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The Potential of Calcium Carbonate from Eggshell as a Pharmaceutical Raw Material in Indonesia

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Abstract

Utilization of Eggshell Waste has the potential as a pharmaceutical raw material. The main content of Eggshell (ES) besides containing macro minerals Calcium Carbonate 14.30% - 36.62%. Micro minerals I 0.00034% and Mo 0.00014%. Boiling at 80°C for 15 minutes and soaking with H₃PO₄ 4-5%. The results of proximate analysis and Scanning Electron Microscope (SEM) showed that the crude protein content of hatchery eggshell waste was 4.85%. The results of Energy Dispersive X-ray (EDX) analysis contained Ca 31.81%, Mg 2.93%, P 4.48%. While bakery waste crude protein 4.84%, Ca 36.62%, Mg not detected, P 0.03%. The difference in results is due to differences in the samples tested taken from several locations and types of chickens that produce EM. The high Calcium Carbonate content of 14.30-36.62% in ES has the potential to be used as a supplementary source of animal calcium in the pharmaceutical industry.

Keywords: Eggshell, Calcium Carbonate, Pharmaceutical Industries.

1. Introduction

The Presidential Regulation Number 59 of 2017 represents the Indonesian government's commitment to implementing the Sustainable Development Goals (SDGs). Environmentally friendly industrial innovation, including the pharmaceutical industry, is a key pillar in achieving sustainability and health for all levels of society. Waste is a global problem; utilizing waste for health supplements represents a breakthrough in improving public health, while simultaneously preserving the environment and supporting the SDGs.

Eggshells are abundant animal waste. Indonesia's free-range chicken egg production in 2022 reached 375,253 tons, an increase of 2.74% and 4.45% compared to 2021 and 2020, respectively. This increase in egg production is also driven by the increase in the free-range chicken population, which reached 7,709,715 over the past year ^[1]. The pharmaceutical industry plays a crucial role in providing quality calcium supplements. Calcium raw materials from ES have the potential to be used as pharmaceutical raw materials.

Animal waste-based pharmaceutical raw materials have not been commercially used with quality that meets the Indonesian Pharmacopoeia. This prompted the authors to explore the potential of calcium from ES as a pharmaceutical raw material. ES contains many mineral nutrients such as calcium, potassium, magnesium, iodine, molybdenum, and chloride ^[2].

The morphology of ES is that it is part of an egg, consisting largely of a single fibre and divided into three layers: the outer, inner, and specific, limited membrane ^[3]. In addition to fibre, it also contains high amounts of essential nutrients ^[4]. The characterization of ES waste from hatcheries has not been widely studied. The use of SEM can determine sample characteristics, including topography and composition ^[5]. To determine the elements in the sample, further testing, namely EDX, is carried out, an analysis method that provides rapid qualitative and quantitative data ^[6]. ES comes from several sources, including bakeries industries (commercial) and hatcheries (broilers).

2. Materials and Methods

EM was obtained from several sources, including bakery (commercial laying hens), hatchery (broiler) free-range chickens, and ducks obtained from the KSPM GURAMI Semanu, Playen, Gunungkidul Regency, and Turi, Berbah, Sleman, Yogyakarta. Laboratory analysis was conducted at the Indonesian Institute of Sciences (LIPI) Natural Materials Technology Centre. Eggshell material came from eggshell waste collected from the PT. Malindo hatchery. In addition, 4-5% H₃PO₄ chemicals were required for proximate analysis and SEM (Hitachi SU 3500) and EDX (EDAX®) preparation. The ion sputter apparatus MC1000 (Hitachi Corp.) was used [Suryani 7]. To increase conductivity [Liu, 8] and prevent

sample discharge phenomena (Endo et al. 9). The materials used for proximate analysis were according to the AOAC (Association of Official Analytical Chemists) method (AOAC, 2005). The equipment used included a 100 kg capacity digital scale, a stove, a bucket, a boiling pan, and a set of proximate analysis tools.

Mineral Determination with XRF

Minerals were determined by XRF using an XRF spectrophotometer (@Malvern Panalytical's), Epsilon 4 50 kV, 3 mA, using the Omnic Standard helium analysis method. The collagen sample was placed in a sample holder, then flattened and inserted into a diffractometer operated with 50 kV X-ray radiation.

3. Research Methodology

To assess the potential of calcium carbonate produced from EM, several journals were reviewed, sourced from databases such as Google Scholar, PubMed, Science Direct, and Wiley, covering various topics related to EM, ranging from the collection and preparation of ES waste (e.g., washing, drying, flouring, calcination), to the analytical treatment process, calcium, and other minerals.

4. Results and Discussion

Table 1: Analysis Result proximate eggshell waste

Parameter	Egg Shell Waste in %			
	Local Chicken	Laying Hens	Duck	Hatchery Native Chicken
Dry matter	98,35	98,58	95,95	99,19
Ash	91,05	90,91	88,59	91,27
crude fat	0,07	0,10	0,07	0,29
Crude fiber	0,84	0,59	1,39	0,79
Crude protein	6,03	6,99	5,77	7,31
Nitrogen-free extract	0,36	1,41	0,12	0,34

Source: Setiyawan et al 2021

On Table 1 of Setiyawan *et al.*' research shows that chicken and duck ES waste was obtained from Playen District, Gunungkidul Regency, Yogyakarta. ES derived from free-range chicken hatchery waste and waste has different mineral and protein contents. ES from hatchery waste is generally higher in protein and collagen, while ES from duck waste has the lowest. However, the highest fibre content is found in duck ES waste, as ducks forage in rivers or rice fields, where there are many fibrous plants.

The nitrogen-free eggshell extract (NFEE) is intended to focus on utilizing its main content, namely calcium carbonate (CaCO₃), which makes up approximately 97% of the total eggshell. NFEE eggshell waste from laying hens has the highest calcium carbonate content, due to the mineral-rich feed provided regularly and in measured doses. For ES waste from native chickens and ES waste from hatching native chickens, the calcium content is almost the same, while for ES waste from ducks, it is the lowest because the ducks in the research area are released to look for a variety of food and do not eat feed that has been given complete nutrition.

Table 2: ES Waste Composition using EDX (EDAX®)

Composition	Source of ES Waste %	
	Hatchery	Bakery
C (Karbon)	-	9,91
N (Nitrogen)	12,5	-
O (Oksigen)	47,76	53,23
Mg (Magnesium)	2,93	-
Si (Silika)	0,28	0,2
P (fosfor)	4,48	0,03
S (Sulfur)	0,24	-
Ca (Kalsium)	31,81	36,62

Source: Setiyawan et al 2021

In the research of Setiyawan *et al.*, table 2 above, ES waste materials taken from Semanu District, Gunungkidul Regency, Yogyakarta, came from two sources, namely chicken ES from hatching and from the bakery industry. The calcium content of ES waste from the bakery industry was the highest at 36.62% compared to ES waste from hatching at 31.81%. Meanwhile, the magnesium and phosphorus content in ES from hatching chickens were the highest at 2.93% and 4.48%, while ES chickens from the bakery industry had magnesium undetectable and phosphorus at 0.03%.

Table 3: Results of XRF analysis of ES waste

Parameter	EM Waste			
	Native chicken	Lying Hens	Duck	Hatchery native chicken
Ca (%)	25,05	27,73	26,21	22,52
Mg (%)	0,72	0,72	0,77	0,56
P (%)	0,23	0,21	0,24	0,19
Na (%)	0,11	0,22	td	td
Si (ppm)	70,2	80,3	35,6	144,1
Cl (ppm)	35,9	24,9	td	td

Td: Not detected

Source: Setiyawan et al 2021

Setiyawan *et al.* also conducted another study, the results of the analysis are listed in Table 3 and the source of ES for free-range chickens, layers, ducks, and hatching chickens, obtained from Playen District, Gunungkidul Regency, Yogyakarta. What is interesting about this study is that the ES waste comes from more varied poultry and the mineral content examined is not only macro elements but also micro elements. In contrast to ES waste from other areas, ES waste from Playen District has the highest calcium content in laying hens at 27.73%, while the lowest is from hatching chicken ES at 22.52%. The magnesium content of ES for free-range chickens, layers, and ducks is 0.72-0.77%, while for ES for free-range chickens from hatching is smaller at 0.56%. The silicon content of ES for free-range chickens from hatching is 144.1 ppm, the highest among other EM, and the lowest is for duck ES at 35.6 ppm.

Table 4: Highest mineral content in ES from various types of poultry and various regions in Gunungkidul Regency, Yogyakarta.

Minerals	Content	Location of EM	Origin EM	Researcher
Calcium	36.62%	Subdistric Semanu Regency Gunungkidul	bakery	Setiyawan <i>et al</i>
Magnesium	2.93%	Subdistric Semanu Regency Gunungkidul	Hatchery Native chicken	Setiyawan <i>et al</i>
Phospor	4.48%	Subdistric Semanu Regency Gunungkidul	Hatchery Native chicken	Setiyawan <i>et al</i>
Silicon	144.1 ppm	Subdistric Playen Regency Gunungkidul	Hatchery Native chicken	Setiyawan <i>et al</i>

Source: Data collected from various sources

The data shows the highest mineral content in several areas of Gunungkidul Regency. The highest calcium content in Gunungkidul Regency can be found in ES produced by the bakery industry, while other minerals, such as magnesium, phosphorus, and silicon, come from ES produced by native chicken hatcheries.

5. Discussion

The research results of Setiyawan *et al.*, which combined ES from free-range chickens, incubators, laying hens, and ducks, were very helpful in identifying mineral content for use in the development of mineral supplement products in the pharmaceutical industry that are environmentally friendly and support the SDGs. Chicken eggs are better used as calcium supplements for osteoporosis sufferers because they contain a higher Mg/Ca ratio (0.026 ± 0.005) compared to duck eggshells ($p < 0.05$). (Tyas *et al.* 2018).

The limited ES sampling in Gunungkidul Regency requires expansion to East Java, Central Java, and West Java, as East Java is the largest chicken egg production center in Indonesia. The highest calcium carbonate content in ES is 36.62%.

The comparison of commercial calcium carbonate derived from soil and chemical reactions with natural calcium carbonate from waste is as follows:

Table 5: Calcium Carbonate content requirements according to Monograph

No	Content in % Calcium	Indonesia	China	Europe	USA
1	Natural Calcium Carbonate (ES)	36.62	-	-	-
2	Ground Calcium Carbonate (GCC)	40.00	40	40	40
3	Precipitated Calcium Carbonate (PCC)	40.00	40	40	40

Source: Compiled from various sources:

Indonesian Pharmacopoeia 6th Edition (PI), China Pharmacopoeia (CP), European Pharmacopoeia (Ph. Eur.), United States Pharmacopoeia (USP).

From the comparison of the calcium content of various types of calcium carbonate and taking into account the requirements of the Indonesian Pharmacopoeia, such as:

- Calcium carbonate content contains not less than 98.5% and not more than 100.5% (CaCO_3). The content is calculated based on the dried substance.
- Description:** White or colourless crystalline powder; odourless; tasteless. Stable in air.
- Solubility:** Practically insoluble in water. Its solubility is increased by the presence of ammonium salts or carbon dioxide. Insoluble in ethanol. Soluble in dilute acetic acid, dilute hydrochloric acid, and dilute nitric acid, producing foaming (releasing CO_2).
- Identification:** Provides a calcium reaction (powder dissolved in acid and tested by adding ammonium oxalate solution will form a white precipitate that is insoluble in

dilute acetic acid).

- Heavy Metal Impurity Limits:** Meets standards for pharmaceutical use (antacids/supplements). Barium: Meets test requirements (generally filtered and tested with sulfuric acid). Magnesium and Alkali Metals: Not more than 1.0%. Acid-Insoluble Substances: Not more than 0.2%.
- Containers and Storage:** With the above requirements, calcium carbonate derived from ES needs to be evaluated by the Food and Drug Authority (BPOM) so that ES calcium carbonate can be used as a natural source of calcium in Indonesia, both in food and pharmaceutical preparations.

6. Conclusions

EM has the potential to be a source of calcium. By utilizing ES waste, we can reduce waste and simultaneously create a new mineral source that is not derived from earth minerals or chemical reactions, but from animal sources, supporting the SDGs.

7. Recommendations

Future research should analyse the chondroitin sulphate and hyaluronic acid, Collagen content of ES, with Eggshell Membrane sources from native chicken eggs, laying hens, hatcheries, and ducks. To The government, specifically the Ministry of Health and the Food and Drug Administration (BPOM), can evaluate the potential of waste as a natural calcium ingredient for inclusion in the Indonesian Pharmacopoeia by providing calcium content requirement range, specifically for natural products like ES, with a range of 35-40%. This allows ES waste to be utilized and reduce waste in Indonesia.

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