SYSTEMATIC REVIEW

Application of Eggshell Membrane as an Adsorbent for Pollutants Removal; A Systematic Review

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ABSTRACT

Introduction: Introduction: The eggshell membrane (ESM) has gained attention for its utility as an adsorbent in various pollutant removal. **Material & Methods:** This study systematically reviewed 17 articles published from 2011 to 2021 that use ESM as adsorbent in the wastewater treatment. **Result and Discussion:** The review found that both modified and unmodified ESM-based adsorbents are promising for removing various types of contaminants. The pollutants targeted by the studies were heavy metals, dyes, and metalloids. Most of the research indicated that the ideal temperature for adsorption was at room temperature, and a lower pH range was favourable. More than 52% of the articles found that the Freundlich isotherm model best fits the adsorption data, indicating that the process is multilayer and heterogeneous. **Conclusions:** ESM-based adsorbents hold potential for pollutant removal. However, additional research is required to explore the applicability of these adsorbents for addressing different categories of contaminants.

Keywords: Eggshell membrane; Adsorbents; Pollutants; Systematic literature review

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INTRODUCTION

Water pollution has been a great concern in most parts of the world, as water is essential for living sources and economic activities (1). The significant growth of the world population and increased industrial development have increased water demands, leading to water pollution. The world's population has expanded fourfold in the twentieth century, while water demand has climbed ninefold (2). Numerous countries are likely to suffer water shortages by 2025. Therefore, due to this situation, a highly stringent water quality standard has been introduced as a preventive and control measure (3). Water pollution happens due to the uncontrolled release of effluents with unknown pollutants content into water bodies, such as rivers and oceans. The pollution arises from a myriad of sources, including industrial wastewater, household sewage, stormwater runoff, septic tanks, and agricultural practices (4). Among these contributors, industries play a crucial role by releasing a diverse array of harmful substances into the aquatic ecosystem. These substances comprise toxic chemicals, both organic and inorganic matter, radioactive sludge, sulphur, asbestos, hazardous solvents, polychlorinated biphenyls, lead, mercury, nitrates, phosphates, acids, alkalies, dyes, pesticides, benzene, chlorobenzene, carbon tetrachloride, toluene, and volatile organic chemicals (4). Pollutants originating from industries have the potential to persist in the environment for an extended duration, leading to the bioaccumulation of these substances within the bodies of aquatic organisms (5). When these wastes are discharged into the water without undergoing sufficient treatment, they become highly hazardous for human consumption and other purposes. This pollutant can be linked to various illnesses, including anemia, low blood platelets, headaches, an increased risk of cancer, several skin conditions, etc. (4). Therefore, the need to extensively remove pollutants in the water is highly necessary and recommended.

In the last few decades, researchers have been trying out various pollutant removal methods in addressing this problem (6). Depending on its sources, the types of pollutants that exist in the bodies of water can be different. Researchers have been figuring out many methods, considering the type of pollutants to ensure high removal efficiency. Many different strategies have been used, such as reverse osmosis, coagulation, advanced oxidation, phytoremediation, and electrodialysis (7,8). Despite these methods being highly effective, there are many additional problems caused by these removal methods. Since almost all these methods are highly advanced technology, they require high energy consumption and costly reagents, increasing the cost needed to cover this process (8,9). Some of these techniques also pose additional problems, which produce by-products that require another removal method. As an alternative, adsorption has been an excellent method to remove various pollutants in the water. This is because adsorption is a highly effective process, very easy to handle, requires low operating costs, and does not release secondary by-products (10). Various adsorbents have been utilized in the adsorption process to study their capability and efficiency of adsorbing pollutants in the water.

The adsorption process is an environmental-friendly pollutants removal technique as many potential wastes can be used as adsorbents (11). Kitchen waste is commonly studied for its capability as an absorbent. Eggshells are a common waste material generated in huge quantities from the food industry, such as restaurants, bakeries, and poultries. The high production of eggshell waste in landfills has raised concerns among the authorities (12). Therefore, the utilization of eggshell membranes as an adsorbent can significantly help in reducing the amount of waste generated by the food industry. The eggshell membrane is primarily made up of protein fibers with numerous functional groups, which significantly helps in binding various heavy metals under certain experimental conditions (13,14). Despite many studies conducted to investigate the application of food waste as an adsorbent for pollutants removal, there is yet a lack of review papers done on this matter, especially on the application of eggshell membrane as an adsorbent. Reviewing the application of eggshell membrane for wastewater treatment is justified due to its potential as a sustainable and renewable resource, abundant availability as a byproduct of the egg processing industry, and inherent adsorption capacity for various contaminants in wastewater. Utilizing eggshell membrane offers a low-cost and environmentally friendly solution, reducing reliance on synthetic chemicals and contributing to the development of eco-friendly wastewater treatment methods. Furthermore, its biodegradability ensures minimal environmental impact after use. Investigating the effectiveness of eggshell membrane in wastewater treatment can lead to innovative, cost-effective, and eco-friendly approaches, addressing water pollution challenges and promoting sustainable practices in the realm of environmental engineering.

MATERIALS AND METHODS

Review Protocol

A systematic literature review approach has been selected to widen the understanding of the application of ESM as an adsorbent in removing water pollutants. This approach offers many benefits compared to the conventional method, as it is a systematic, transparent, and replicable method. According to Qazi et al. (15), a systematic literature review provides three common purposes, (a) To conduct an equitable evaluation of research articles on the desired topic using a precise and systematic method, (b) To assist in identifying research gaps for further improvements in the future research and (c) To summarize and provide background for new research activities. Many formal reviews coordinating protocols have been developed to maximize the quality of reviews for policy and practice and guarantee that they adhere to defined standards and procedures (16). The review protocol used for this systematic literature review was Reporting Standards for Systematic Evidence Syntheses, also known as ROSES. According to Mohamed Shaffril et al. (17), ROSES is designed to encourage researchers to provide sufficient information with accurate detail. Based on this protocol, the review process has three main steps, which are, Systematic Searching Strategy, which was divided into three parts, namely, identification, screening (inclusion and exclusion criteria) and eligibility, Quality Assessment, and Data Extraction and Analysis. This review covered all these steps, as shown in Figure I.

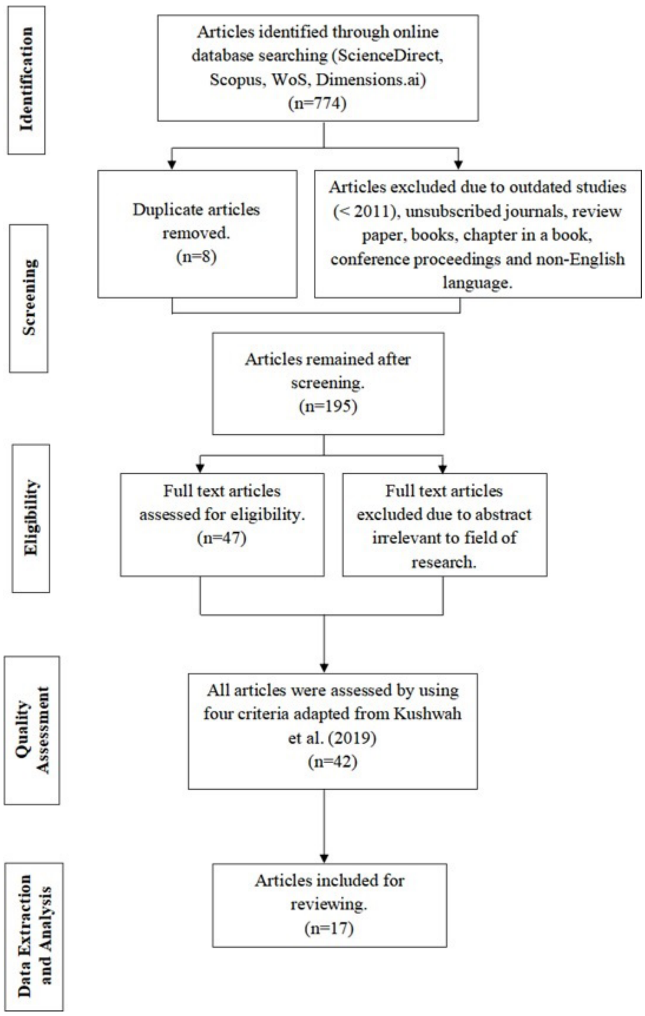

Figure 1 : Research flow diagram.

Resources

Four online databases were used in this review: ScienceDirect, Scopus, Dimensions.ai, and Web of Science (WoS). Researchers are recommended to use many online databases to increase the likelihood of obtaining more relevant articles (17). ScienceDirect is a database with over 2500 peer-reviewed journals covering many disciplines such as physical sciences and engineering, life sciences, health sciences, social sciences, and humanities. While Scopus is the world's largest abstracts and citation database that has over 75 million peer-reviewed journals. Another database used in this review is WoS. WoS is maintained by Clarivate Analytics which allows access to over 171 million records. In addition, these three databases were accessed through institutional subscription-based services for an advance and rigorous searching experience. In contrast, Dimensions.ai is a free online database that was also very helpful in the article searching process. This free online database has approximately 119 million publications from 87,000 journals. All these four online databases were chosen considering their prominence in providing the most relevant articles for this review paper.

Systematic Searching Strategy

Identification

The identification process consisted of two stages: identification of the main keywords and enrichment of the main keywords. This process is crucial as it ensures a rigorous search strategy through the selected databases. The main keywords constructed for this study were unmodified, modified, eggshell membrane, adsorbent, and pollutant. These keywords were developed by referring to this review's research question, objectives, and keywords used in past studies. The main keywords were then enriched through a few methods: finding the synonyms through an online thesaurus and using variations words. The variation technique was applied to the keyword 'eggshell membrane'. 'Eggshell membrane' and 'eggshell membrane' provided a significant difference in the article search result. Both main and enriched keywords were tabulated in Table I. Next, after finding both main and enriched keywords, a full search string was developed based on field code function, truncation, phrase searching, wild card, and Boolean operator. The full search string used for all databases was tabulated in Table II.

Table I : The main and enriched keywords

Main keywords	Enriched keywords
Unmodified	Natural
Modified	Altered, adjusted
Eggshell membrane	Eggshell membrane
Adsorbent	Adsorption
Pollutant	Hazardous waste

Table II : The search string

Database	Search string
ScienceDirect	(Unmodified OR natural) AND (modified OR altered OR adjusted) AND ("eggshell membrane OR "eggshell membrane") AND (adsorbent OR adsorption) AND (pollutant OR hazardous waste)
Scopus	TITLE-ABS-KEY ((unmodified OR natural) AND (modified OR altered OR adjusted) AND ("eggshell membrane OR "eggshell membrane") AND (adsorbent OR adsorption) AND (pollutant OR hazardous waste))
Web of Science	(Unmodified OR natural) AND (modified OR altered OR adjusted) AND ("eggshell membrane OR "eggshell membrane") AND (adsorbent OR adsorption) AND (pollutant OR hazardous waste)
Dimensions.ai	(Unmodified OR natural) AND (modified OR altered OR adjusted) AND ("eggshell membrane OR "eggshell membrane") AND (adsorbent OR adsorption) AND (pollutant OR hazardous waste)

Screening

The screening included and excluded retrieved articles in the identification process based on the inclusion and exclusion criteria selected by the author. This process was done automatically using the sorting function available in every database. Any duplicate articles were removed during this process. The first inclusion criterion is publication timeline, where only articles published between 2011 and 2021 were included in this review. It was impossible to include all published articles on this topic, explaining why the ten-year period was chosen for the timeline criteria. The next inclusion criteria are that only subscribed journals were included to ease obtaining articles for the review process. Next, the document type is also one of the criteria, where only research articles were selected in this study. Research articles have primary data that can ensure the quality of the review. Therefore, all reviews, books, chapters in a book, and conference proceedings were excluded. Most importantly, only articles written in English were included to avoid the hassle of understanding the research articles. Both inclusion and exclusion criteria were tabulated in Table III.

Eligibility (Manual screening)

Eligibility is a process where authors manually screened all the retrieved articles from the previous method to ensure all articles aligned with the criteria and topic

Table III : The inclusion and exclusion criteria

Criteria	Inclusion	Exclusion
Publication Timeline	2011 - 2021	< 2011
Subscription	Subscribed journal	Unsubscribed journal
Document Type	Research articles	Review, books, chapter in a book, conference proceedings
Language	English	Non-English

of the review. It was done by reading the title, abstract and main contents of all articles to ensure that it is related to the objectives of this review. Articles that are irrelevant to the field of research were excluded from this review.

Quality Assessment

The objective of quality assessment is to ensure that all retrieved articles align with this review's objectives. It is also essential to evaluate the quality of an article before including it in the review process. Quality assessment was performed by two experts in this study and the score for each question was assessed using four criteria adapted from Kushwah et al. (18). Each criteria's score was calculated and added up to obtain a final score. Only the article with a score exceeding fifty percent of the ideal score of 8 was included in this review (19). All criteria used in this process were tabulated in Table IV.
Table IV: Quality assessment tool

Data Extraction and Analysis

After identifying the eligible articles for this review through the quality assessment process, all the required data from these articles were extracted by referring to the objectives and research questions of this review. To achieve a thorough data extraction, a thematic analysis was conducted to generate themes and sub-themes from all included studies. This was done by thoroughly reading their abstract, results and discussion section. During this analysis, any similar patterns among the included articles were identified. These similarities between the articles were then gathered and grouped into main themes and sub-themes accordingly. As a result, all data extracted were grouped based on these main themes and sub-themes. All themes were re-examined to ensure that those themes could answer the research questions of this review and focus on the objectives. Consequently, four main themes were identified, and six sub-themes were successfully extracted from these main themes

RESULT AND DISCUSSION

Description of Studies

Figure 1 shows the flow diagram of the article search result for eggshell membrane-based adsorbent for pollutants removal. In the identification process, 774 articles were retrieved from 2011 until 2021 through all the mentioned online databases. Eight duplicate studies were removed, while another 571 articles were excluded in the screening stage due to the following reasons: outdated studies, not a research article and not written in the English language. This made up 195 articles that remained after the screening stage. Then, another 151 articles were further removed as the abstracts are

Table IV : Quality assessment tool

Quality Evaluation	Description
QE 1	Inclusion of empirical study in the article. The possible answers are quantitative, qualitative and no evidence with scores of +2, +1.5 and 0, respectively.
QE 2	Description of benefits and limitations of the article. The possible answers are yes, partially and no with scores of +2, +1 and 0, respectively. When one of the other two has been elaborated, the partial score will be considered.
QE 3	The justifiable of the study output. The possible answers are yes, partially and no with scores of +2, +1 and 0, respectively. When a technique is explained in a constrained context, the partial score will be considered.
QE 4	Publication avenues rating. The possible answer and scores are +2 if the summation of citation and H index exceeds 100, +1.5 if the summation of citation and H index is between 99 and 50, +1 if the summation of citation and H index is between 49 and 1, and lastly 0 if the summation of citation and H index is 0.

irrelevant to this field of research. That made only a total of 47 articles eligible to be assessed in the quality assessment process, which was adapted from Kushwah et al. (18). These articles were proceeded with a full-text reading process, and 30 articles were removed as they did not pass 50 % of the total score. Finally, 17 articles were selected for the following reviewing process, data extraction and analysis.

Through the full-text reading process, four main themes were selected to be extracted, and from these main themes, a total of six sub-themes were yielded. As shown in Table V and Table VI, the four main themes are (a) type of ESM modifications (two sub-themes) (b) category of pollutants, (c) optimal conditions (four sub-themes), (d) adsorption isotherms and (e) adsorption capacity or removal percentage. In terms of publication year, two articles were published in 2011 (20,21), an article was published in 2012 (22), and three articles were published in 2013 (13,23,24). Then, 2014 has two published articles (25,26), while there is none in 2015, followed by two articles published in 2016 (27,28). In 2017, two articles were published (29,30), while there was only one article in 2018 (2). There were two articles published in 2019 (12,31) and only one article in both years, 2020 and 2021 (32,33). Table V below shows the characteristics of all articles that were selected for this review paper.

Unmodified and Modified Eggshell Membrane as Adsorbent Toward Different Types of Pollutants

The retrieved articles show that the use of ESM as an adsorbent to remove water pollutants is not limited to one type, but instead, two types of ESM, which are unmodified ESM and modified ESM. As observed in Table V, five studies were conducted on unmodified ESM, eleven studies on modified ESM, and one study investigated both modified and unmodified ESM. Based on the reviewed articles, the adsorption properties of unmodified ESM were investigated

Table V : Characteristics of included studies

No	Type of Modifications	Modification Agent/Method	Pollutants Category	Authors
A1	Chemically modified ESM	Hydrochloric acid + sodium hydroxide	Dye	Parvin *et al.* (2019)
A2	ESM derived- magnesium ferrite	$Mg(CH_3COO)_2.H_2O$ + $Fe(NO_3)_3{\cdot}6H_2O$	Antibiotic	Li *et al.* (2017)
A3	Amino-ESM-27	Ammonia + nickel chloride	Heavy metal	Chen & Huang (2016)
A4	Activated ESM (ESAC 3)	Activated by Pyrolysis treatment	Organic compound	Giraldo & Moreno-Pirajбn (2014)
A5	Methyl esterified ESM (MESM)	Methanol + Hydrochloric acid	Heavy metal	Chen et al. (2013)
A6	Polyethyleneimine modified ESM (PEI-ESM)	Polyethyleneimine + methanol + glutaraldehyde	Heavy metal	Zou & Huang (2013)
A7	Thiol-functionalized ESM (TESM)	Ammonium thioglycolate	Heavy metal	Wang et al. (2013)
A8	Layered double hydroxide ESM (LDH/ESM)	Mg(NO3)2·6H2O + Al(NO3)3·9H2O + urea	Heavy metal	Guo et al. (2011)
A9	Methyl esterified ESM (MESM)	Methanol + Hydrochloric acid	Metalloid	Al-Ghouti & Khan (2018)
A10	Methyl esterified ESM (MESM)	Methanol + Hydrochloric acid	Dye	Choi (2017)
A11	Polyethyleneimine modified ESM (PEI-ESM)	Polyethyleneimine + methanol + glutaraldehyde	Heavy metal	Liu & Huang (2011)
A12	Unmodified ESM	-	Dye	Candido et al. (2019)
	ESM impregnated with silver-based flower nanoparticles (ESM + SFNP)	Silver nitrate aqueous solution + ascorbic acid	Dye	
A13	Unmodified ESM	-	Dye	Salman et al. (2012)
A14	Unmodified ESM	-	Dye	Saratale et al. (2021)
A15	Unmodified ESM	-	Heavy metal	Lahmar et al. (2020)
A16	Unmodified ESM	-	Heavy metal	Ho et al. (2014)
A17	Unmodified ESM	-	Heavy metal	Baláž et al. (2016)

against two pollutants: heavy metals and dyes. This can be explained as ESM has been known for its good adsorbent properties, especially against these two pollutants. For example, ESM has a large surface area due to its highly fibrous and porous structure that provides a strong affinity towards many adsorbates (33). The interwoven fibrous structure was exhibited due to the high content of highly crosslinked protein fibers, mainly collagens and glycoproteins (34,35). According to the Scanning Electron Microscope (SEM) and Transmission Electron Microscope (TEM) analysis conducted by He et al. (34), ESM's interwoven fibrous structure has a diameter ranging from 0.5 to 2 μm, and this fibrous structure contributes to the retention of heavy metals and dye molecules (32). Another factor contributing to ESM's good adsorbent properties is it is composed of many functional groups, such as carboxylic, hydroxyl, amines and amides, leading to a potential adsorbent for various types of pollutants.

In addition, the adsorption capability of many waste materials, including ESM, is typically limited. However, the adsorption efficiency shown by ESM can be improved through several modification processes (27). As seen from Table V, the articles on modified ESM are the most commonly studied type in this review, constituting more than 70 % of the reviewed articles. According to Xin et al. (36), adsorbent modification can be done physically or chemically; however, chemical modification is the most common modification type, as it is more efficient and simple. In this review, the type of ESM modifications used in all of the included studies varied from each other. As all of the reviewed articles conducted the adsorption against different pollutants, the modification agent or method will depend on the targeted pollutants to achieve maximum adsorption efficiency. For example, Li et al. (30) used a modified ESM-derived magnesium ferrite (MgFe2O4) for the removal of doxycycline (DC) antibiotic due to the availability of a strong surface complexation between the pollutant and MgFe2O4 that leads to a highly effective adsorption process.

Another example is a study by Chen & Huang (28) where the porous ESM's surface was functionalized with amino groups, producing an adsorbent called Amino-ESM-27. In this study, the adsorption capability of Amino-ESM-27 was investigated against copper ions, which is heavy metal. This can lead to a successful adsorption process; as mentioned by Huang et al. (37), any functional groups with nitrogen, such as amino groups, can produce strong coordination with heavy metals due to the presence of chelating ligands -N. Another study that uses the same functionalization method was carried out by Wang et al. (23). The only difference is that the authors chose thiol groups due to their considerable affinity for numerous heavy metal ions due to Lewis's acid-base interaction.

In addition, there are also a few studies that share a similar modification method. For example, studies conducted by Chen et al. (13), Choi (29) and Al-Ghouti & Khan (2) used the same modification method, which is by using methanol and hydrochloric acid to produce a methyl esterified ESM (MESM). This modification is called esterification, where the negatively charged carboxylic groups on the ESM surface are esterified to produce an improved ESM with a high cationic charge density (29). According to Chen et al. (13), MESM also showed an improvement of adsorption capacity, which offers a 200-fold improvement against its targeted pollutant, arsenic, compared to the unmodified ESM. Another type of modification is shared by Liu & Huang (21) and Zou & Huang (24), where these studies used a polymer, which is polyethyleneimine (PEI), to functionalize the ESM through a crosslinking reaction. As these studies were against chromium (VI) and copper (II) ions, PEI-ESM is the ideal adsorbent. This is because PEI can act as an anion changer and a chelator with metal ions (38), allowing it to adsorb both metal anions and metal cations (24).

Adsorption Conditions

Effect of Temperature

According to Akhtar et al. (39), the changes in temperature can affect the adsorption process in three ways, which are by a) modifying molecular activity at the solid interface, b) disrupting the connection between functional units of solutes and adsorbent species, and (c) altering the adsorbent's nature. The optimum temperature for all included studies has been listed in Table VI. Based on the results, the optimum temperature ranges from 20 °C to 80 °C, with 25 °C being the most optimum temperature among all the reviewed studies. Articles by Salman et al. (22), Chen & Huang (28), Choi (29) and Saratale et al. (33) have shown an increased adsorption capacity due to the increased temperature, indicating that the adsorption process is endothermic. The rising temperature of the sample solution has caused the mobility of pollutants molecule to increase as it gains sufficient energy to interact with the adsorbent (40). Other than that, Zulfikar & Setiyanto (41) mentioned that high temperatures would also cause a swelling effect in the internal structure of the ESM, allowing many pollutants to diffuse further in the pore of ESM. In other words, high temperatures will promote more interaction between the adsorbents and adsorbates, hence improving the adsorption capacity.

In contrast, the adsorption process in articles by Al-Ghouti & Khan (2), Ho et al. (26) and Lahmar et al. (32) favored a low temperature condition, indicating an exothermic nature of adsorption. In these articles, as the temperature of the solution increases, the adsorption capacity decreases. A higher temperature can disrupt certain functional groups responsible for adsorption in some adsorption processes, hence reducing the pollutants uptake. This has also been mentioned in the previous paragraph where temperature can affect the adsorption capacity through various ways, such as altering the nature of the adsorbent. For example, in a study conducted by Al-Ghouti & Khan (2), the high temperature has caused the ester functional group on the esterified ESM to be ineffective, decreasing the boron removal during the adsorption process. The same phenomenon also happened in a study by Ding et al. (42) in which the amine group on the adsorbent has decomposed due to the increased temperature. This has shown that temperature suitability will depend on the type of pollutants used against the ESM-based adsorbent.

Effect of pH

The pH of the solution is also an important condition that can affect the adsorption capacity of the ESM in many ways, such as speciation of pollutants, changes in electrostatic charges and ESM's surface characteristics and degree of ionization (43). The maximum and minimum pH for all studies were observed to be at 1 and 11, respectively. The majority of the studies favored acidic conditions, pH 1 to pH 6. In contrast, pH 7 is the least favored condition seen among all the reviewed articles, with only three studies listed. The effect of pH also differs based on the type of pollutants used. As summarized in Table VI, the optimum pH for dye removal ranges from pH 8 to pH 11, a basic condition. In general, cationic dyes such as methylene blue and methyl orange used by Candido et al. (31) and Salman et al. (22), respectively, show a high removal at a high pH of solution due to the electrostatic bonding formed between positively charged species by the cationic dyes with the ESM. The electrostatic bonding managed to develop as the functional groups of ESM have been deprotonated in the increasing pH condition, hence inducing the release of negative charges on the ESM's surface (44).

Table VI : Experimental conditions of adsorption process with ESM-based adsorbent

Ref ID	Pollutants	Optimal Conditions				Adsorption Isotherm	Q_m or R%
		T (°C)	pH	Adsorbent Dosage	Initial Conc. (mg/L)		
A1	Congo red dye	-	9	7.5 g/L	-	Langmuir	117.7 mg/g
A2	Doxycycline (DC)	25	-	-	-	Langmuir	308.5 mg/g
A3	Cu^{2+}	50	6	0.2 g/L	120	Freundlich	214.6 mg/g
A4	Phenol	25	-	-	-	Langmuir	191.9 mg/g
A5	As(III) As(V)	25	4 6	- -	- -	- -	- -
A6	Cu^{2+}	-	5	0.10 g	-	-	110.7 %
A7	Cr(VI) Hg(II) Cu(II) Pb(II) Cd(II) Ag(I)	30	1 6 6 6 6 6	- - - - - -	- - - - - -	Langmuir Langmuir Langmuir Langmuir Langmuir Langmuir	113.6 mg/g 138.9 mg/g 46.7 mg/g 32.5 mg/g 23.7 mg/g 129.9 mg/g
A8	Cr(VI)	20	5.1	-	-	Langmuir	27.9 mg/g
A9	Boron	25	4	-	17.5	Freundlich	33.33 mg/g
A10	Sulfur blue dye	40	6	0.01 g/L	10	Langmuir	187.6 mg/g
A11	Cr(VI)	35	3	20 g/L	-	Langmuir	161.3 mg/g
A12	Methyl orange dye Methylene blue dye	-	7 11	-	10	Freundlich Freundlich	10.3 mg/g 25.5 mg/g
	Methyl orange dye Methylene blue dye	-	7 11	-	10	Freundlich Freundlich	23.8 mg/g 6.5 mg/g
A13	Methylene blue	80	10	0.75 g	20-40	-	93.3 %
A14	Reactive Red 120 dye	65	3	-	-	Freundlich	191.5 mg/g
A15	Cr(VI)	25	3	-	5	-	-
A16	Ni^{2+} Ag^{+}	45 45	9 4	0.8 g 0.8 g	25	-	91% 100%
A17	Ag^{+} Cd^{2+}	-	6 7	-	-	Langmuir Freundlich	52.91 mg/g 23.42 mg/g

As for heavy metals, these pollutants achieved a high removal efficiency in an acidic condition, ranging from pH 1 to 6, with pH 6 being the optimum pH for heavy metals removal. This can be explained as most heavy metals used in the included articles are positively charged, for example, $Ni^{2+}$and Cu^{2+} in the studies by Ho et al. (26) and Zou & Huang (24), respectively. These positively charged pollutants need to compete with the H^+ ions available in the acidic condition for any vacant active sites on the ESM's surface (28). Lowering he pH even more will produce more hydrogen ions, making the competition even harder, leading to a decreased removal of pollutants. In contrast, a further increase in the pH will cause precipitation by most heavy metals, such as Cu(II), reducing the percentage of pollutants removal (24). These situations have shown that pH 6 is the most suitable pH for heavy metal removal.

Effect of Adsorbent Dosage

According to an article written by Vesali-Naseh et al. (45), knowing the optimum adsorbent dosage is important, especially from an economic standpoint. Improved interaction and contact of the pollutants with the adsorption sites can effectively increase the adsorption capacity of the adsorbent. This can be achieved by experimenting with the best adsorbent dosage in a particular adsorption study. All articles included in this review mentioned that the pollutants removal increased with a higher ESM dosage. However, it only rises until a saturated point, in which the removal capacity will decrease after then. This situation has been explained in one of the included articles by Choi (29), where the author mentioned that an excessive amount of adsorbent dosage could lead to particle aggregation, overlapping and overcrowding. This can result in a reduction of surface area and an increase in the diffusion path length. Therefore, it can be seen from Table VI, the adsorbent dosage used for all included studies is low, ranging from 0.01 g/L to 20 g/L. However, in one of the reviewed studies by Liu & Huang (21), the amount of modified ESM dosage was not changed throughout the study. These authors experimented with the effect of polyethyleneimine (PEI) dose, which is the modification agent for the ESM adsorbent. From this article, the adsorption capacity increased from 102 to 161 mg/g with an increase in the amount of PEI used for PEI–ESM preparation from 0 to 20 g/L. However, the adsorption capacity decreased with a further increase in the amount of PEI used, which is from 20 to 40 g/L. This situation can be explained as with the rise in the PEI dosage, some pore channels on the ESM were blocked due to the amino groups from PEI. This has reduced the surface area on the ESM's surface, hence decreasing the adsorption capacity.

Effect of Initial Concentration

The initial concentration of the pollutant is also a crucial parameter in the adsorption process. It plays an essential role in removing water pollutants by affecting the number of available active or binding sites on the adsorbent's surface. Generally, a higher initial concentration will reduce adsorption capacity due to the higher competition of binding sites within the pollutant molecules. The higher competition is caused by the elevated ratio of the number of pollutants molecules versus the available active sites on the ESM's surface (43). This situation has also shown that the adsorption process is dependent on the availability of binding sites. On the other hand, at a low concentration, there will be sufficient unoccupied active sites on the surface of ESM for the adsorbent to adsorb all the pollutants molecules. Hence, this explains why the optimum initial concentration used in all included studies is low, ranging from 5 mg/L to 25 mg/L, as seen in Table VI. However, there is only one article by Chen & Huang (28) that favors a higher initial concentration, which is 120 mg/L. According to the authors, the high initial concentration of the pollutant, Cu(II) ions, has caused an increased driving force to tackle Cu(II) ions' mass transfer resistance between the aqueous and solid phases. This has resulted in a greater chance of chelating capacity between the pollutant ions and active functional groups available on the adsorbent surface, increasing the adsorption capacity.

Adsorption Isotherms

The adsorption isotherms are commonly applied to predict the adsorption mechanism and the interaction between the adsorbent and adsorbate during the adsorption process at a constant temperature and pH (46). The adsorption isotherms behaviour of all included studies was determined by two isotherm models, namely the Langmuir and Freundlich models. As seen in Table VI, eight included articles best fitted the Langmuir model, indicating that their adsorption process exhibited a monolayer formation of adsorbate on the homogeneous adsorbent surface. The maximum adsorption capacity (Qm) of these Langmuir fitted studies ranges from 23.7 mg/g to 308.5 mg/g. As for Freundlich, nine included articles best fitted this type of isotherm model, with the maximum adsorption capacity (Kf) ranging from 6.5 mg/g to 214.6 mg/g. Freundlich isotherm model follows a multilayer trend of adsorbate formation and a heterogeneous adsorption process.

CONCLUSION

The presence of various pollutants in the water offers many harmful impacts on humans, aquatic organisms, and the environment; hence, it is necessary to mitigate this issue. Adsorption along with a practical choice of adsorbent will significantly help in removing all the pollutants that existed in the water. In this paper, four main themes and six sub-themes derived from these themes were successfully extracted. All main themes, which comprised ESM-based adsorbent modification and its optimum operating parameters, were systematically reviewed. The types of modification conducted on the eggshell membrane were extracted and presented based on their classifications of modification agents and methods. Most studies that use modified ESM-based adsorbent have concluded that modified ESM promotes better and more effective adsorption capacity than unmodified ESM. However, the unmodified ESM still works excellently in removing water pollutants due to its good adsorbent properties. As for the operating parameters, the temperature and pH of the solution had more significant impacts on the adsorption capacity than adsorbent dosage and initial concentration. Both adsorbent dosage and initial concentration can still elevate the adsorption capacity of ESM-based adsorbent, however only in a limited range. The Freundlich adsorption isotherm is the most suited model to explain the adsorption behaviour for all of the included articles, indicating that the adsorption follows heterogenous and multilayer adsorption systems. Overall, this review has gathered all the patterns and trends commonly used in adsorption relating studies. It is able to provide a basic understanding of this topic for any concerned parties in implementing new strategies to solve the water pollution issues. Finally, new disciplines and variables arising from this review's findings could also assist in providing new knowledge for future research.

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