powder is similar to that of lime, it can be a good alternative to industrial lime as a stabilizing material [60]. In a study using eggshell powder as a stabilizing material to increase the strength of floors, it was concluded that a floor containing 6% eggshell powder was the optimum combination. It was concluded that eggshell powder could improve the performance of clay in terms of plasticity index, compression properties, and strength aspects [60].

Vilarinho et al. [61] developed eco-ceramic wall tiles using bio-calcium carbonate from eggshell as a raw material. Eggshell (0, 25, 50, 75, and 100 wt%) was used instead of limestone in the study. Through the characterization of the samples, the properties of biocalcium carbonate with the same granulometry were within industrial limits. Therefore, the study has proven that limestone can be completely replaced by eggshell in the production of ecoceramic wall tiles. Reintroducing the eggshell to the ceramics industry contributes to a real industrial symbiosis as well as a circular economy [61].

Tutus et al. [62] in the study where precipitated calcium carbonate and eggshell calcium carbonate mixtures were used in the production of wallpaper, they reported that water-resistant opaque papers were obtained as a result of reducing the spaces between the fibers by using eggshells [62].

The use of pH modifiers in mining and related industries is very important because certain reactions require certain pH to occur. Also, without the required pH, some undesirable chemical reaction byproducts can occur which tend to make the atmosphere unsafe. The calcined eggshell increased the pH of a solution with a native pH of 9.71 to 12.44 [63]. Technologies used in acid mine drainage have many limitations in terms of high capital cost, large production, volumes of sludge operational sensitivity, and applicability for active treatment only [64; 65; 66; 67]. Calugari et al. [64] reported that thermally activated eggshells are effective in acid mine drainage neutralization and processing of heavy metals in acid mine drainage in cold climates, and can be used in active or passive solution methods, post-closure, and restoration, barrier and drainage processes. Choi and Lee [67] found that the combined use of calcined eggshells and microalgae to remove heavy metals from acid mine drainage (AMD) and simultaneously increase biomass productivity, removal percentage of Fe, Cu, Zn, Mn, As, and Cd from acid mine drainage wastewater is 99.47 to 100%.

Fernandes et al. [68] used eggshells in the production of foam glass and proved to be a good alternative to conventional foaming agents such as CaCO3, MgCO3, or SiC used in foam glass production. In addition, eggshells have advantages such as being a waste material, potentially cost-effective, contributing to the cleaning of the environment, and saving energy by releasing gas at low temperatures (700oC) [68].

Due to the fact that polyurethanes can have different properties depending on the type and composition of the components that make them up; They are valuable engineering materials used in many fields, especially in the automobile, adhesive, furniture, paint, construction, and cooling sectors [69]. Victor et al. [70] reported that eggshells have a higher flexibility and ability to resist deformation as a filler in the production of flexible polyurethane foam. These features are the most needed features in foam mattresses. Erdem et al.[69] reported that rigid polyurethane foams with 7% eggshell powder additives may be the most likely structure to be recommended for many applications. Egg shells have good strength and tensile strength to strengthen the mechanical stability of polymer matrices [70].

Minakshi et al. [71] reported that the eggshell can be used for energy storage, the shape of the electrochemical curves obtained in the study is similar to that of the commercially available activated carbon electrode, and the CaCO3 obtained from the eggshells can be used as an electrode in Liion capacitors. Abdi et al. [72] studied 1% and 2% eggshell powder, eggshell ash, acid-activated eggshell powder, acid-activated eggshell ash, and commercial bleaching earth for bleaching neutralized soybean oil. Due to its high efficiency in removing unwanted components (pigments, iron, copper, oxidation products, and free oil) and environmental protection against acid pollution, eggshell powder can be used as an adsorbent and can be considered as a good alternative for bleaching edible oils, according to the study [72]. The application of organic fertilizers provides valuable nutrients to the plants and not only improves the physical and chemical condition but also increases the crop yield and reduces the use of chemical fertilizers [73]. Eggshell is an enriched source of calcium, essential for plant growth [74]. In agriculture, potassium is called potash, which is used as fertilizer. The combination of potassium and calcium in eggshell can make it a suitable raw material for fertilizer production [6]. Biswas et al. [74] reported that the combination of eggshell, hydrolyzed chicken feather protein, and vermicompost (100:10:3 ratio) increased the agronomic parameters of the capsicum plant approximately four-fold compared to its chemical counterpart. In addition, the combination used increased the yield parameters of the crops by enriching the soil with both micro and macronutrients [75].

Eggshell and Chlorella Vulgaris algae increased the Ca content of tomatoes as well as minerals such as B, K, Mg, and Zn in soilless agriculture [76]. Holmes et al. [76] reported that ground eggshells proved to be an effective liming material in the region with acidic surface soil and were applicable for soil pH correction. Laohavisuti et al. [77] obtained low and high-purity calcium carbonate from eggshell to produce dicalcium phosphate, monocalcium phosphate, and tricalcium phosphate for the fertilizer, mineral feed, and food additive industries. The study showed that eggshell can be used in the fertilizer, animal feed and food additive industries that require very large quantities [77]. It contains high amounts of bioavailable calcium, such as egg shells, shellfish, bones, and fish scales. The eggshell is easy to separate from the protein part, namely the yolk and albumen. Considering the separation of muscle and other protein masses from the bones and the removal of the proteinaceous portion from fish scales and shellfish shells, it takes more time and less efficient separation processes [78]. Although oyster shells contain some toxic elements such as Pb, Al, Hg and Cd, egg shells do not contain such toxic elements. Eggshell calcium has good solubility [79] and CaCO3 extraction from eggshells is relatively inexpensive, physically feasible, and efficient compared to other sources [78]. In rat studies, calcium from eggshell powder was discovered to have a high bioavailability compared to commercially available CaCO3 [79]. Zerek [81] reported that the ash content and calcium mineral content of cookies containing 3%, 6%, and 9% eggshell powder increased with increasing eggshell powder. As a result of the microbiological evaluation of sterilized egg shells, total mesophilic aerobic bacteria (TMAB), mold-yeast, Salmonella spp. were not detected. Hassan [82] reported a significant difference in calcium (Ca) content and Ca bioavailability in biscuits made from wheat flour with eggshell powder additives. On the other hand, Perez et al. [83] developed functional yogurt that was purified from pathogenic microorganisms from micro powders obtained from egg shells and suitable for human consumption. The calcium content of eggshell-added yogurt was found to be twice as much as normal yogurt. Eggshells are an inexpensive and readily available source of calcium that can be prepared at home, provided that appropriate precautions are taken (Salmonella), leaving no major contaminants behind.

2. Conclusions

Eggshell waste generation is growing rapidly and will continue to increase. Instead of destroying the eggshells, which create an important environmental

problem, it should be aimed to use them in appropriate areas, and at the same time to improve their reuse capabilities. The positive results obtained from the studies support the perspective of the zero waste operation to produce material recycling, waste management, sustainable development, and valueadded products in the future. When waste is evaluated, it will have an economic contribution as it will be transformed into high value-added materials. Literature studies have shown that eggshells are an important bio-waste in providing the transition from garbage to value-added products. By organizing the collection of eggshells from industrial egg uses, food factories, poultry farms, and bakeries, they can become fully usable raw materials for many of the above-mentioned applications.

Author Statements:

- **Ethical approval:** The conducted research is not related to either human or animal use.
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