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Physico-chemical and Biological Techniques of Bisphenol A Removal in an Aqueous Solution

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ABSTRACT

Bisphenol A (BPA) is widely used in everyday life and can be found everywhere, including in the ecosystem and manufactured goods. BPA not only has a negative impact in low doses, but it also has biological and pathophysiological implications for obesity and hormonal effects. The objectives of this paper were to review the BPA removal technology and the factors that influence the BPA removal based on biological methods. BPA elimination from water is crucial for environmental protection, in terms of biological treatment. In addition, the future prospect of biological removal of BPA indicates that effective microorganism cultures could disturb the pathogen growth and increase composition rate of BPA. The biological technology by the implementation of microorganisms for the removal of BPA through break down of organic contaminants is straightforward, money saving, and widely acknowledged by the public.

Keywords: physico-chemical technology, biological technology, Bisphenol A; Effective Microbes (EM); Epoxy resins.

INTRODUCTION

Although bisphenol A (BPA) was synthesised in the 1890s, it is only found to be estrogenic in the early 1930 with lesser effectiveness compared to estradiol-17β by Dodds and Lawson. Later, Dodds revealed diethylstilboestrol as a valuable synthetic estrogen, BPA has no longer been applied as a drug. The International Union of Pure and Applied Chemistry (IUPAC) name for BPA is 2,2-bis(4-hydroxyphenyl) propane-4,4-(1-methylethlidene) bisphenol. The physical properties for BPA are white solid, molecular formula $C_{15}H_{16}O_2$, molecular weight, 228.29 g/mol, density 1.20 g/mL (at 25 °C), melting point 158–159 °C, and hardly soluble in water. BPA is a common chemical component in plastics and it has been used to harden the plastic strength properties.

In other words, BPA is widely used in daily life and is available around us either in the ecosystem and in manufactured goods. BPA is frequently used as a monomer in the manufacturing of polycarbonate plastics and epoxy resins. Despite its vast usage in manufacturing of products, BPA is categorized as an endocrine-disrupting compound (EDC). This is because, adsorption of BPA into one's body can contribute to interference of cellular pathway, immune toxicity, neurotoxicity and low sex-specific neurodevelopment. Thus, the existence of BPA and its derivatives in the scosystem has gained worlwide awareness. Biodegradation of BPA in the environment has been found to be dependent on such factors as pH, salinity, temperature, and oxygen availability. In industry, the manufacturing procedures of polysulfone, epoxy resin and some polyester resins caused the existence of BPA as an intermediate product.

According to Yuksel et al. [2013], the rate of BPA could be emitted by polycarbonate bottles is in the range 0.20–0.79 ng h^{-1} at room temperature, even if the bottles have never been used. Plus, the rate of BPA emitted in boiling water is 55 times larger compared to room temperature; hence, it has become a serious environmental threat. The manufacturing process of BPA is shown in Figure 1 and is as follows: Equation (1) depicted in the appearence of an acid catalyst (hydrochloric acid or cation exchange resins), the condensation of phenol and acetone generates BPA. Equation (2) shows the polycarbonate plastics manufacturing where almost 65% of BPA is consumed. The second largest use is epoxy resins, which accounts for about 30% of the consumption, as in Equation (3). Another major concern is the water pollution by chemicals since industries use large amount of water in the production of their products [Sonune & Ghate 2015; Crini and Badot 2015; Cox et al. 2015; Sharma et al. 2015; Rathoure and Dhatwalia 2016].

Endocrine disrupting compounds (EDCs) are exogenous elements that change and disturb the function of the endocrine system, thus leading to serious effect on individual health and surroundings [Zamri et al. 2021]. Furthermore, EDCs not only cause health problems to humans but also bring problems to the animals and their habitat through the contamination of EDCs in the water sources [Hasib and Othman 2020; Akhter et al. 2021]. Low fertility in humans and animals could be observed besides congenital malfunctions and cancer. For these reasons, research must be carried out to overcome this health and environmental issue. Microbes have played an important role in the biodegradation of inorganic, organic substance and nutrient cycle in our natural environments. During fermentation, the existence of microbes enables waste recycling process to occur while treating wastewater and boosting alternative energy production. In general, the heterotrophic bacteria obtain the energy to grow and synthesis new cells through carbonaceous organic matter degradation in wastewater effluent.

FATE OF BISPHENOL A

Volatilisation, photo-oxidation, adsorption, bioaccumulation, and chemical oxidation onto sediment particles are the examples of BPA released pathway into the environment. On the basis of the assessment and the process impact on the environment, microbial transformation and degradation processes are considered possible for successfully removing BPA from the environment. In the aquatic environment, BPA has half-life ranging from 2 to 7 days. The efficacy of biological wastewater treatment is often dependent on the metabolism of bacteria. However, it is also notable that the complexity of microbial structures and their compositions are highly influenced by the operating parameters during wastewater treatment. For example, the biomass structure of certain species influences the metabolic processes that may take place in the technological arrangement as well as the final standard of wastewater effluent.

Level of Bisphenol A concentration

The concentration levels of BPA from different sources are summarised in Table 1. The sources of BPA released from household sewage contribute the highest concentrations of BPA, while river sediment has the lowest concentration. The data used was reported between 2015 and 2018.

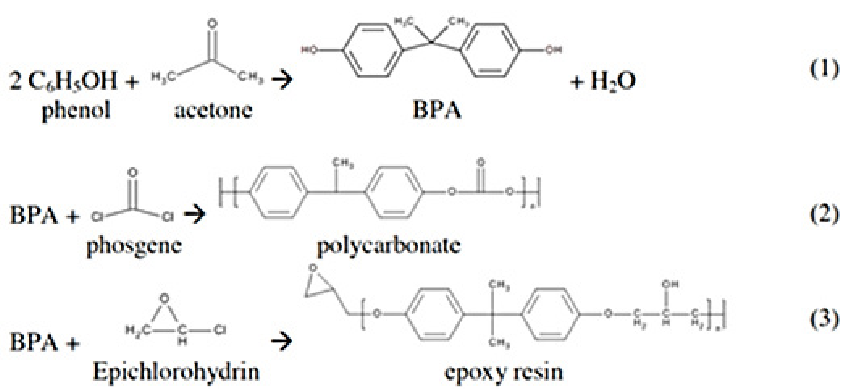

Figure 1. Production pathway of BPA [Sonune & Ghate, 2004]

Table 1. Concentration levels of BPA

Source of BPA released	Concentration	Reference
Stormwater	0–56 μg/L	Lu and Chen, 2018
River	0–12 mg/L	
Municipal wastewater treatment plants	0.01 μg/L–86 μg/L	
Plant	0.01–0.20 μg/L	
Industrial wastewaters	25–150 μg/L	Lee and Peart, 2000
Household sewage	25–75 mg/L	Sabrine et. al., 2018
Landfill leachates	0–172 μg/L	Yamamoto et al., 2001

According to Lu and Chen [2018], it was reported that integration of biochar and biofilter able to remove BPA from stormwater. The BPA concentration found in surface water was as high as 56 μg/L. In addition, the findings stated that the lowest adsorption capacity of BPA in stormwater occurred at pH 9 with 10 mg/L humic acid (HA). An experiment was also carried out at influent BPA concentration ranging from 0–12 mg/L to investigate the potential aerobic granules as well as the morphology and structure of species that exists in granules. It was found that the treatment of BPA using a sequencing reactor employing aerobic granules functioned with 8-h cycles manage to eliminate 97% of BPA with removal rate of 0.8 mg/g. As much as 0.01 μg/L to 86 μg/L of BPA was found in municipal wastewater treatment plant effluent; meanwhile, the bacterial concentrations in wastewater treatment plants are typically 109/ml. In general, the heterotrophic bacteria in wastewater effluent obtain their energy to grow and synthesise new cells through the degradation of carbonaceous organic matter. The toxicological effect of BPA on plants is important to observe the influence of BPA on plant survival and metabolism. In order to achieve such objective, plant tissues are usually undergone several processes namely: drying, grinding, methanol extraction, centrifugation, acetonitrile extraction, before the extracts were analysed using High Performance Liquid Chromatography (HPLC). It was found that approximately 90% of recovery rates was achieved when standard-added analyte was verified using this technique with the limits of detection (LOD) and quantification (LOQ) ranged from 0.01 to 0.20 μg/kg and 0.04 to 0.60 μg/kg respectively; thus, the suitability of this technique in determination of trace concentration of regular estrogen pollutants for instance BPA in plants was shown.

Effect of Bisphenol A

BPA pollutants have been identified due to their existence and persistence in the environment. It is an endocrine disruptor because BPA able to mimic the body's hormones and can affect with natural hormone secretion, production, transport, function, elimination and action [Chin et al. 2018]. Moreover, BPA also can imitate the function of estrogen and other hormones in the human body. Unfortunately, BPA can severely affect infants and young children. When BPA enters the body, it can initiate metabolic disorders in particular low sex-specific neurodevelopment [Gurmeet et al. 2014] immune toxicity, neurotoxicity, and interference with cellular pathways. Thus, the existence of BPA and its derivatives in the ecosystem has recently drawn worldwide awareness. According to Rivero et al. [2014], minimal exposure of BPA to laboratory animals shows increment in the size of the fetal mouse prostate. Other effects are increased in growth of post-natal, early sexual maturation in females, female offspring's mammary gland development is aroused, males' daily sperm generation and fertility are reduced, as well as disturbance in the function of immune system, transformations in the brain (increased the progesterone receptor mRNA), reduction in antioxidant enzymes, and behavioural consequences (involving hyperactivity, escalation in assertiveness, and lessened maternal behaviour) occur. These negative impacts show that despite having a generally short half-life, long-term exposure of BPA could accumulate to a certain concentration that is detrimental to health.

According to Chen et al. [2020], for efficient removal of BPA a β-cyclodextrin (β-CD) modified graphene oxide (CDGO) membrane that possessed high flux and adsorption was successfully established. Generally, vacuum filtration is used to fabricate the CDGO membranes. CDGO nanosheets was first staked on porous substrates in which the fabrication of CDGO nanosheets are done through chemically grafting β-CD molecules onto both sides of GO nanosheets. Due to the stable formation of β-CD molecules with BPA molecules via host-guest recognition, the planned CDGO membranes are highly efficient for BPA removal. It is critical to eliminate BPA because Asadgol et al. [2014] proved an increase in the risk of obesity in rodent is caused by BPA. This is believed to occur due to activity of BPA that triggers the brain and fat cells to control the deposition of adipose tissue and increased food consumption in rodents. Plus, further research also found that the exposure to low-dose BPA over a period of time can lead to cardiovascular difficulties which include angina, coronary artery heart disease, heart attack, peripheral artery disease and hypertension.

TECHNOLOGY OF BPA REMOVAL

Over the years, research has found a common method applied in BPA removal out of the water namely, biological degradation, chemical oxidation and membrane separation. Physical, chemical, and biological technologies have been widely discussed as methods of treatment. Among the treatment methods, there are flotation, oxidation, precipitation, solvent extraction, evaporation, ion exchange, carbon adsorption, phytoremediation, membrane filtration, biodegradation and electrochemistry.

Physical technology of BPA removal

Aluminium-based metal-organic framework / sodium alginate-chitosan (Al-MOF/SA-CS)

Some of the earliest research on low-dose BPA documented adverse effects in animals. BPA is a synthetic compound derived from carbon that contains two 4-hydroxyphenyl rings. It is able to affect the biological processes especially metabolic, thyroid hormone, and androgen system. Zifen Luo et al. [2019] provided a detailed explanation about effective removal of BPA from an aqueous solution using aluminium-based MOF/ sodium alginate-chitosan composite beads. On the basis of the study, BPA was successfully removed by utilizing a synthesized aluminium-based metal-organic framework (MOF)/sodium alginate-chitosan (Al-MOF/SA-CS) composite beads as an adsorbent.

In order to characterize the adsorbent, Fourier transform infrared spectroscopy (FTIR), scanning electron microscope (SEM), X-ray photoelectron spectroscopy (XPS), nitrogen adsorption-desorption isotherms and X-ray diffraction (XRD) were examined. The findings show that the porosity of the beads could be changed in the presence of CS. Plus, it is also believed that the total pore volume and larger surface area of AL-MOF/SA-CS beads is obtained in comparison with the beads without CS. On the basis of several characteristics such as good water stability, high adsorption properties, good recyclability, and ease in separation, AL-MOF/SA-CS composite beads are believed to be extremely effective at removing BPA from contaminated water. The exposure of BPA on food can occur through the migration of BPA from contact with food materials and is an alarming concern due to the toxicity and hormonal properties of BPA.

Membrane Filtration

Utilisation of membrane filtration in water purification treatment often limited due to the concern of membrane clogging [Sun et al., 2015] that shorten the life of membrane and filtration cycle. Besides, membrane fouling frequently occurs due to the existence of organic substances in wastewater, which interferes with the separation of low molecular weight organic micropollutants. In order to curb such problem, hybrid process is often chosen for BPA and other EDCs removal. Combination of membrane filtration and Fenton's process is one of the examples of hybrid system to remove BPA.

Nano Filtration

Nano Filtration (NF) has the ability to remove both adsorbed and soluble BPA. However, the efficiency of BPA removal by using NF is dependent on the concentration of BPA available that is adsorbed onto a suspended solid. The BPA adsorbed on the suspended solid eventually will increase the size of respective matter. If this happened, microfiltration (MF) would be enough for post- treatment purpose because MF is able to separate the adsorbed BPA on the suspended solid from the effluent. A comparison between MF and NF ceramic membrane was carried out for BPA removal from reactor effluent with immobilised biomass, conducted at a hydraulic retention time (HRT) of 1.5 h. The results found showed that suspended solids in high concentrations are observed in the effluent, and it is believed to originate from the adsorbed BPA and other hydrophobic compounds. Due to high concentration of solids, membrane fouling can occur in which can be used to study the susceptibility of ceramic membrane to fouling. The same method can also be used to find out the best membrane combination for BPA removal. MF membrane with pore size of 0.45 m could eliminate BPA that is retained, since BPA is immobilised on particulates. However, high BPA loading is present in the retentate from the membrane process; therefore, it is important to find the possible way out to curb the respective problem, such as by recycling the retentate back to the biological reactor.

Electrocatalytic

According to Ju et al. [2015] an electrocatalytic technique is often used in BPA degradation and separation from water. Carbon electrodes have been introduced in the electrochemical oxidation of BPA. It was found that polymerization of BPA occurred in the solution by a deposition of polymer film was observed in which caused inactivation of the carbon electrodes. Besides carbon electrodes, β-MnO_2 nanowires have superior mechanical consistency and BPA was successfully oxidised and broken down using this method. Nevertheless, the effects of the β-MnO_2 nanowires are suppressed by the occurence of humic acid and metal ions. As a result, ionic liquids (ILs) were used to enhance the electrodes' constancy and reusability.

Chemical technology of Bisphenol A removal

Polyphenol Oxidase

Removal by polymerisation of polycyclic aromatic hydrocarbons, chlorophenols and phenols were proven possible with peroxidase enzymes as catalyst. Previously, Kimura et al. [2015] has reported on quinone oxidation of BPA by polyphenol oxidase (PPO) before using chitosan beads to remove quinone derivatives. The oxidation of BPA by PPO was optimum at 40 °C and pH 7.0. Besides PPO, a microbial peroxidase enzyme, *Coprinus cinereus peroxidase*, was able to successfully remove BPA out of an aqueous solution within 30 minutes at optimum condition of pH 9 to 10 (40 °C) in the existence of H_2O_2 at mole ratio to BPA of 2:1 . Moreover, Sonoki et al. [2011] reported that they had made artificial tobacco plants, comprising a gene for lignin peroxidase which was generated in their roots. Sonoki et al. [2011] also claimed that lignin peroxidase produced by the plants has the ability to eliminate aqueous BPA four times more effectively than the control plants. In addition, tyrosinase is used in the presence of H_2O_2 to oxidise BPA to quinone. Chitosan gels, powders, and porous beads added to a BPA and tyrosinase solution can completely remove BPA. Adsorption of quinone derivatives on chitosan beads for example, can completely remove BPA within 4 to 7 hours. Multiple copper atoms that were possessed by laccases on its activation sites could also be used in the oxidation of BPA to quinone derivatives. However, enzyme inactivation could be the barrier in utilizing laccases enzyme.

Magnetic Vermiculite-Modified

According to Saleh et al. [2019], Magnetic vermiculite (MV) was used as a novel capable adsorbent to treat wastewater including BPA. MV was modified by poly(trimesoyl chloride-melamine) (MP) and synthesised before being applied in wastewater treatment. In the synthesis of MV-MP adsorbent, Fe_2O_3 nanoparticles were improved with trimesoyl chloride and melamine via interfacial polymerisation technique. Upon completion of interfacial polymerisation process, the morphology, and chemical properties of the adsorbent were analysed by FTIR and SEM, meanwhile the factorial design analysis was employed to examine the consequence of experimental factors on the yield of adsorption Langmuir isotherm were the better fit for BPA than the Freundlich isotherm based on the isotherm model investigation. Moreover, pseudo-second-order (PSO) kinetic model depicted a good association for the adsorption of BPA compared to psedo-first-order (PFO) model in the kinetic study. The thermodynamics study revealed that BPA adsorption on MV-MP was an impulsive and exothermic process at the temperatures tested. Plus, it was also found that the MV and MV-MP composites able to excellently remove BPA even after 7 reuse cycles with a highest adsorption capacity and adsorption/desorption capacity. By considering all the results, the synthesised MV-MP composite is a adsorbent with good potential for elimination of BPA from contaminated wastewater. Figure 2 shows the preparation steps of MV-MP adsorbent.

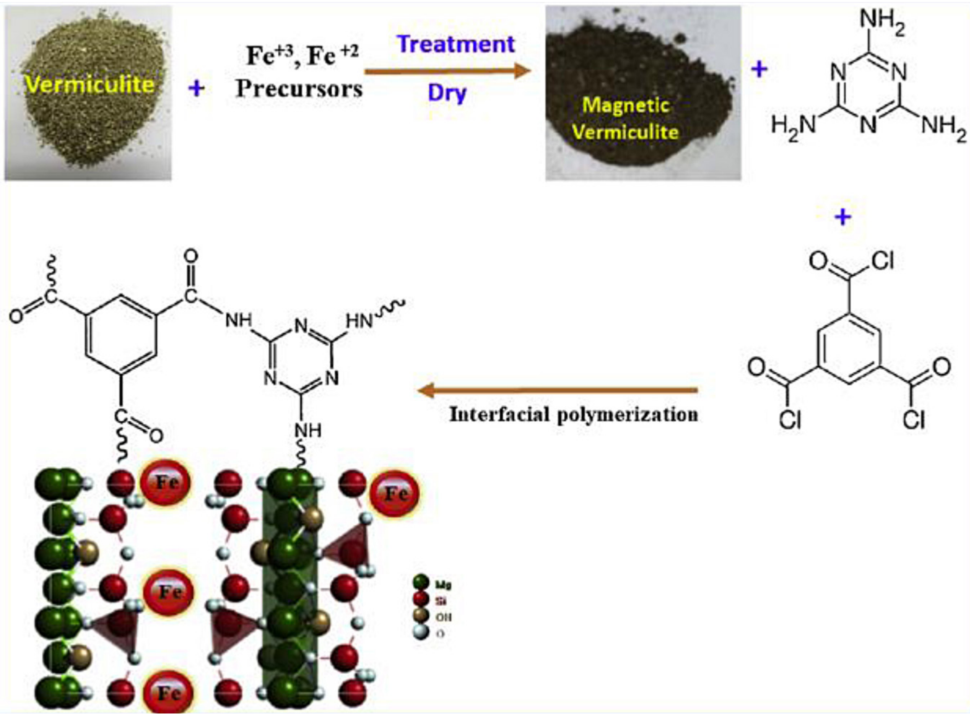

Figure 2. Preparation steps of magnetic vermiculite adsorbent [Saleh et al. 2019]

Electro-oxidised

Aravind et al. [2019] reported that non-biodegradable BPA was electro-oxidised in a membrane-less cell using a Ti-TiO_2/IrO_2/RuO_2 anode and a Ti cathode. The process parameters that were optimised were pH9, chloride concentration of 17 mM, practised current density of 20 mA cm^{-2}, and reaction time of 10 minutes. By combining OCl ions and OH radicals, BPA was electro-transformed into dihydroxy benzoic acid, dichloro-2-hydroxy acetophenone, 4-hydroxy benzoic acid, 2-hydroxy proponoic acid and trihydroxy benzene. Through a silico toxicity analysis, it was found that dihydroxy benzoic acid, dichloro-2-hydroxy acetophenone, and trihydroxy benzene have the mutagenic and carcinogenic properties. The electro-transformed biodegradability of the BPA solution was adjusted to 0.459.

Furthermore, by utilising a consortium of naphthalene-degrading microorganisms in a partially packed bed reactor for 50 hours able to degrade 91.9% of the electro-transformed BPA products. Moreover, the bacteria enzyme system aids in the

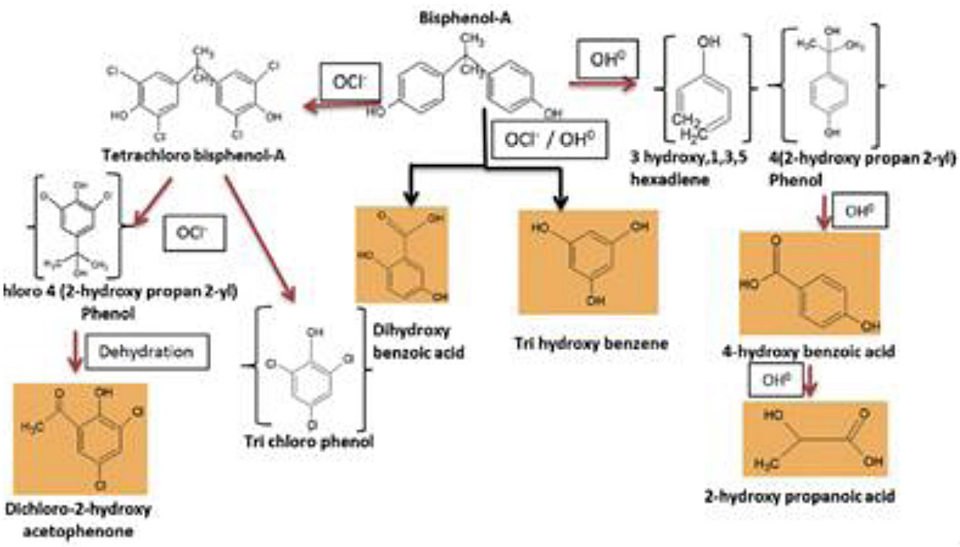

Figure 3. Proposed electrochemical degradation pathway for BPA [Aravind et al. 2019]

conversion of toxic intermediates produced during the electro-oxidation process into aromatic hydrocarbons such as acetopyruvate, maleyl acetate, fumarate, oxodipic acid, and malate. Through this method, approximately 91% and 56% of COD and TOC removal, respectively, was achieved. Furthermore, a silico toxicity analysis revealed that the final discharge has no toxicity and no estrogenic activity. Figure 3 shows the BPA electrochemical degradation process proposed.

Cu(II)-EDTA

According to Zhang et al. [2020], Cu(II)-EDTA has good stability even at wide range of pH (pH 3.0 to pH 12.0) and is hardly removed via the traditional method of precipitation. However, Fe(III) displacement/UV photolysis/alkaline precipitation [Fe(III)/UV/NaOH] was highlighted as an effective technique to remove Cu(II)-EDTA. Despite the possibility, a significant volume of Fe(III) is required in the process, which results in a high emissions of harmful sludge production. Fe(III) photochemistry is known to be ligand-dependent. Fe(III)-oxalate complexes, for example, are highly light-sensitive; even so, introducing oxalic acid to the Fe(III)/UV/NaOH process was beneficial. Apart from oxalic acid, acetylacetone (AA) can also be used as AA is considered as a effective chelating ligand for various metal types and an efficient photo-activator. Small dosage of AA ([AA]/[Cu]=1.5) into Fe(III)/UV/NaOH procedure for example, able to reduce Fe(III) dosage ([Fe]/[Cu]) from 10.4 to 3.2. From this finding, Fe(III) mixed with AA/UV/NaOH was observed to be a more eco-friendly and more effective method to treat metal-organic complexes, despite the concern on the recovery of heavy metals and precipitate sludge.

Silica Microspheres

A water-soluble azobenzene-containing 4-[(4-methacryloyloxy) phenylazo]benzene sulfonic acid is used as functional monomer in the surface polymerisation of photoresponsive surface molecular imprinting polymer (SMIP) using silica microsphere. Good response on photo properties, high specific affinity to BPA (maximum adsorption capacity of 6.96 μmol g^{-1}), and fast binding kinetics (binding constant: 2.47×10^4 M^{-1}) in aqueous media are among the advantages of SMIP microspheres. Plus, at 365 and 440 nm irradiation run alternately shows quantitatively binding and release of BPA by SMIP microspheres. According to Yang et al. [2014], the use of SMIP microspheres to recognise the BPA levels in tap and mineral water was discovered to be simple and quick.

Biological technology of Bisphenol A removal

Caco-2 Cells

Ungureanu et al. [2018] presented a study demonstrating the concentrations of BPA in cultured Caco-2 cells. It was found that BPA higher than 200 mM shows a cytotoxic effect in cells, meanwhile, BPA at lower concentration shows minimum toxic effect on the cells. The fact that the BPA concentrations cannot be toxic at a concentration below than 0.1 mM is because it is hard to conclude the severity of low dosage of BPA over a longer period of time, since the exposure period is no longer than 24 hours. It is critical to address the issue of BPA contamination in food and beverage packaging as soon as possible, as the current legislation imposes a limit of 0.6 mg/L (~ 0.26 mM) and portrayed a severe negative side effect even after only 24 hours of exposure.

Bacterial Strains

Sarma et al. [2019] stated that three bacterial strains (HAWD1, HAWD2, and HAWD3) and a bacterial consortium (BCC1) were isolated straight from the river sediment in order to remove BPA. El-Bestawy al. [2015] reported that organic loading significantly affects the nitrification process due to the removal of organic matter and nitrification often run simultaneously within the same reactor. Low nitrification process is often caused by high organic loading of wastewater. Moreover, in high organic loading wastewater, loss of ammonia can be observed due to assimilation by heterotrophs which leads to the inhibition of ammonia oxidation. Plus, consumption of oxygen during oxidation of organic matter reduced the availability of oxygen and resulted in low nitrification rate. Biological treatment in wastewater showed excellent efficiency to remove all nitrogen forms through aerobic digestion and usage of ammonia in the nitrification process helps in the complete removal of nitrogen. On the other hand, chemical treatment helps to precipitate most of phosphate content in wastewater.

Polymeric Microspheres

Xie et al. [2015] stated that an increase in BPA levels and its metabolite were observed during the biodegradation of BPA by acclimated activated sludge, imprinted polymeric microspheres (MIPMs). The greater concentration of BPA substrates and its metabolites presents depicted that the biodegradation efficiencies had increased and at the same time decrease the half-life of biodegradation process. MIPMs was observed to have the abilities to enhance degradation efficiencies at low levels of pollutants and with interferences like heavy metals and humic acid in wastewater. Moreover, the use of MIPMs as an enhancer in biodegradation of BPA were better compared to active carbon, a non-selective sorbent. Therefore, the use of MIPMs merged with activated sludge are easy, beneficial, and good for the environment but able to break down low-level pollutants in surface water.

Magnetic Nanoflower Biocatalyst

Han et al. [2019] stated that a magnetic nanoflower biocatalyst with a hierarchical flower-like surface of the core-shell magnetic composite microspheres was consistent with the organic component (horseradish peroxidase, HRP) and the inorganic component, through self-build in the phosphate buffered saline (PBS) solution. Through a series of characterisation technique, the pattern, structure and crystallisation characteristics of the magnetic nanoflowers were justified. The optimised findings revealed that the hierarchical structure of the magnetic nanoflowers formation conditions is able to increase the enzyme activity besides durability, stability, and reusability. Upon utilisation of the magnetic nanoflowers in elimination of BPA from wastewater, it was found that 92.1% of BPA can be removed with enzymatic activity at 183% compared to free HRP. In addition, reusability and reproducibility characteristics of magnetic nanoflowers showed that it has potential application in biocatalysts, despite time consuming procedures involved and low activity recovery.

Rhamnolipid

Chang et al. [2011] mentioned that the degradation of BPA was possible in the river environments due to aerobic condition. In laboratory scale test, biodegradation of BPA often depends on parameters such as sodium chloride, yeast extract, brij 30, cellulose, brij 35, and surfaction or rhamnolipid. However, it was observed that the addition of rhamnolipid leads to higher degradation of BPA compared to the other aforementioned factors. Besides rhamnolipid, inoculating sediment containing bacteria with potential to degrade BPA also helps in increasing the BPA biodegradation efficiency. Unfortunately, despite varying stimulating factors under anaerobic conditions for 140 days, the BPA in stream water was not reduced.

Trametes versicolor 8979

Brazkova et al. [2019] stated that *Trametes versicolor 8979* in submerged cultivation is able to completely remove BPA from the respective medium. *Trametes versicolor 8979* possesses high activity of laccase and manganese-dependent peroxidase which was believed to enhance the BPA removal. After 6 hours of incubation time at 28 °C and 220 rpm, BPA was found to have been almost totally eliminated from the reaction media.

Biogenic Manganese Oxides and Engineered Escherichia Coli Cells

Zhang et al. [2019] claimed that using a dual oxidation-action matrix of biogenic manganese oxides and engineered Escherichia coli cells comprising surface-expressed multicopper oxidase, it was possible to eliminate EDCs (CotA). The CotA gene was extracted from a Mn^{2+} oxidising bacterium and used to construct a fusion gene "inaQ-N/CotA" with an attaching motif in a Q-N from Pseudomonas syringae, which was then expressed in E. coli cells to display catalytic CotA on the cell surface. Under the Mn^{2+}-enriched culturing conditions for an extended period of time, the engineered cells can form microspherical, aggregated composites primarily composed of ramsdellite (MnO_2). Through the analysis, 7 and 10 degraded intermediates using the 13C isotope were identified from 13C-labeled BPA and 13C-labeled NP, respectively. The mineralisation pathways of BPA and NP were proven with the appearance of $13CO_2$ by the composite. Utilisation of *Caenorhabditis* elegans, an indicator organism in bioassays depicted the degradation processes eliminate the estrogenic activity of BPA and NP under acidic pH and at room temperature.

BIOLOGICAL TECHNOLOGY AS MAIN SELECTION FOR BISPHENOL A REMOVAL

Biological technology is well accepted by the public and the utilisation of microorganisms to biodegrade organic contaminants is easy and economical. Furthermore, microbiological processes are considered as a key element in technological developments for removing emergent contaminants from water.

Factors affecting removal

Bacterial metabolisms are able to identify the effectiveness of biological treatment of wastewater. The operating conditions of wastewater treatment frequently affect the species composition of microorganisms and their microbial structures. In addition, the biomass from the species structures determined the metabolic pathways; thus affecting the quality of effluent produced. Furthermore, the extracellular polymeric substances produced by the microorganisms were directly connected by the formation of microbial aggregates. Generally, EPS composed of biological products from cell lysis, excretion, shedding or material from cell surfaces, adsorption of substances from the environment [Abu Bakar et al. 2021]. The properties of EPS and biomass structure are associated to the composition of colloidal polysaccharide and protein in the EPS. Besides, the operating parameters used during treatment varied its production. The use of organics to yield EPS, preserve biomass structure, and protect cells from the adverse effects of free ammonium and free nitrous acid is required due to starvation over a long cycle length, a limited COD/N ratio, and a high nutrient load.

According to Hacıosmanoğlu et al. [2019] the adsorption capacity of phosphonated Halomonas Levan (PhHL) for BPA was almost unaffected under neutral and acidic pH conditions. However, it was noticed that the adsorption capacity of BPA drastically reduced upon increasing the solution to pH 9 and was dependent on the adsorbent material resulted from the interaction mechanisms between BPA, adsorbent material and the charges presents at the respective pH value. At 50 mg/L of BPA, the range of pH values between 5 to 6, and the first 30 minutes, was efficiently removed. Meanwhile, 99.3% of BPA was removed in 30 minutes (pH 5.5) and was totally eliminated after 60 minutes at 40 °C. Trace organic chemicals were observed to reduce at non-optimal PH values. This was believed to occur because of inactivation of enzyme which reduced the interaction rates between BPA and the enzyme.

The percentage removal for each different type of physical, chemical, and biological technology is shown in Table 2. On the basis of

Table 2. Percentage removal of BPA by different technologies

Technology	Method of removal	Percentage removal, %
Physical	Aluminium-Based Metal-Organic Framework (MOF)/Sodium Alginate-Chitosan (Al- MOF/SA-CS) Composite Beads	79
	Membrane Filtration	82
	Nano Filtration	78
	Photo-Fenton-Like Treatment	83
	Electrocatalytic	74
Chemical	Polyphenol Oxidase (PPO)	85
	Magnetic Vermiculite-Modified (MV)	83.5
	Electro Oxidized	87
	Cu(II)-Edta	84
	Silica Microspheres	81
Biological	Microalgal Biochar	93
	Caco-2 Cells	89
	Bacterial Strains (HAWD1, HAWD2, and HAWD3)	98
	Polymeric Microspheres (MIPMS)	91
	Magnetic Nanoflower Biocatalyst	94
	Rhamnolipid	92
	Trametes Versicolor 8979	91
	Biogenic Manganese Oxides And Engineered *Escherichia Coli* Cells	97

Table 2, the biological methods have the maximum percentage removal of BPA which proves their efficiency in not only removing BPA, but also removing other factors such as BOD, COD, TDS, TSS, pH, salinity and conductivity.

Effective microbe involvement

Microorganisms can be single-celled or multicellular, and can be made up of prokaryotic or eukaryotic cells. Bacteria, archaea, fungi (yeasts and moulds), algae, protozoa, and viruses are the primary features of microorganisms. Microorganisms play a variety of unique and complex roles within an ecosystem, including photosynthesis, waste breakdown, and infection of other organisms. Microorganisms benefit us in several ways, divided into four categories: commercial, medicinal, agricultural, and environmental. There are three general types of microbes: degenerative forming microorganisms, constructive regenerative microorganisms, and neutral opportunistic microorganisms. EM can also be classified as constructive regenerative microorganisms due to regenerative type. It can prevent decomposition of substances while keeping living organisms and environment healthy. The degenerative forming microorganisms behave in the opposite way to the constructive regenerative microorganisms. The neutral opportunistic type of microorganisms forms the largest group and adheres to the dominance principle in the system. Dr. Higa of the University of Ryukyus in Okinawa, Japan, discovered a unique group of naturally produced beneficial microbes capable of reviving, restoring, and preserving the environment in 1982. Later, Dr. Higa named this specific group Effective Microorganisms, better known as Effective Microbes (EM). EM are mixed cultures of valuable, naturally present microorganisms that can be used as inoculants to increase an ecosystem's microbial diversity. In other words, EM is a mixed culture of aerobic and anaerobic microorganisms that coexist to the mutual benefit of both through symbiosis. In addition, microbes are crucial to maintaining the ecological balance and exist everywhere in nature. The microbes are not harmful or pathogenic, have not been genetically or chemically modified (non-GMO), and are not chemically derived. Lactic acid bacteria and phototropic bacteria are the two main groups of microorganisms that form EM. Table 3 shows the effective microbes involved in the removal of BPA.

Table 3. Effective microbes used in removal of BPA

Effective microbe	Example
Lactic acid bacteria	*Lactobacillus plantarum, L. Casei, Streptoccus lactis*
Photosynthetic bacteria	*Rhodopseudomonas palustrus, Rhodobacter spaeroides*
Fungi	*Aspergillus oryzae, Penicillium sp., Mucor hiemalis*
Yeast	*Saccharomyces cerevisiae, Candida utilis*
Actinomycetes	*Streptomyces albus, S. griseus*

Bacteria exist in variation of forms, namely rods, spheres, and spirals with general width of individual cells in the range of 0.5 to 5.0 micrometres (μm; millionths of a metre). Bacteria are unicellular, but they frequently exist in sets, chains, tetrads, or clusters, with flagella or capsules. Gram staining often used to characterise bacteria to be classed as gram-positive (purple colour stain) or gram-negative (pink colour stain) depending on the existence of cell wall of bacteria.

Algae on the other hand are classified as eukaryotes, plant-like structure which contains chlorophyll and rigid cell walls, being able to carry out photosynthesis. Algae can be discovered in wet soil and aquatic ecosystems and can even be found as unicellular organisms up to 120 m in length. A cluster of algae can appear in many different of shapes, whereas a unicellular species can be spherical, rod-shaped, club-shaped, or spindle-shaped. Multicellular algae can be seen as filaments of cells joined together, but they can also be found in colonies as simple assemblages of single cells or as different types of cells with distinct functions.

Fungi are unicellular or multicellular eukaryotic organisms with rigid cell walls. Fungi can either be in microscopic size or larger structure like mushrooms and bracket fungi which can be found in soil or damp logs. Furthermore, fungi lack chlorophyll, rendering them incapable of photosynthesis. As a result, fungi absorb dissolved nutrients from their surroundings. Moulds are multicellular fungi that produce filamentous, microscopic structures, whereas yeasts are unicellular fungi.

Protozoa, also known as protozoans, are single-celled, eukaryotic microorganisms that can be oval or elongated in shape. Protozoa can have diameters as small as 1 mm and as large as 2,000 m, or 2 mm in the absence of cell walls. At some point in their life cycle, they can move and ingest food particles. Some phytoflagellate protozoa, on the other hand, are plant-like and obtain their energy through photosynthesis. The unicellular yeasts range in size or shape from spherical to egg-shaped to filamentous. Yeasts are well-known for their ability to ferment carbohydrates, producing alcohol and carbon dioxide in products such as wine and bread.

EM consist of five families of microorganisms: fungi, actinomycetes, yeast, photosynthetic bacteria and lactic acid bacteria. Bacteria are single-cell organisms. Some bacteria need oxygen to survive, and others do not. The lactic acid bacteria are notable for their disinfecting abilities. These lactic acid bacteria can both suppress harmful microorganisms and decompose organic molecules. Furthermore, *Fusarium*, a harmful fungus, can be inhibited in its reproduction by the lactic acid bacteria. *Lactobacillus plantarum, Streptoccus lactis*, and *Lactobacillus casei* are examples of the lactic acid bacteria. The photosynthetic bacteria are the bacteria that contribute in the activity of EM. These bacteria produce useful substances from root secretions, organic matter, and harmful gases such as hydrogen sulphide by using sunlight and soil heat as energy sources. The photosynthetic bacteria also aid in the better utilisation of sunlight, a process known as photosynthesis. These microorganisms produce metabolites that plants directly absorb. These photosynthetic bacteria also serve as nitrogen binders and increase the number of other bacteria. The examples of the photosynthetic bacteria are *Rhodopseudomonas palustrus* and *Rhodobacter spaeroides*. During the fermentation processes, fungi can rapidly degrade organic substances. Fungi can also suppress odours and protect against the damage caused by harmful insects. The examples of fungi are *Aspergillus oryzae*, *Penicillium sp*. and *Mucor hiemalis*. Yeasts are manufactured anti-microbial substances that aid in plant growth. Certain bacteria, including the lactic acid and actinomycete groups, consume yeast metabolites. Yeasts include *Saccharomyces cerevisiae* and *Candida utilis*. Actinomycetes suffocate unsafe fungi and bacteria that coincide with photosynthetic bacteria. The examples of actinomycetes are *Streptomyces albus* and *S. griseus*.

Mechanism of removal

Microalgae are an effective bio-indicator in ecotoxicity assessments because of their susceptibility to a variety of micropollutants such as EDCs and heavy metals. *Tetraselmis sp.* strain V2 is a marine alga used in the evaluation of BPA detoxification by strain G320. Cell density and dry weight method were performed to monitor the growth of algae in cultures. As a result, the initial toxicity of BPA was decreased upon inoculating strain G320 in MSM medium. According to Muszynski et al. [2015] substrate affinity of nutrient-removing bacteria reveals their species composition. Ammonia-oxidising bacteria (AOB) have higher diversity in domestic wastewater compared to a municipal wastewater treatment plant with *Nitrosomonas sp.* as the main player in nitrification. Temperature is considered as a vital factor that can affect abundancy of AOB while balancing the composition of *Nitrosospira sp.* and *Nitrosomonas sp*. in wastewater treatment plants besides the types of treatment system used.

Although autotrophic nitrifiers are in low abundance, successful nitrification depicted the removal of ammonium in a heterotrophic process. In the reactors of activated sludge processing high-organic tannery, ammonium-rich and coking wastewater, heterotrophic nitrifiers from the *Paracoccus, Thauera, Azoarcus* and *Comamonas* genera were found to grow. *Pseudomonas sp.* and *Paracoccus sp*. performed heterotrophic nitrification in laboratory-scale aerobic granules under high nitrogen loads. It was discovered that BPA biodegradation was greater in the no sediment samples in contrast to the sediment-containing samples, reducing the BPA bioavailability to microorganisms and thus slowing the biodegradation process.

FUTURE PROSPECTS OF BIOLOGICAL BISPHENOL A REMOVAL

From a future perspective, it is important to enhance the monitoring tools of environment and risk assessment. This can be done by identifying the BPA stress and characterisation of the dose-sensitivity interactions. In addition, the properties of BPA on plants need to be studied as there is limited research on the plant field, compared to human and environmental effects. In addition, the future prospects for biological methods of

BPA removal found that EM cultures can inhibit pathogens, hasten organic waste decomposition, rise the supply of inorganic nutrients and valuable organic compounds to plants, and boost the activity of valuable microorganisms (such as mycorrhizae and nitrogen fixing bacteria). This mixture increases the genetic susceptibility of soil, plants, water, humans, and animals. All over the world, EM developments are used in agriculture, crops, environmental clean-up (polluted waterways, lakes, and lagoons), and wellbeing industrial sectors. This review also discussed the benefits and drawbacks of the available technologies.

CONCLUSIONS

Microbes like bacteria and fungi biodegrade BPA into simpler substances which can be consumed by other mirooraganisms, thus keeping our planet clean. In the sewage treatment process, the utilisation of bacteria helps to degrade organic matter. In addition, EM composed of bacteria, yeast, and fungi (>80 strains) with diverse and versatile scope of application in wastewater treatment has been thoroughly tested and proven safe, causing no harm to humans and animals. The biological technology involved in the removal of BPA through the use of microorganisms for organic contaminant biodegradation is simple, cost-effective, and widely accepted by the public.

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