



# Valorizing papaya seed waste for wastewater treatment: a review

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## Abstract

The use of papaya seed as a precursor material for the production of adsorbents and coagulants, and their applications in wastewater treatment processes to remove pollutants, such as heavy metals, dyes and microorganisms, are reviewed in this manuscript. Recent studies have shown that adsorbents derived from papaya seed can be used to replace high-cost adsorbents as low-cost alternatives. The raw papaya seed biomass can be chemically modified to alter its surface functional group to improve the adsorption efficiency. In this work, the adsorption isotherms correlated with heavy metal and dye adsorption onto papaya seed adsorbent are discussed. The remarkable performance of papaya seed natural coagulant in removing turbidity and dye in the coagulation and flocculation process is also presented. The development of a highly efficient adsorbent and coagulant using papaya seed remains a critical field area of ongoing research. This review sheds light on the recent development of various feasible approaches for incorporating papaya seed into various wastewater treatment processes, thereby transforming it into a value-added product.

**Keywords** Papaya seed · Wastewater treatment · Adsorption · Coagulation · Isotherms

## List of symbols

%	Percentage	g	Gram
ACH	Polyaluminium chlorohydrate	g/L	Gram per litre
<i>C. papaya</i>	<i>Carica papaya</i>	IC	Indigo carmine
Cd	Cadmium	MB	Methylene blue
Cd <sup>2+</sup>	Cadmium (II) ions	mg/g	Milligram per gram
COD	Chemical oxygen demand	mg/g min	Milligram per gram minute
CR	Congo red	mg/L	Milligram per litre
Cr	Chromium	mm	Millimetre
Cu	Copper	Mn	Manganese
DDR	Drimarene dark red	N/A	Not applicable
FAO STAT	Food and Agriculture Organization of the United Nations	NaOH	Sodium hydroxide
Fig.	Figure	Ni	Nickel
		PAC	Polyaluminium chloride
		PASS	Polyaluminium silicosulphate
		Pb	Lead
		Pb <sup>2+</sup>	Lead (II) ions
		PFS	Polyferric sulphate
		R <sup>2</sup>	Coefficient of regression
		TDS	Total dissolved solids
		UNESCO	United Nations Educational, Scientific and Cultural Organization
		US EPA	United States Environmental Protection Agency
		Zn	Zinc

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## Introduction

The population of the world is expected to reach 9.8 billion by the middle of the century, posing a global challenge as safe, clean and potable water is scarce even in developed countries (Edogbanya and Obaje 2020). Water pollution is mainly caused by rapid urbanization, agricultural expansion, pesticide use, land degradation, high population density, as well as inefficient waste disposal, resulting in deteriorated water quality and water scarcity (Villaseñor-Basulto et al. 2018). According to the United Nations Educational, Scientific and Cultural Organization (UNESCO) water quality statistics, 2.4 billion people in the world live without access to proper sanitation. Inadequate sanitation significantly worsens water pollution. In developing countries, up to 90% of sewage is discharged untreated directly into water bodies, where 2 million tonnes of sewage and other effluents are discharged into the water bodies daily, while the industry discharges between 300 and 400 megatonnes of sewage into water bodies annually (UNESCO 2018).

It is critical to manage wastewater carefully at every stage of the water cycle, from collecting fresh water, pre-treating it, distributing, using, collecting, and recovering, to using treated wastewater before returning it to the environment for further cycle. As a result of population growth, urbanization, and economic development, the amount of water generated and its overall pollution load are increasing globally (UN Water 2021). The problem is exacerbated by the fact that wastewater sources include both domestic and urban wastes, as well as industrial waste such as textiles, and pulp and paper. There are many low-income areas of cities and towns in developing countries that dump wastewater directly into local surface water drains or informal drainage channels, sometimes with or without treatment. The wastewater system frequently contains a high concentration of highly toxic chemicals from small-scale mining and the automobile industry, as well as household effluent and human waste. According to the Environmental Protection Agency (EPA), there are approximately 23,000 to 75,000 sanitary sewer overflows every year in the USA. Untreated waste from sewage treatment plants is estimated to be between 3 and 10 billion gallons annually (UpKeep 2019).

Wastewater treatment is an important process in sanitation systems. The removal of hydrophobic colloids from water presents a major challenge in water treatment systems since these particulates, which are mostly organic in nature, are usually abundant and present in large quantities when compared to other pollutants (Villaseñor-Basulto et al. 2018). Biological treatment, chemical treatment, and physicochemical treatment are among the widely adopted

wastewater treatment processes. Coagulation and flocculation are physicochemical processes that are commonly used at the start or end of wastewater treatment processes. Depending on the chemical composition of the contaminants present in the water, various types of coagulant are commonly used in the conventional treatment processes. Coagulants are generally classified as inorganic, synthetic organic, or natural organic polymers. As a result of new wastewater treatment initiatives, more environmentally friendly materials are being developed to treat wastewater, including natural organic polymers that can reduce costs, prevent pH variations, prevent sludge production, and increase biodegradation. Despite recent research on natural coagulants for wastewater treatment, systematic information on the main benefits and drawbacks, challenges, and future prospects of using such natural materials for wastewater treatment remains lacking (Villaseñor-Basulto et al. 2018).

Extensive efforts have been done in the past to investigate the effectiveness of natural coagulants as a substitute for chemical coagulants, owing to their properties of being toxic-free and safer to be used, as well as being sustainable and biodegradable (Sillanpää et al. 2015; Hoong and Ismail 2018). Beans, cowpea, chitosan, peanuts, *Plantago ovata*, *Leucaena leucocephala*, maize seed powder, *Moringa oleifera*, *Vicia faba* and surjana seed powder are among the natural coagulants derived from naturally available sources or waste materials (Patel and Vashi 2012; George and Chandrn 2018; Kristanda et al. 2020). Among these sources, papaya seed has also been reported as a potential source of natural coagulant for wastewater treatment. Generally, the food and fruit processing industries discard papaya seeds as waste, accounting for approximately 20% of the total weight (Kumoro et al. 2020). According to Kumoro et al. (2020), papaya seeds contain approximately 27.3–28.3% proteins, 19.1–22.6% fibre and 28.2–30.7% fat. Papaya seeds are rich in monosaturated fatty acids, like oleic acid. They also contain polyphenols, flavonoids, alkaloids, tannins and saponins, which are strong antioxidants and exhibit anti-inflammatory properties (Jain 2020). From the work of George and Chandrn (2018) and Muda et al. (2020), it was possible to achieve turbidity removal efficiency up to 89.14–95.5% using papaya seed as the coagulant.

Adsorption, on the other hand, is a noteworthy process that is gaining popularity in wastewater treatment nowadays. Various synthetic dyes can be effectively isolated from industrial effluents using adsorbents, such as activated carbon, silica-based adsorbents, fly ash and biosorbents. Numerous studies have been conducted over the last few decades to investigate the efficacy of biosorbent as an alternative to activated carbon and conventional adsorbents. These biosorbents are derived from natural sources and



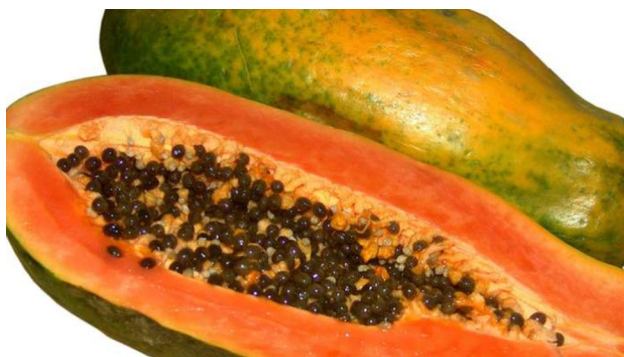
waste materials, including *Amaranthus hybridus* (African spinach), peanut shells, walnut shells, banana waste, pine cone shells, and micro-grooved chitosan beads (Egila et al. 2011; Deniz 2014; Santos et al. 2016; Ahmad and Danish 2018; Wong et al. 2020). Papaya seed is among the identified potential biosorbents for the removal of various dyes and heavy metals from wastewater as well.

This review aims to provide valuable insights into the use of *Carica papaya* seeds for wastewater treatment, especially in the areas of adsorption and coagulation–flocculation processes. Aside from emphasizing the importance of papaya seed as natural adsorbents and coagulants, the impact of various process parameters on its performance is presented. This review will be of interest to researchers and practitioners interested in the biochemical properties of papaya seed, as well as its potential for wastewater treatment.

## Papaya

### Properties, composition and general applications

Papayas (*C. papaya*) grow wild in many tropical regions and thrive in nearly every type of rainforest soil. The major papaya producing countries are India, Brazil, Nigeria, Indonesia and Mexico which produce approximately 6,642 tonnes of papaya fruits each year to meet the overwhelming global demand (Singh 2011). It is an extremely important fruit for human consumption and its cultivation is on the rise all over the world (Liu et al. 2012). In general, papaya fruit (Fig. 1) tastes mildly sweet and musky, with a nice tang that varies depending on the species and climate. It is estimated that there are over fifty different types of papaya. According to Tan et al. (2020), uncontrolled pollination has resulted in a decrease in the pure breeding of papaya varieties. Papaya fruit can be oval or elongated in shape, measuring 75 mm to 500 mm in length, and weighing between 500 and 1000 g. The flesh is deep yellow or orange in colour,



**Fig. 1** Longitudinal section of papaya fruit (*C. papaya*) (Britannica 2021)

and can occasionally be salmon-coloured. The papaya fruit contains a large number of small black-coloured seeds. Various round, wrinkled black seeds are attached to the walls of the vast central cavity.

Papaya is a popular fruit due to its delicious flavour and high nutrient content. In addition to vitamins A, B, and C, papayas contain papain, chymopapain, and a number of other probiotics, as well as antimicrobial, antioxidant, antibacterial, and anti-inflammatory properties (Vij and Prashar 2015). Researchers are becoming more interested in papaya wastes due to their superior medicinal properties, despite the fact that the wastes are made up of skin and seed. Antioxidant properties have been discovered in both papaya peel and seed (Cheok et al. 2016). In addition, papaya seeds are also reported to possess antimicrobial properties against *Salmonella choleraesuis* and *Staphylococcus aureus*, making them a potential wound-healing agent (Nayak et al. 2012). *C. papaya* is traditionally used to treat a wide array of skin disorders, including wounds. In traditional medicine, the active ingredient found in *C. papaya* is papain, an enzyme that protects against ulcers. In the past, a decoction made from papaya seeds was used to treat skin ulcers and inflammation (Nayak et al. 2012). It is widely used in developing countries nowadays to treat various wounds, particularly burns, as it is readily available and effective.

Apart from the flesh, Marfo et al. (1986) analysed the chemical composition of papaya seeds to assess both their nutritional value and toxicity. The seeds (Fig. 2) in papaya fruit, accounting for approximately 15% to 20% of the total flesh weight, contain high levels of lipids and protein (Hameed 2009). According to Jain (2020), 100 g of dried papaya seeds provides approximately 558 cal of energy. Papaya seeds, on the other hand, have anti-inflammatory and wound-healing properties, are good for digestion, help



**Fig. 2** Papaya seeds



prevent cancer and kidney disorders, boost immunity and promote heart health (George and Chandrn 2018). This is due to the fact that they are high in vitamins, such as Vitamin A, Vitamin C, Vitamin P and a variety of B vitamins and minerals such as iron (Fe), magnesium (Mg), calcium (Ca), phosphorus (P), zinc (Zn), and many more. Besides, papaya seeds have a high oil content that ranges from 13.9% to 30.7%, which primarily consists of nutraceuticals and monosaturated fatty acids, such as oleic acids (Kumoro et al. 2020).

Most of the papaya seeds are discarded as agricultural waste during fruit processing. They are inedible due to the presence of toxic chemicals, such as phytates, glucosinolates and tannins (Kristianto et al. 2018a). However, its anti-inflammatory properties help to speed up digestion and eliminate toxins more effectively (Shorts 2021). In one study, papaya seeds were shown to protect and preserve kidney health and function, whereby papaya seed extract could prevent kidney damage in rats given toxic medications. This could be attributed to the antioxidant properties and fatty acid content of the seeds.

There are a variety of ways to use papaya seeds, including food and non-food applications. As an alternative to black pepper in the food industry, papaya seed is used as a substitute for this spice. In contrast to raw papaya seed, which has a spicy-pungent flavour, papaya seed oil has a distinct aroma composition (Yanty 2014; Tan et al. 2019). The oil is usually extracted by ultrasound-assisted extraction, extrusion and expelling processes, as well as solvent and aqueous enzymatic extraction (Puangsri et al. 2005; Lee et al. 2011; Samaram et al. 2015). As described by Samaram et al. (2014), the papaya seed oil is resistant to oxidation, making

it a highly beneficial oil in the food industry with exceptional health benefits. On the other hand, papaya seed flour is a nutritious food ingredient with excellent foaming, emulsifying, and protein content. It is recommended for use in the food product formulation due to its high nutritional content (Alobo 2003; Santos et al. 2014). When papaya seed flour is used in hamburgers, for example, it increases the moisture content while reducing the shrinkage of the hamburger meat (Azevedo and Campagnol 2014).

Table 1 shows the proximate contents of dried papaya seed meal. Different ripening and maturing stages, as well as climates, may result in different proximate composition of papaya seeds.

## Papaya production

Papaya is one of the most popular fruits in the world, accounting for approximately 15.36% of total tropical fruit production worldwide (Fatombi et al. 2019). Figure 3 shows the top 10 major papaya producers in the world, along with their average production from 2015 to 2019. India contributed to the highest production, with 5.7 million tonnes, approximately 5 times higher than Brazil which produced the second most, with approximately 1.2 million tonnes. Asia has been the leading papaya producing region (56.7%) followed by the Americas (32.1%), Africa (11.1%) and Oceania (0.1%). In Asia, the production of papaya in Malaysia increased steadily from 2015 to 2017, but then decreased drastically from 2017 to 2018 and then slightly increased in 2019 (Fig. 4). The global papaya production has recorded an increase of 13.19% from 2015 to 2019. According to the Food and Agriculture Organization of the United Nations (FAOSTAT), the global *C. papaya*

**Table 1** Proximate contents of dried papaya seed meal

Nutrients (%)				References
Crude protein	Crude fat	Crude fibre	Crude ash	
27.3–28.3	28.2–30.7	19.1–22.6	2.4	Kumoro et al. (2020)
26.3	28.2	30.8	2.09	Muazu and Aliyu-Paiko (2020)
27.4	28.6	8.02	5.21	Moses and Olanrewaju (2018)
35.69	7.68	3.88	7.95	Onuoha et al. (2018)
33.0	N/A	18.8	9.00	Seshamamba et al. (2018)
11.67 ± 0.04	2.51 ± 0.03	32.51 ± 0.03	5.98 ± 0.03	Martial-Didier et al. (2017)
25.4	21.0	24.3	6.43	Azevedo and Campagnol (2014)
28.6	29.7	8.78	6.94	Santos et al. (2014)
25.1 ± 0.08	0.00 ± 0.01	45.6 ± 0.05	8.2 ± 0.08	Maisarah et al. (2014)
43.61–48.42	N/A	17.95 ± 0.125	6.08 ± 0.0805	Parni and Verma (2014)
31.3	32.5	5.19	8.89	El-Safy et al. (2012)
29.1–31.9	29.4–31.6	7.80–9.40	9.94–11.5	Adesuyi and Ipinmoroti (2011)
14.21	28.73	33.62	10.30	Dakare et al. (2011)
30.1	34.8	1.67	7.11	Bolu et al. (2009)
32.4 ± 0.48	N/A	4.2 ± 0.06	5.3 ± 0.13	Alobo (2003)
44.4 ± 0.50	28.3	31.8 ± 1.74	4.48 ± 0.01	Marfo et al. (1986)



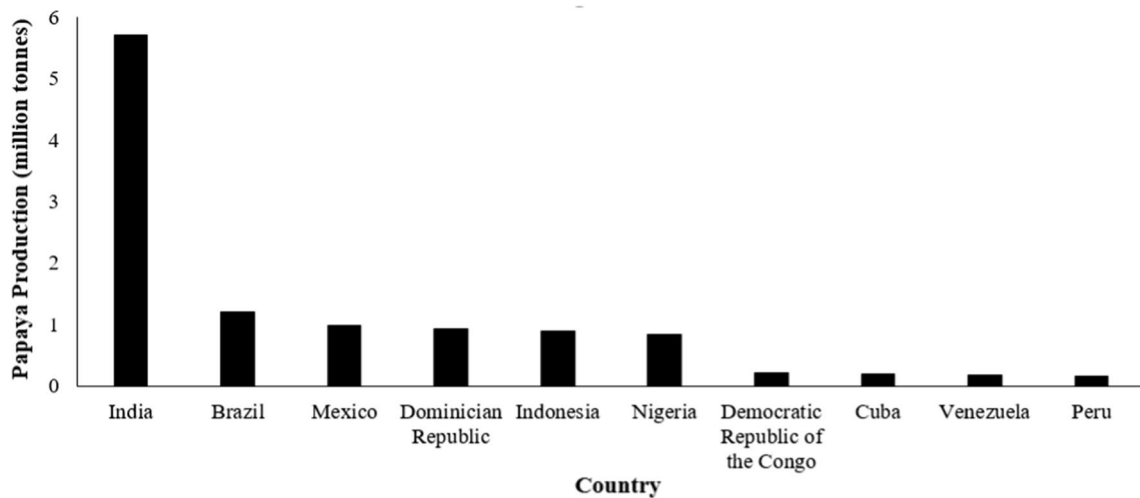


Fig. 3 Top 10 global papayas producers with the average production rate between 2015 and 2019 (FAOSTAT 2021)

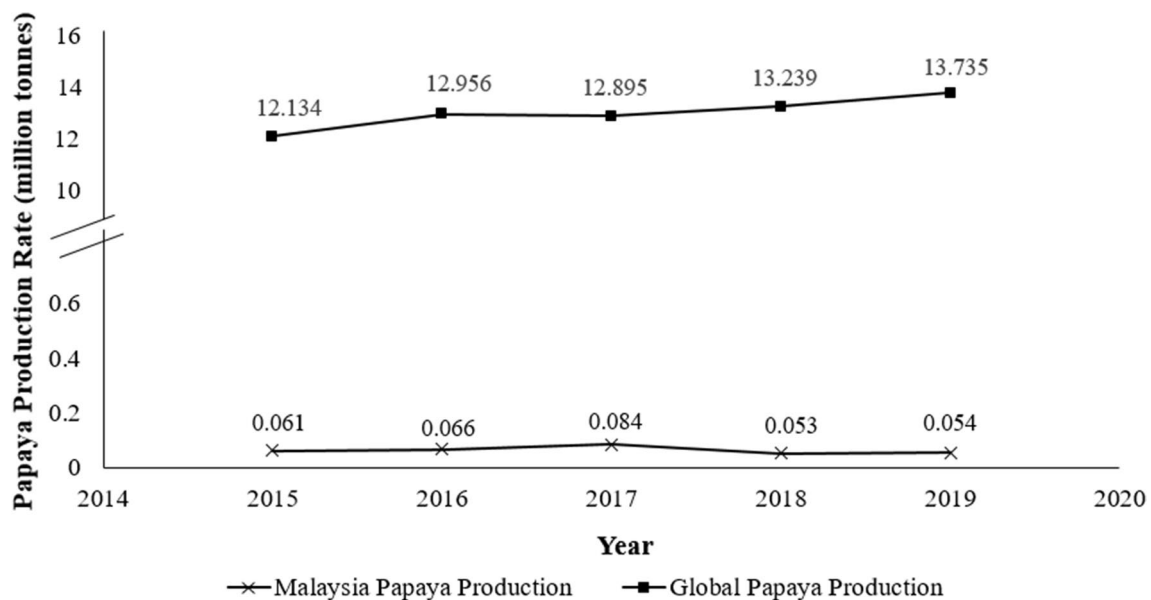


Fig. 4 Global and Malaysian papaya production from 2015 to 2019 (FAOSTAT 2021)

production in 2015 exceeded 60,625 tonnes, and the production rate is expected to rise in the coming years. With such a large amount of papaya produced and an approximate culling rate of 30% to 50%, there is a massive amount of papaya wastes such as papaya peels, seeds and leaves produced at the same time. Typically, papaya wastes are discarded as agricultural wastes that must be disposed of in a landfill.

### Papaya seed as a bio-based material in wastewater treatment

Papaya seed is mainly prepared in a few steps or stages before being used in adsorption or coagulation process, which involve seed harvesting, drying, crushing, sieving



and extraction if it is needed. The effectiveness of the papaya seeds in removing pollutants from different wastewater sources using various processes differs depending on how they are prepared. It is essential for the seeds to be matured and ground prior to harvesting. A grinder, mortar, blender, and mixer can all be used to obtain papaya seed powder with specific particle size. Papaya seeds with small particle sizes are preferred as the amount of lipids in the seeds is reduced, lowering the colour and turbidity of the treated water. From the literature, findings by numerous researchers showed that particle sizes ranging

from 40 to 450  $\mu\text{m}$  were suitable for wastewater treatment applications (Hameed 2009; Nasuha et al. 2011; Foletto et al. 2013; Ibrahim et al. 2014; Weber et al. 2014). Past studies have revealed that coagulating compounds from papaya seeds could be extracted with sodium hydroxide (NaOH), hexane, acetic acid, ethanol, distilled water and water (Unuabonah et al. 2009; Adie Gilbert et al. 2011; Garba et al. 2016; Fatombi et al. 2019; Mrad et al. 2020). After the extraction process, papaya seed powder is sometimes processed with washing, drying, crushing and sieving (Adie Gilbert et al. 2011; Fatombi et al. 2019). The

**Table 2** Papaya seed processing methods used in different wastewater treatment processes

Seed processing method	Source of pollutant	Water treatment process	References
Added in iron sulphate and iron chloride hexahydrate, washed (distilled water and ethyl alcohol), dried and kept	Synthetic methylene blue (MB)	Adsorption	Jong et al. (2020)
Extraction with 1 M NaOH, washed, dried and sieving with particle size 50 $\mu\text{m}$	Synthetic Indigo carmine (IC) and Synthetic Congo red (CR) dyes from single and binary solutions	Adsorption	Fatombi et al. (2019)
Drying, crushing and defatted using Soxhlet extraction method with hexane as solvent	Copper (II) and lead (II)	Adsorption	Garba et al. (2016)
Drying and sieving with particle size lower than 212 $\mu\text{m}$	Synthetic MB from aqueous solution	Adsorption	Ibrahim et al. (2014)
Drying, grinding and sieving with particle size between 350 and 450 $\mu\text{m}$	Hazardous pharmaceutical dyes	Adsorption	Weber et al. (2014)
Drying, crushing and sieving with particle size between 350 and 450 $\mu\text{m}$	Large pollutant molecule from aqueous solution	Adsorption	Foletto et al. (2013)
Drying and crushing in a knife mill	Tannery dye from aqueous solution	Adsorption	Weber et al. (2013)
Extraction with hexane, dried and crushed	Lead (II) $\text{Pb}^{2+}$ and Cadmium $\text{Cd}^{2+}$	Adsorption	Adie Gilbert et al. (2011)
Esterification with pure methanol, washed with distilled water and dried	Synthetic cationic and anionic dye adsorption from aqueous solution	Adsorption	Nasuha et al. (2011)
Washing (distilled water), drying and sieving with particle size 125–250 $\mu\text{m}$	Synthetic cationic and anionic dye from aqueous solution	Adsorption	Nasuha et al. (2011)
Washing (distilled water), drying and sieving with particle size 125–250 $\mu\text{m}$	Synthetic MB solution	Adsorption	Hameed (2009)
Drying, crushing and defatted using Soxhlet extraction method with hexane as solvent	Synthetic MB solution	Adsorption	Unuabonah et al. (2009)
Drying, crushing and sieving with size 20 mesh	Amalgamation slurry waste	Coagulation–flocculation	Nurcholis et al. (2020)
Extraction with water, acetic acid, or ethanol	Contaminated water	Coagulation–flocculation	Mrad et al. (2020)
Papaya seeds were made into powder	Surabaya River water	Coagulation–flocculation	Suprawito et al. (2020)
The papaya seeds are remained	Surabaya River water	Coagulation–flocculation	Suprawito et al. (2020)
Washing (water), drying, crushing and sieving with finer particles	Kallada River water	Coagulation–flocculation	George and Chandrn (2018)
Washing, drying and sieving with particle size 40–50 mesh	Synthetic Drimarene dark red (DDR)	Coagulation–flocculation	Kristianto et al. (2018a)
Washing, drying and grinding with mesh size -0 + 40, -40 + 50, -50 + 60	Synthetic turbid water	Coagulation–flocculation	Kristianto et al. (2018b)
Drying and sieving with mesh size 0.5 mm	Municipal wastewater	Coagulation–flocculation	Maurya and Daverey (2018)
Grinding and sieving with a pore size of 0.4 mm	Synthetic wastewater	Coagulation–flocculation	Unnisa and Bi (2018)



papaya seeds contain a high protein content, and some authors have speculated that the active coagulating agents in the extract are proteins (George and Chandrn 2018).

Table 2 summarizes the papaya seed processing methods used in adsorption and coagulation–flocculation processes as reported by various researchers. Although papaya seed coagulant is commonly extracted with distilled water, the main disadvantage of using it to remove turbidity from wastewater is the high chemical oxygen demand (COD) in the treated water due to dissolved organic carbon content from the papaya seed-coagulant.

## Papaya seed as an adsorbent in adsorption processes

### Mechanism of adsorption process

Adsorption is the process through which a substance, originally present in one phase, is removed from that phase by accumulation at the interface between that phase and a separate (solid) phase. The biosorption process is one of the most promising alternatives for removing heavy metal ions from aqueous solutions by using biomass or biosorbents instead of precipitation, membrane filtration, electrolysis or reverse osmosis (Garba et al. 2016). When compared to other types of the wastewater treatment processes, such as chemical precipitation and electrochemical treatment, the adsorption process is reported to be flexible and simple to operate, with fewer sludge disposal problems (Ojovan and Lee 2005; Powell Water Systems 2016; Krishnan et al. 2021). It has been discovered to be the most efficient and cost-effective method of all dye removal approaches due to its long use in wastewater treatment and demonstrated remarkable effectiveness (Kandisa and Saibaba 2016). The adsorption process occurs when molecular species are deposited onto a surface. Adsorbates are molecular species that become adsorbed on surfaces, and the surfaces on which adsorption occurs are known as adsorbents.

Adsorbents such as activated carbon, silica-based materials, fly ash and biosorbents can isolate dye from industrial wastewater effluents effectively. For instance, in the case of activated carbon, Adegoke and Bello (2015) and Nassar and Geundi (2007) concluded that the material could adsorb compounds, but lacked functional groups suitable for dye molecules. Besides, since the particle size of dye molecules are usually very large, the steric effect will obstruct the molecular transport during the process. When activated carbon prepared from mahogany sawdust and prickly pear seed cake was used to remove dyes, the adsorption capacity was found to be higher than 300 mg/g and 336.12 mg/g for direct dyes and methyl orange,

respectively (Malik 2004; el Maguana et al. 2020). Natural adsorbents, in addition to activated carbon, have been also discovered to remove dye molecules from wastewater. For example, Rodríguez-Arellano et al. (2021) found that cocoa bean powder has a maximum adsorption capacity of 95.79% for removing CR dye. Papaya seed, on the other hand, has the ability to remove synthetic dyes in the range of 80.47% to 98.82% (Unuabonah et al. 2009; Nasuha et al. 2011; Pavan et al. 2014; Mounir et al. 2017; Zaharaddeen et al. 2019; Jong et al. 2020). Natural adsorbents are often used due to their environmentally friendly properties, ease of availability in nature and economic effectiveness.

### Dye removal application

Papaya seeds contain functional groups such as the O–H, C=O, C–O, C=C aromatic, C–H aliphatic and C–H aromatic groups, making them a good candidate for dye removal from wastewater as an adsorbent. The dye removal percentage demonstrated by papaya seed ranged between 80.47 and 98.82% for the MB, CR, IC, tannery, malachite green, Procion red, crystal violet, tartrazine and amaranth. Nasuha et al. (2011) used esterification to chemically modify papaya seeds. It was observed that the esterification process produced ions with negative charge, which was the opposite of that positive ions carried by MB dye. As a result of the decreasing repellent surface functional group of papaya seed and the cations of MB dye, the removal of MB dye has increased. Thus, in comparison with raw dried papaya seeds, esterified papaya seeds demonstrated a higher dye removal percentage (96.29%) than the raw dried papaya seeds (91.79%). It was shown that altering the carboxyl group in the papaya seed improved the adsorption capacity for removing cationic dye from aqueous solutions. Jong et al. (2020) used magnetized papaya seeds to achieve a high MB dye removal of 80.47%. The high percentage of dye removal may be caused by electrostatic interactions between the positively charged MB dye molecules and the negatively charged surface of the magnetized papaya seeds. This is due to the presence of a large number of negatively charged surface sites in plant lignocellulosic materials, such as papaya seeds, including hydroxyl and carboxyl groups in cellulose fibres and phenolic compounds in proteins (Egila et al. 2011; Lindholm-Lehto 2019). Because of its chemical composition, papaya seed has been shown to reduce or mitigate MB dye pollution problems.

Besides, researchers have also found that dyes can be removed using raw papaya seeds in powder form, both defatted and undefatted (Hameed 2009; Unuabonah et al. 2009; Paz et al. 2013; Weber et al. 2013, 2014; Pavan et al. 2014; Mounir et al. 2017; Zaharaddeen et al. 2019). The major parameters affecting dye removal efficiency are the initial



concentration, contact time, pH and adsorbent dosage. As the solution concentration decreases, the dye removal percentage increases, indicating that dye removal is directly proportional to the initial dye concentration. By increasing the initial MB dye concentration from 10 to 400 mg/L, for example, the amount of MB dye adsorbed on defatted papaya seed adsorbent increases from 0.98 to 39.56 mg/g (Unuabonah et al. 2009). Besides, Weber et al. (2013) discovered that increasing the dye concentration from 300 to 400 mg/L resulted in reducing the dye removal percentage from 67 to 52%. This was probably because dye molecules aggregated on the surface of the adsorbent, leading to less accessible active sites on the surface (Gaffar et al. 2004).

The contact time will also have an impact on the dye removal efficiency. With the exhaustion of surface adsorption sites, the rate of dye molecules transporting from the surface to the interior section of the adsorbent particle determines the uptake rate (Mounir et al. 2017). It was observed by Hameed (2009) that MB dye uptake began rapidly and then gradually slowed down until it reached saturation. It was possible that the initial rapid phase occurred because there were more vacant sites available at the start. However, as time passed, it became more difficult to occupy those that remained (Zaharaddeen et al. 2019).

The pH of the adsorption medium affects not only the surface charge of adsorbent, but also the degree of ionization of the solution and the dissociation of functional groups at adsorbent and dye active sites (Onyango et al. 2004). According to Hameed (2009), the adsorption of MB dye was minimum at pH 3 and increased with pH as pH

was increased to 4, where it remained nearly constant over the pH range of 4 to 10. The low MB dye adsorption rate on papaya seed at  $\text{pH} < 4$  may be attributed to the positive charge on the surface which made hydrogen ( $\text{H}^+$ ) ions highly competitive with dye molecules, resulting in a reduction in the amount of dye adsorbed (Hameed 2009).

The dye removal efficiency is also affected by the adsorbent dosage. Unuabonah et al. (2009) showed that increasing the dosage of defatted papaya seeds from 0.05 g to 2 g increased the percentage of MB dye adsorbed from 98 to 100%. The result could be explained by the larger surface area and increased adsorption sites available following the increase in adsorbent dosage (Mounir et al. 2017). In a separate work by Zaharaddeen et al. (2019), malachite green dye was removed using defatted papaya seed adsorbent and the authors reported that the dye removal increased when defatted papaya seed adsorbent dosage was increased from 25 to 175 mg.

Table 3 tabulates the summary of adsorption capacities of papaya seed biosorbent for various dye removal processes.

## Heavy metal removal application

The heavy metals that were found to be removed by using papaya seed biosorbent were inclusive of copper (II) ions, lead (II) ions, chromium (VI) ions, nickel (II) ions, cadmium (II) ions, manganese (II) ions and zinc (II) ions (Adie Gilbert et al. 2011; Egila et al. 2011; Ong et al. 2012; Chithra et al. 2014; Garba et al. 2016; Rahmawati et al. 2016).

**Table 3** Adsorption capacities of papaya seed biosorbent for the removal of various dyes

Adsorbent	Adsorbate	$q_m$ (mg/g)	Temperature (°C)	pH	Removal Percentage (%)	References
Magnetized papaya seed	Synthetic MB	–	–	–	80.47	Jong et al. (2020)
Papaya seed	Synthetic CR from binary solution	635.99	30	–	–	Fatombi et al. (2019)
Papaya seed	Synthetic CR from single solution	319.08	30	–	–	Fatombi et al. (2019)
Papaya seed	Synthetic Indigo carmine (IC) from single solution	168.51	30	–	–	Fatombi et al. (2019)
Papaya seed	Synthetic IC from binary solution	102.65	30	–	–	Fatombi et al. (2019)
Defatted papaya seed	Synthetic Malachite green dye	126.05	–	–	89	Zaharaddeen et al. (2019)
Papaya seed	Synthetic MB	52.28	–	10	98.82	Mounir et al. (2017)
Formosa papaya seed	Synthetic crystal violet	85.99	25	8–10	98.4	Pavan et al. (2014)
Papaya seed	Synthetic tartrazine and amaranth	51 and 37.4	24.85	2.5	–	Weber et al. (2014)
Papaya seed	Synthetic Procion Red	73.26	–	–	–	oletto et al. (2013)
Papaya seed	Synthetic MB	637.29	–	–	–	Paz et al. (2013)
Papaya seed	Synthetic Tannery dye	440	–	–	–	Weber et al. (2013)
Esterified papaya seed	Synthetic MB	250	30	–	96.29	Nasuha et al. (2011)
Papaya seed	Synthetic MB	200	30	–	91.79	Nasuha et al. (2011)
Papaya seed	Synthetic MB	555.57	30	3–10	–	Hameed 2009)
Papaya seed	Synthetic MB	1250	30	6.25	99	Unuabonah et al. (2009)





Heavy metals, both as metals and as metallic compounds, are known to cause adverse health effects in the industry. According to the International Labour Organization (ILO) (2004), lead (Pb) have long been used as paint pigments, but their use has been drastically reduced to avoid health risks. Chromium (Cr) is widely used in various industries, particularly in the production of salt and chromic acid, but it can cause cancer. Nickel (Ni), on the other hand, is a sensitizer and a cancer-causing substance. Besides, cadmium (Cd) is one of the environmental and marine pollutants that affect human health by gradually reducing kidney function when stored in the kidneys over a lifetime. Meanwhile, an excessive amount of manganese (Mn) weakens the body's immune system and can harm the nervous system. Zinc (Zn) and copper (Cu) are frequently found in the fumes that cause "metal fever".

The heavy metal removal percentage achieved by papaya seed biosorbent ranged between 95 and 99.96% (Adie Gilbert et al. 2011; Ong et al. 2012; Garba et al. 2016). The pH, biosorbent dosage, initial metal ions concentration, contact time and agitation rate were among the major parameters that affected heavy metal removal. Chithra et al. (2014) reported that the removal of Cr (VI) decreased as the initial pH of the solution increased from pH 2 to pH 8. An increase in initial pH reduced the adsorption capacity of dichromate due to the presence of hydroxide (OH<sup>-</sup>) ions, which hindered the diffusion of the dichromate ions to the *C. papaya* seed biosorbent. Ucin et al. (2002) speculated that this could be due to repulsive forces between the *C. papaya* seed biosorbent and Cr (VI) ions. The adsorption capacity for Ni (II) in the study reported by Chithra et al. (2014) showed an increase followed by a reduction from pH 4 to 10, with the maximum adsorption capacity occurring at the initial pH of 6. At alkaline region, the Ni (II) ions started to precipitate to nickel hydroxide, reducing the uptake of Ni (II) ions by the adsorbent.

The effect of biosorbent dosage on the heavy metal removal was demonstrated by Chithra et al. (2014) who achieved a high removal percentage of Ni (II) and Cr (VI) ions when the biosorbent (*C. papaya* seed) amount was

increased from 0 g/ 100 mL to 1.5 g/ 100 mL. Based on the results, both metal ions were removed at a higher percentage with increasing amount of biosorbent, and were then maintained at a constant percentage. It could be because the number of adsorption sites and surface area of the adsorbent increased with increasing biosorbent dosage, resulting in a better removal of heavy metal ions at higher concentration levels (Chithra et al. 2014).

The initial concentration of metal ions will affect the heavy metal removal percentage as well. Egila et al. (2011) has demonstrated *C. papaya* seeds could be used to remove Mn (II) and Pb (II) ions from aqueous solutions. The results showed that when the heavy metal ions concentration increased from 10 mg/ 100 cm<sup>3</sup> to 50 mg/ 100 cm<sup>3</sup>, the Mn (II) ions was removed at a higher removal rate than Pb (II) ions under the same condition. The amount of heavy metal ions bound by lignocellulosic substrates was found to be dependent on the type and concentration of heavy metal ions. The amount of heavy metal presence or available in the solution increased as its initial concentration was increased, thus enhancing the removal efficiency. It is also to note that at low initial heavy metal ion concentrations, adsorption sites on the biosorbent remain unsaturated during adsorption, but as the concentrations of heavy metal ions increase, the number of competing ions on the active sites increases, resulting in fewer binding sites available for complexation (Chithra et al. 2014).

Last but not least, contact time and agitation rate are among the significant parameters affecting the performance of *C. papaya* seed for heavy metal ions removal as an adsorbent. Ong et al. (2012) found that higher agitation rate and longer contact time promoted higher Zn (II) uptake. As the agitation rate increased, the resistance in the boundary layer decreased, allowing the papaya seed biosorbent to become more mobile. As a result, metal cations diffused more readily towards the binding sites on surfaces of the papaya seed biosorbents. Besides, Adie Gilbert et al. (2011) has reported a very high initial adsorption rate for Pb (II) and Cd (II) ions using defatted papaya seed biosorbent, with the rates of 13.8 to 44.4 mg/g.min, respectively. More heavy metal ions

**Table 4** Adsorption capacities of papaya seed biosorbent for the removal of heavy metal ions

Adsorbent	Adsorbate	Q <sub>m</sub> (mg/g)	Concentration	pH	Metal removal percentage (%)	References
Defatted papaya seeds	Cu <sup>2+</sup> and Pb <sup>2+</sup>	17.29 and 53.02	150 mg/L	–	97.55 and 99.96	Garba et al. (2016)
Papaya seeds	Cr (VI)	–	–	2	–	Rahmawati et al. (2016)
<i>C. papaya</i> seed	Cr (VI) and Ni (II) ions	5.85 and 5.58	0.4 g/ 100 mL and 1.0 g/ 100 mL	2 and 6	–	Chithra et al. (2014)
<i>C. papaya</i> seed	Zn ions	19.88	–	5	98.33	Ong et al. (2012)
Defatted papaya seeds	Pb <sup>2+</sup> and Cd <sup>2+</sup>	1666.67 and 1000	–	–	95	Adie Gilbert et al. (2011)
Papaya seeds	Mn (II) and Pb (II) ions	–	–	–	–	Egila et al. (2011)



were adsorbed onto the papaya seed biosorbent with increasing contact time. The author explained that because of their smaller ionic radius, Cd (II) ions had a higher adsorption rate than the Pb (II) ions.

Table 4 tabulates the summary of adsorption capacities of papaya seed biosorbent for different heavy metal ions removal processes.

### Adsorption isotherm of dyes and heavy metals removal using papaya seed adsorbents

An adsorption isotherm describes the properties of solute-surface interactions as well as the specific relationship between adsorbate concentration and the amount of adsorbate that accumulates on an adsorbent surface at a constant temperature (Li and Wang 2009). When the process reaches equilibrium, an adsorption isotherm can be used to determine the adsorbent capacity and to better understand the interaction between the adsorbate and the adsorbent. As a result, a variety of models have been used to explain the adsorption isotherms. Among these adsorption isotherms, the Langmuir and Freundlich isotherms are the most commonly used models by researchers.

The parameters of the isotherm models reveal significant information about adsorption mechanisms as well as adsorbent surface properties. The Langmuir model proposed by Langmuir (1918) has been widely used to describe the adsorption occurred on homogenous surface by monolayer sorption with a finite number of identical sites such as for the adsorption of 2,4,6-trichlorophenol on coconut husk-based activated carbon (Hameed et al. 2008). Adsorption is assumed to be a dynamic process in this model. The number of molecules being adsorbed will be equal to the number of molecules leaving the adsorbed state when the system reaches equilibrium. According to the Langmuir isotherm, adsorption occurs at homogeneous sites and forms a monolayer. The solid adsorbent has a limited adsorption capacity. Each active site is identical, which can only accommodate one solute molecule (monolayer adsorption) (Benjelloun et al. 2021). There is also no interaction between the molecules that have been adsorbed (Langmuir 1918). In this isotherm model, adsorbates are deposited as monolayers onto a surface with a finite number of adsorption sites and uniform adsorption energies, with no adsorbate transmigration across the surface plane (Baccar et al. 2010). Equation 1 is the nonlinear Langmuir equation that is used to describe the equilibrium adsorption results.

$$q_e = \frac{q_m K_a C_e}{1 + K_a C_e} \quad (1)$$

where  $q_e$  is the amount of dye adsorbed at equilibrium time (mg/g),  $C_e$  is the equilibrium concentration of dye in solution (mg/L),  $q_m$  is the maximum adsorption capacity (mg/g), and  $K_a$  is the isotherm constants for Langmuir (L/mg).

The linearized form of Langmuir isotherm that can be written as two different forms are expressed in Eq. 2 and Eq. 3, respectively:

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{K_a q_m} \quad (2)$$

The slope and intercept of the plot between  $C_e/q_e$  vs.  $C_e$  will give  $q_m$  and  $K_a$ , respectively:

$$\frac{1}{q_e} = \frac{1}{K_a q_m} \frac{1}{C_e} + \frac{1}{q_m} \quad (3)$$

In this form,  $q_m$  and  $K_a$  are plotted between  $1/q_e$  vs.  $1/C_e$ .

The separation factor ( $R_L$ ) of the Langmuir isotherm defined by Webber and Chakravorti (1974) is used as an indicator of favourable adsorption if  $0 < R_L < 1$ , unfavourable if  $R_L > 1$ , reversible if  $R_L = 1$  and irreversible if  $R_L = 0$ . The expression of  $R_L$  is written as:  $R_L = \frac{1}{(1 + K_L C_0)}$ . The adsorption capacity of the adsorbent, the interaction between the solute and solution, as well as the nature of adsorbed materials on the surface of the adsorbent can all be explained using isotherm models.

On the other hand, the Freundlich model that was empirically developed by Freundlich (1907) would be appropriate to describe the sorption of several compounds to heterogeneous surfaces or surfaces with sites of varying affinities, assuming that stronger binding sites are occupied first and the binding strength decreases as the degree of site occupancy increases (Srividya and Mohanty 2009; Silva et al. 2013; Yagub et al. 2014). There are various adsorption energy sites, but they all have the same entropy and are distributed according to an exponential law as a function of adsorption heat (Benjelloun et al. 2021). The density of sites also decreases exponentially (Freundlich 1907). In other word, Freundlich isotherms can be used to describe adsorption on heterogeneous surfaces as well as multilayer sorption formation. The principle is based on the assumption that adsorbate uptake is associated with a heterogeneous adsorbent surface (Ewecharoen et al. 2008). The logarithmic form of Freundlich model is given in Eq. 4:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (4)$$

where  $q_e$  is the amount of metal ion adsorbed at equilibrium time (mg/g),  $C_e$  is the equilibrium concentration of dye in solution (mg/L),  $K_f$  is the capacity of the adsorbent, and  $n$  is the intensity of adsorption constant for Freundlich.

This isotherm can be represented using the nonlinear equation or linear equation models (Freundlich 1907; Foo

and Hameed 2010; Sahin and Tapadia 2015; Qian et al 2018). The linearized forms of Freundlich are expressed in Eqs. 5 and 6.

$$Q_e = k_F(C_e)^{\frac{1}{n}} \quad (5)$$

$$\ln q_e = \ln K_f + \frac{1}{n}(\ln C_e) \quad (6)$$

Most of the results in the literature showed that the adsorption isotherms of dyes and heavy metals removal using papaya seed adsorbents were best fitted to the Langmuir isotherm compared to the Freundlich isotherm. Nasuha et al. (2011) has analysed the experimental data using both Langmuir and Freundlich adsorption models. The adsorption isotherm data for MB dye were best fitted to the Langmuir isotherm as the adsorption of MB dye on raw and esterified papaya seed has a homogeneous distribution on the active sites. It could be supported by the maximum monolayer adsorption capacity for both raw and esterified papaya seed with Langmuir isotherm (200 mg/g and 250 mg/g) fitting experimental data better than Freundlich isotherm (2.59 mg/g and 3.25 mg/g). Mounir et al. (2017) has also shown that the Langmuir isotherm has a higher correlation coefficient ( $R^2$ ) (99.4%) than the Freundlich model (95.4%) for the adsorption of MB dye using papaya seed biosorbent and the adsorption kinetic model follows a pseudo-second order with a maximum adsorption capacity of 52.28 mg/g. The results demonstrated that the adsorption process has occurred in a monolayer. On the other hand, the Freundlich isotherm model can be applied to adsorption on heterogeneous surfaces that have interactions between the adsorbed molecules, and it is not restricted to monolayer formation. Hameed (2009) who worked on the adsorption of MB dye on papaya seed reported that the experimental data fitted the best to the Langmuir model with a maximum adsorption capacity of 555.57 mg/g. The  $R^2$  value obtained for Langmuir isotherm and Freundlich isotherm was 0.9863 and 0.9507, respectively. Again, this indicated that the MB dye adsorption on papaya seed occurred as a homogeneous monolayer on the surface.

Furthermore, Ong et al. (2012) used the Langmuir, Freundlich and Brunauer–Emmett–Teller (BET) equations to investigate the equilibrium data of Zn adsorption using papaya seed biosorbent. The Langmuir isotherm produced the best correlation, with the  $R^2$  value of 0.979, while Freundlich and BET had  $R^2$  values of 0.842 and 0.804, respectively. The authors explained that the adsorption process occurred only at specific localized sites on the surface, and that the saturation coverage corresponded to full occupancy of these sites. Each site could only accommodate one molecule or atom at a time. There was no interaction occurring

between neighbouring adsorbed molecules or atoms. In addition, when fitting the experimental data of Pb (II) and Cd (II) ions adsorption onto defatted *C. papaya* biosorbent, Adie Gilbert et al. (2011) discovered the adsorption results followed the Freundlich model better than the Langmuir model. According to the authors, the adsorption process was heterogeneous. Hence, the fact that the defatted *C. papaya* seed biosorbent had multiple adsorption sites was further supported.

The process of adsorption is generally accompanied by surface adsorption as well as diffusion into the pores (Benjelloun et al. 2021). The adsorption kinetics play an important role in designing the adsorption experiments. According to Shroff and Vaidya (2011), adsorption kinetics is determined by a series of steps, which include the transfer of solutes to the adsorbent surface, transfer from the surface to the intraparticle active sites and retention on the active surfaces with adsorption, complexation or intraparticle precipitation mechanisms. When metal ions are removed by adsorption, it is particularly important to determine the process kinetics, as this can aid in the design of the process under real conditions (Bartczak et al. 2018). Kinetic studies are important in the adsorption process because they reveal the rate of adsorbate adsorption and regulate the residual process time (Demirbas et al. 2009). The pseudo-first-order, pseudo-second-order, and Elovich kinetic models have all been suggested as good representations of dye and heavy metal adsorption on adsorbents.

The adsorption of various dyes and heavy metals using papaya seeds adsorbents is widely explored using different types of adsorption isotherm, inclusive of pseudo-first-order, pseudo-second-order, intraparticle diffusion, and the Elovich model. A pseudo-first-order equation based on the Lagergren kinetic is the most commonly used equation for liquid–solid adsorption (Lagergren 1898). Rodrigues and Silva (2016) proposed the pseudo-first-order model in which an ordinary first-order differential equation (Eq. 7) was used to describe the adsorption kinetics of a species within an adsorbent particle.

$$\frac{dQ}{dt} = K_1(Q_e - Q_t) \quad (7)$$

As expressed by the differential equation (Eq. 7), the adsorption capacity is proportional to the “distance to equilibrium”, which is defined as the difference between the average concentration of the species adsorbed in equilibrium with the fluid phase and the final concentration of the species adsorbed in equilibrium with the fluid phase (Benjelloun et al. 2021). By integrating this differential equation at the boundary conditions,  $qt=0$  to  $t=0$  and  $qt=qt$  to  $t=t$ , the following pseudo-first-order Lagergren



equation is obtained in Eq. 8 (Çisfçi and Henden 2015; Moussout et al. 2018).

$$\ln(Q_e - Q_t) = \ln(Q_e) - K_1 t \quad (8)$$

where  $Q_e$  is the amount of adsorbate in the adsorbent at equilibrium (mg/g);  $Q_t$  is the amount of adsorbate in the adsorbent at time  $t$  (mg/g);  $K_1$  is rate constant of Lagergren's first order; and  $t$  is time of contact (min).

Ho's pseudo-second-order kinetics model, on the other hand, specifies the adsorption rate as the square of the difference between the equilibrium concentration of adsorbate and the concentration being absorbed (Ho and McKay 1999). The pseudo-second-order kinetics model is also known as "Blanchard's model". When the adsorption occurs at two surface sites, the second-order differential equation (Eq. 9) can be used to express it (Rout et al. 2015; Naderi et al. 2018).

$$\frac{dQ}{dt} = K_2(Q_e - Q_t)^2 \quad (9)$$

By integrating this differential equation at the boundary conditions,  $qt=0$  to  $t=0$  and  $q_t=q_t$  to  $t=t$ , the mathematical equation of the pseudo-second-order kinetic model is shown in Eq. 10 (Çisfçi and Henden 2015; Moussout et al. 2018).

$$\frac{t}{Q_t} = \frac{1}{K_2 Q_e^2} + \frac{1}{Q_e} t \quad (10)$$

where  $Q_e$  is the amount of adsorbate in the adsorbent at equilibrium (mg/g);  $Q_t$  is the amount of adsorbate in the adsorbent at time  $t$  (mg/g);  $K_2$  is rate constant of pseudo-second order; and  $t$  is time of contact (min).

The initial adsorption rate  $h$  (mg/(g.min)) is defined as follows:

$$h = K_2 Q_e^2 \quad (11)$$

The Elovich equation has been extensively used in the recent studies to determine the kinetics of gases adsorbing on solids as well as the adsorption of pollutants from aqueous solutions (Ho 2006). This model corresponds to the following equation (Eq. 12) (Chien and Clayton 1980):

$$Q_t = \beta \ln(\alpha\beta) + \ln(t) \quad (12)$$

where  $Q_t$  is the amount of adsorbate in the adsorbent at time  $t$  (mg/g);  $\beta$  refers to the number of sites available for adsorption;  $\alpha$  is the initial adsorption rate (mg/g.min); and  $t$  is time of contact (min).

Most of the results in the literature showed that the adsorption kinetics of dyes and heavy metals removal using papaya seed adsorbents was best fitted to pseudo-second-order kinetics and the Elovich models compared to pseudo-first-order kinetic model.

For example, Hameed (2009) found that the pseudo-second-order kinetics model was the best presentation of MB dye adsorption using papaya seed. It was observed that the  $R^2$  values for pseudo-first-order kinetics model (0.8486) was lower than pseudo-second-order kinetics model (0.9998) at 360 mg/L of initial MB dye concentration. The adsorption of MB dye onto defatted papaya seed was found to be exothermic and spontaneous by Unuabonah et al. (2009). The pseudo-first-order rate constant increased with increasing initial MB dye concentration, while the pseudo-second-order rate constant and the adsorption capacity of defatted papaya seed were observed to follow the same trend. The authors also discovered that there was an increasing deviation from linearity with increasing initial concentration of MB dye. The  $R^2$  values of 0.7397 and 1.0000 were obtained for pseudo-first-order and pseudo-second-order kinetics model, respectively, at an initial MB dye concentration of 200 mg/L. Consistent with the findings by Hameed (2009), the pseudo-second-order kinetics model best described the kinetic data obtained by Unuabonah et al. (2009). Pavan et al. (2014), on the other hand, studied the adsorption of crystal violet on papaya seed powder using a pseudo-second-order kinetics model, and the results indicated that the experimental data and pseudo-second-order model agreed very well. Pseudo-first order, pseudo-second order and the Elovich kinetics models were used to analyse the adsorption results in the study. At a crystal violet concentration of 40 mg/L, the  $R^2$  values were 0.955, 0.974 and 0.806 for pseudo-first order, pseudo-second order and the Elovich model, respectively. Based on the  $R^2$  values, the pseudo-second-order kinetics model was identified as the best fit for representing the adsorption of crystal violet on papaya seed.

Table 5 tabulates a list of adsorption isotherms and kinetics models for different adsorption processes of different pollutants.

## Papaya seed as coagulant or flocculant in coagulation–flocculation process

### Mechanism of coagulation–flocculation

Coagulation–flocculation is a standard pre-treatment process used in the industrial wastewater treatment plants to remove suspended and dissolved solids. It is normally used in conjunction with sedimentation to aid impurities coagulate and form flocs that settle to the bottom (Mazille and Sphuler 2020).

Coagulants and flocculants are chemicals that must be added at a specific dosage to wastewater for treatment. Coagulants of various types, such as inorganic and organic polymers, are used in wastewater treatment plants. Aluminium sulphate or alum( $Al_2(SO_4)_3$ ), iron chloride



**Table 5** List of adsorption isotherms and kinetic models for various adsorption processes using papaya seed adsorbent

Adsorbent	Adsorbate	Isotherm model	Kinetics model	References
Magnetized papaya seed	Synthetic MB	Langmuir	–	Jong et al. (2020)
Papaya seed	Synthetic Congo red (CR) from single solution	Langmuir–Freundlich	Pseudo-second order and Elovich	Fatombi et al. (2019)
Papaya seed	Synthetic CR from binary solution	Langmuir–Freundlich	Pseudo-second order and Elovich	Fatombi et al. (2019)
Papaya seed	Synthetic IC from binary solution	Langmuir–Freundlich	Pseudo-second order and Elovich	Fatombi et al. (2019)
Papaya seed	Synthetic Indigo carmine (IC) from single solution	Langmuir–Freundlich	Pseudo-second order and Elovich	Fatombi et al. (2019)
Defatted papaya seed	Synthetic Malachite green dye	Langmuir	–	Zaharaddeen et al. (2019)
Papaya seed	Synthetic MB	Langmuir	Pseudo-second order	Mounir et al. (2017)
Defatted Papaya seeds	Cu <sup>2+</sup> and Pb <sup>2+</sup>	Langmuir	–	Garba et al. (2016)
<i>C. papaya</i> seed	Cr (VI) and Ni (II) ions	Langmuir	Pseudo-second order	Chithra et al. (2014)
Papaya seed	Synthetic MB	Langmuir	Pseudo-second order	Ibrahim et al. (2014)
Formosa papaya seed	Synthetic crystal violet	Langmuir	Pseudo-second order	Pavan et al. (2014)
Papaya seed	Synthetic tartrazine and amarant	Langmuir	Pseudo-second order	Weber et al. (2014)
Papaya seed	Synthetic Procion Red	Langmuir	Pseudo-second order	Foletto et al. (2013)
Papaya seed	Synthetic MB	Langmuir	Pseudo-second order	Paz et al. (2013)
Papaya seed	Synthetic Tannery dye	Langmuir	Pseudo-second order	Weber et al. (2013)
<i>C. papaya</i> seed	Synthetic zinc	Langmuir	Pseudo-second order	Ong et al. (2012)
Defatted Papaya seeds	Pb <sup>2+</sup> and Cd <sup>2+</sup>	Freundlich	–	Adie Gilbert et al. (2011)
Esterified papaya seed	Synthetic MB	Langmuir	–	Nasuha et al. (2011)
Papaya seed	Synthetic MB	Langmuir	–	Nasuha et al. (2011)
Papaya seed	Synthetic MB	Langmuir	Pseudo-second order	Hameed (2009)
Papaya seed	Synthetic MB	–	Pseudo-second order	Unuabonah et al. (2009)

(FeCl<sub>3</sub>), polyaluminium chloride (PAC) are among the examples of inorganic coagulants that have been commonly used. On the other hand, organic coagulants include polyaluminium chlorohydrate (ACH), polyferric sulphate (PFS) and polyaluminium silicosulphate (PASS) (Kweiner Tetteh and Rathilal 2020). Alum is an inorganic coagulant and is used as a conventional coagulant. Alum, aluminium chloride, sodium aluminate, iron sulphate, iron chloride, and iron chloride sulphate are a few examples of chemical coagulants. These conventional chemical coagulants are effective in raw water with a wide pH and temperature range. In fact, they are less sensitive to low water temperatures and require lower dosages to achieve water treatment goals. According to International Water Association (IWA) (2021), the total dissolved solids (TDS) would be lower if a conventional chemical coagulant is used in the treatment.

Natural coagulants have been used to treat water in Africa, China and India for over 2,000 years (Asrafuz-zaman et al. 2011). They can be categorized into plant-based and non-plant-based. The plant-based natural coagulants derived from naturally available sources or waste materials include beans, cowpea, chitosan,

peanuts, *Plantago ovata*, *Leucaena leucocephala*, maize seed powder, *Moringa Oleifera*, *Vicia faba* and surjana seed powder (Patel and Vashi 2012; George and Chandrn 2018; Kristanda et al. 2020). Coagulants that are not derived from plants, such as chitosan and alginate, come from marine sources (Kristianto et al. 2018a). There has been a lot of interest in the synthesis of natural coagulants in recent years (Mohd Asharuddin et al. 2021). This is due to the discovery of natural coagulants as a replacement in sustainable water and wastewater treatment for waste minimization, prevention and renewability. According to Pricilla (2019), the use of natural coagulants can significantly reduce sludge handling costs because, unlike alum, they do not consume alkalinity, so pH adjustments can be omitted, resulting in additional cost savings. Furthermore, they are readily available, biodegradable and less harmful to the environment and human health (Martínez et al. 2019).

Coagulants and flocculants must be precisely calibrated to the specific composition of the water. Generally, coagulants are positively charged particles that are attracted to negatively charged suspended solids in turbid water. The positive charge of the proteins in papaya seeds, which is known





as cysteine protease, is appropriate for the purpose because coagulation is induced when the papaya seed coagulant coagulates with negatively charged particles, like silt, clay, bacteria and toxins (George and Chandrn 2018). The floc will settle and clarified water will be produced as a result of the adsorption and neutralization process. As described by Johnson et al. (2019), the coagulation process occurs based on mechanisms such as polymer bridging and charge neutralization (Fig. 5). The larger the floc produced, the heavier they are, and thus the shorter the time required for the coagulated particles to settle down. The basic processes that occur during the coagulation process are shown in Fig. 6.

### Papaya seed as natural coagulant

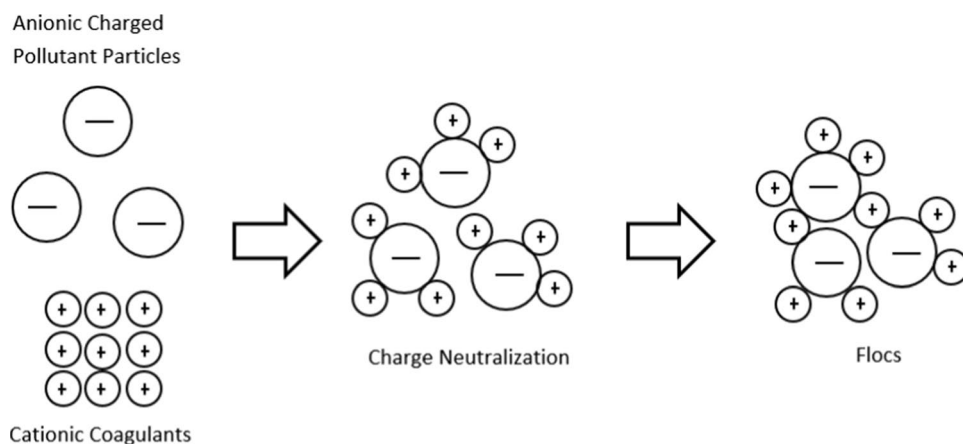
According to Unnisa and Bi (2018), cysteine protease is a highly water-soluble protein found in some *C. papaya* seeds that can be used as a coagulant for water and wastewater treatment. The authors reported that process variables including pH, turbidity, TDS, *E. coli* and coliform counts, all of which are important to the effectiveness of coagulation activity, should be optimized for use with *C. papaya*

seeds as natural coagulants (Unnisa and Bi 2018). Similarly, Kristianto et al. (2018a) reported that an optimal coagulation–flocculation process condition could be achieved using *C. papaya* seeds dosage at 0.57 g/L and water pH of 1.97, resulting in 84.77% of CR dye removal. At the dosage of 0.6 g/L, *C. papaya* seeds were found to remove 89.14% of turbidity and 90.29% of TDS (George and Chandrn 2018). Muda et al. (2020) reported that the turbidity removal efficiency was increased up to 95.5% with an optimum dosage of papaya seed coagulant at 130 mg/L. Based on the past researches, it was possible to achieve turbidity removal efficiency ranged between 84.77 to 95.5% using papaya seed as the natural coagulant.

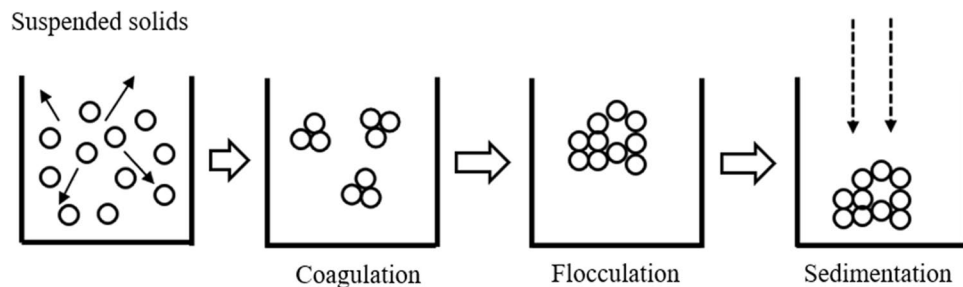
### Dye removal application

The dye removal efficiency demonstrated by papaya seed in the coagulation and flocculation process was between 80 and 84.77% (Amir et al. 2018; Kristianto et al. 2018a). pH and coagulant dosage were the two most important parameters affecting dye removal efficiency. Kristianto et al. (2018a) investigated the removal of Drimarene dark red

**Fig. 5** Process of charge neutralization



**Fig. 6** Simple illustration of coagulation, flocculation and sedimentation processes



**Table 6** Conditions and efficiency of synthetic dye removal using papaya seed coagulant

Synthetic Dye	Coagulant Dosage	pH	Removal (%)	Reference
Synthetic MB dye	0.4 g/ 200 ml	–	80	Amir et al. (2018)
Synthetic DDR	0.57 g/L	1.97	84.77	Kristianto et al. (2018a)



(DDR) dye from a wastewater treatment plant for synthetic textile colouring agents. Lower pH was found to enhance the dye removal efficiency by protonating the active coagulating agent in papaya seed powder, thus providing better electrostatic interactions with dyes (Kristianto et al. 2018a). The increase in dosage also resulted in better removal percentage until its optimum condition. Beyond the optimal condition, the removal percentage could be decreased due to colloidal re-stabilization (Szyguła et al. 2009; Zafar et al. 2015). Therefore, when adding coagulant to water, it is essential to introduce an acceptable coagulant dosage so that there are just enough polyelectrolytes to neutralize the particle, resulting in the zeta potential being nearly zero.

Table 6 shows the conditions and efficiency of synthetic dye removal using papaya seed coagulant.

### Turbidity and microorganisms removal application

The percentage of turbidity removed by papaya seed ranged between 41.89 and 100%. The major parameters influencing the turbidity removal efficiency were pH, coagulant dosage, mixing time and stirring speed. The relationship between different parameters is critical in identifying the optimum conditions for the coagulation process. George and Chandrn (2018) found that the water acidity level was reduced from 24 to 19 mg/L after treatment with *C. papaya* seeds for turbidity removal, which was comparable to raw water acidity level. In a separate study by Amran et al. (2021b), the turbidity removal percentage was increased from 43 to 90% as pH decreased from 6.5 to 4 with the deshelled *C. papaya* seed bio-coagulant dosage kept constant at 125 mg/L. Nevertheless, it is to note that the coagulation activity could be negatively affected by high doses of coagulant. This was particularly highlighted by Maurya and Daverey (2018), whereby the turbidity removal percentage reduced from 59 to 14.6% as the papaya seed coagulant dosage increased from 1.2 to 1.4 g/L. Likewise, at higher dosages of natural coagulants, TSS and COD removal efficiency also decreased (Unnisa and Bi 2018). On the other hand, George and Chandrn (2018) stated that an increase in stirring speed inhibited the shearing of initial floc formation, resulting in a reduction in turbidity from 35 NTU to 4.9 NTU at 80 rpm, enhancing the turbidity removal efficiency. The treated wastewater has a value of 4.9 NTU, which is lower than the recommended value of 5 NTU by the World Health Organization (WHO).

One of the interesting applications of *C. papaya* seeds in wastewater treatment includes the removal of microorganisms, such as heterotrophic bacteria, yeast and faecal coliforms. Papaya seed coagulant was capable to remove total coliform and *E. coli* in the range of 57.6% to 100% and 62.1 to 100%, respectively (Yongabi et al. 2011; Amir et al. 2018; Unnisa and Bi 2018; Amran et al. 2021a; Yimer

and Dame 2021). The papaya seeds contain proteolytic enzymes of papain and chymopapain which have antiviral, antifungal and antibacterial properties (Vij and Prashar 2015). Deshelled *C. papaya* seed-derived natural coagulant removed a surprisingly high percentage of *E. coli* and total coliform from the polluted water, despite the increase in organic content that may lead to an increase in the microbial activity (Antov et al. 2010). According to the findings, the natural coagulant extracted from deshelled papaya seeds possesses antibacterial properties. Researchers found that phenolic compounds can act as antibacterial agents that eliminate *E. coli* as well as total coliforms (Ifesan et al. 2013). Yongabi et al. (2011) have removed microorganisms using papaya seed in the range of 40 to 98%. Yimer and Dame (2021), on the other hand, discovered that a high total coliform removal percentage (96.32%) was achieved using papaya seed with defatted salt extract. A decrease in the total coliform removal was observed when the coagulant dosage was increased up to the optimum dosage of 20 ml. The trend of bacterial load reduction that was proportional to coagulant dosage was also reported by Unnisa and Bi (2018). The authors used papaya seed powder as the coagulant, and obtained persistent improvement in total coliform as the coagulant dosage increased by 5 mg/L intervals, and the best reduction was achieved with the optimum dosage of 15 mg/L. As a result, it was suggested that higher coagulant dosages were marginally more effective than lower dosages, which was related to the positive surface charge of the coagulant that could destabilize colloidal particles (Kakoi et al. 2016; Zaidi et al. 2019). Table 7 tabulates the summary of turbidity and microorganisms removal process using papaya seed coagulant.

### Conclusion

This review has unveiled the utilization of papaya seed from agricultural wastes as adsorbents or coagulants for the removal of various types of pollutants from wastewater. Surface modification has been widely investigated as a means of improving the performance and effectiveness of papaya seed, but detailed work on how the alteration of papaya seed's surface functional groups affects the adsorption and coagulation efficiency is limited in the literature. Papaya seed-based adsorbents demonstrated promising results in the adsorption process; however, information on using them for the simultaneous removal of co-existing pollutants is still scarce. Since industrial effluents contain a variety of pollutants, there is a need to further investigate papaya seeds as “general purpose” biosorbents capable of removing more than one pollutant at one time. Even though there is an increasing trend in tapping the full potential of papaya seed for wastewater



**Table 7** Turbidity and microorganisms removal from the textile industry and dye effluents using papaya seed coagulant

Source of water	Characteristics	Coagulant Dose	Turbidity Removal (%)	References
River water	57.6% and 62.1% of total coliform and <i>E. coli</i>	196 mg/L	–	Amran et al. (2021a)
Synthetic kaolin water	83% coagulating activity	196 mg/L	88	Amran et al. (2021b)
Tulte River water	Superior total coliform removal of 96.32%	20 mL	96.19	Yimer and Dame (2021)
Water samples collected from the upper basin of the Bogotá river	–	2500 ppm	66.45	Mrad et al. (2020)
Surabaya river water	1235 NTU to 9.82 NTU	0.2 g/L	99.94	Suprawito et al. (2020)
Surabaya river water	1235 NTU to 7.11 NTU	0.3 g/L	99.92	Suprawito et al. (2020)
Water samples collected from Sultan Ismail Water Treatment Plant, located near Universiti Teknologi Malaysia	94% coagulating activity	130 mg/L	95.5	Muda et al. (2020)
N/A	88% <i>E. coli</i> bacteria	100 seeds/ 100 L	90	Amir et al. (2018)
River water	TDS removal of 90.29%	0.6 g/L	89.14	George and Chandrn (2018)
Synthetic turbid water	–	1 g/L	93.6	Kristianto et al. (2018b)
Municipal wastewater	TSS removal of 66.66% and COD removal of 66.67%	0.8 g/L	41.89	Maurya and Daverey (2018)
Turbid water	100% removal of both <i>E. coli</i> and coliforms	0.2–0.6 mg/L	100	Unnisa and Bi (2018)
Water samples collected from Asia River, Ilorin, Kwara State	The trace metals were all below the detection limit	0.47 g	94.92	Eletta et al. (2016)
River water	TDS removal (90.29%)	0.6 g/L	89.47	Arya and Duithy (2015)
River water	98% removal of microorganisms ( <i>E. coli</i> , coliform, pseudomonas and yeast)	100 seeds/ 100L	90	Yongabi et al. (2011)

treatment processes, most of the work are reported based on laboratory-scale results. The feasibility of transforming this naturally abundant waste resource into a valuable adsorbent or bio-flocculant at the industrial scale remains unknown.

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