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Palladium catalyst for treatment of inorganic and organic pollutants in wastewater: a short review

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ABSTRACT

The over-exploitation of water sources combined with rapidly expanding population and urbanization has resulted in increased demand for clean water. Thus, alternatives are only choice to address the issue. The wastewater treatment serves as an ideal alternative to resolve the problem. But the expensive methods are a hurdle to this approach, particularly dealing with inorganic and organic pollutants. The treatment of these pollutants in polluted water can be solved by using palladium catalyst and different support systems for enhanced catalytic activity. In this review, a brief overview of about different Pd catalytic approach in curbing different pollutants has been summarized.

Keywords: Palladium catalyst; Wastewater; Pollutants; Alternatives approach

1. Introduction

Water scarcity has become a leading worldwide crisis with approximately half of a billion people throughout the world faced severe water depletion [1]. The situation has worsened when available freshwater remained inadequate to sustain increasing demand due to accelerating population growth, urbanization, and industrialization [2]. Water stress could be a host for various crises such as shortages in the food supply, hindering economic development and also caused widespread diseases [3,4]. The adverse effects of water shortage became much regions of the world that are facing poverty and could not afford for a good water treatment plant. The numerous great efforts such as enactment of stringent environmental regulation and policy, development of seawater desalination plant and improvement on sanitation technology have been invested to alleviate this issue [5–7]. Reusing wastewater is seen to be a potential practical solution to extend the availability of water supply and reducing dependency to freshwater resources [8].

National Research Council [9] estimated wastewater effluent discharge of approximately 12 billion gallons into surface water per day countrywide. Account for high wastewater generation rate, reusing it serves as a profound alternative to meet the growing water demand for industrial, domestic and agricultural purposes [9]. Trang et al. [10] highlighted the major risk of developing skin diseases, mainly eczema, and fungal infection among farmers who received exposure to wastewater in agriculture. In a worse case, prolonged exposure to high-level wastewater pollutant, for example, arsenic may increase the risk of cancer [11].

Since past decades, wastewater treatment has received a good deal of attention among the researchers. The treatments provide a pathway for utilizing the available resources and transform it into a usable water source for secondary purposes such as irrigation, industrial needs and drinking [9,12,13]. Thus, the treatment serves as a vital option to reduce the existing wastes, maintaining environmental quality and producing treated wastewater that is appropriate and safe for consumption [14]. However, the treatment via environmentally sound technologies, cost-efficient and less complexity remains a challenging problem [15]. Nonetheless, the implementation of treatment technologies are relatively difficult for rural and less developed region due to the topographical limitation

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and economical constrain [16,17]. Wastewater treatment for a large scale application requires costly sophisticated equipment and high usage of energy. In addition, existing conventional treatment technologies such as flocculation, filtration, and precipitation are less effective as they merely dispose of the waste. Therefore, it is essential to develop a cost-effective treatment combined with the complete destruction of pollutants as well as less hassle in recovery.

In this context, catalysis is one of a novel approach that satisfies the requirements of cost-effective treatments [18]. This approach employs the thrust of catalysis where the only limited amount of catalyst is required to enhance the rate of reaction and transform the reactant into benign end-product under mild operating conditions [19]. Various catalytic methods using Pd, Pt, Ru, Rh, Ni, and Ir have been investigated extensively. Among the catalysts, Pd has been studied particularly for hydrogenation of organic and inorganic pollutants.

Palladium catalyst has been synthesized in many various ways based on the literature. Based on the Scopus database, around 611 articles have been published with the searching keywords of palladium, catalyst, and water treatment. Besides that, the publication in this subject area also increases from 9.74% in 2000 to 90.26% in 2018. The publication of this subject area is most seen appeared in the Journal of Applied Catalysis B Environmental, Environmental Science and Technology, Water Research, Journal of Hazardous Materials, Catalysis Today, Chemosphere and Applied Catalysis A general.

The already published reviews about palladium catalyst are shown in Table 1. The review from Nowicka et al. [20] and Alshammari et al. [22] are more focusing on bimetallic catalyst involving palladium. The review from Nowicka et al. [20] is focusing towards the structure of bimetallic, factor affecting the catalytic activity, preparation and characterization of the catalyst. Based on the review from Siddiqi and Husen [21], the focusing area is towards biosynthesis of palladium. The topics on structural feature and method to the synthesis of catalyst are highlighted in the review from Alshammari et al. [22]. In the review from Chaplin et al. [23], the authors are aiming to the topics of proposed reaction pathways for important classes of contaminants, rates of contaminant reduction with different catalyst formulations, long-term sustainability with respect to natural water foulants, microbial fouling processes, and fouling prevention and regeneration strategies and technology applications. All these review papers also included the application of the catalyst.

Table 1
Published reviews about palladium catalyst

Published reviews	References
Designing Pd-based supported bimetallic catalysts for environmental applications	[20]
Green synthesis, characterization and uses of palladium/platinum nanoparticles	[21]
Bimetallic catalysts containing gold and palladium for environmentally important reactions	[22]
Critical review of Pd-based catalytic treatment of priority contaminants in water	[23]

In this review work, the focusing topics are the pollutant in water, palladium catalyst for wastewater treatment, material support for palladium catalyst and performance of palladium catalytic activity with different material support. This review outlines the development in the catalytic treatment of wastewater using Pd catalyst, the insight on fundamental aspects of Pd and its application in wastewater treatment. The overview of water treatment and recent development in treatment technologies have been reported in this review. The pollutants present in wastewater with a central focus on inorganic (ClO_3^-, ClO_4^-, BrO_3^-) and organic (trichloroethylene, perchloroethene, azo dyes) pollutants are also highlighted in this review. Furthermore, the future impact of Pd catalyst has been described briefly.

2. Pollutants in water

2.1. Wastewater composition

Wastewater was acclaimed to contain a high concentration of essential micronutrients that helped to increase soil fertility, induced crop growth and productivity and reduced fertilizer usage [24–32]. These attractive characteristics encouraged farmers to reuse wastewater directly or indirectly for agricultural irrigation purpose [33,34]. However, wastewater, either treated or untreated, poses various threats to public health and environment due to the presence of toxic chemicals and biological contaminants such as heavy metals, inorganic and organic chemicals as well as pathogenic contaminants [35].

Contaminants present in wastewater were much complex compared to groundwater or drinking water contaminants [36]. Significant variation in the complexity of wastewater compositions was much attributed to its source, norm of community and environmental conditions [37]. The composition of domestic wastewater, for instance, is differed to the composition of agricultural wastewater the large part of it consists of nitrogen and phosphorus [38]. Generally, wastewater is comprised of several components including organic pollutants (azodyes, trichloroethylene, perchloroethene), inorganic pollutants (nitrate, bromates, chlorates), heavy metals (mercury, lead, cadmium) and pathogenic contaminants. The contamination of these wastewater pollutants into surface water creates a serious issue that linked its toxicity to the aquatic environment and ecological system [39].

Apart from aforementioned contaminants, emerging contaminants (ECs) also constitutes the wastewater. Emerging contaminants were classified as a newer class of compounds that were unregulated, previously not known and have appeared only recently [40]. Several examples of ECs are endocrine disrupters, pharmaceuticals, flame retardants, artificial sweeteners, industrial additives, personal care products and these pollutants list were expected to be growing as more chemical compounds are continuously being produced [40]. According to US EPA [41], the exposure of ECs even at very low level may demonstrate low acute toxicity and cause significant reproductive effects. The elimination of these pollutants were considered to be intricate as it is water soluble and have poor degradability [42]. ECs are typically introduced to the environment by discharging of effluent from the sewage to receiving watersheds, thus lead to contamination of environment [43]. Exhaustive studies were subjected in the removal of these contaminants by a wide range of methods such as biological, physical and chemical treatment. Among the extensively used treatment technologies include adsorption, catalytic and photocatalytic treatment, ozonation, flocculation, sedimentation and filtration [44–48].

2.2. Inorganic pollutants

The composition of wastewater is considered to be complex with a various cocktail of pollutants [49,50]. The increase in complexity of wastewater composition is contributed by several factors such as population agglomeration and socio-economic status of the population [51]. Generally, these pollutants can be categorized into inorganic and organic pollutants.

The inorganic pollutant is a non-carbonaceous entity that pollutes the water [52]. It can be classified on the basis of its origin (point source and nonpoint source) or (anthropogenic and natural source) [53]. Inorganic pollutants from different sources are summarized in Table 2.

Table 2
Inorganic pollutants in wastewater

Pollutant	Origin	Reference(s)
Selenium, selenate and selenite	By-product of ore refining, agricultural irrigation, metal smelting	[50,51]
Nitrates and nitrites	Inorganic fertilizers and pesticides, industrial effluents and leachate from landfills	[40,52–55]
Phosphate	Inorganic fertilizer	[56,57]
Chlorate, chlorite and perchlorate	Disinfection by-products, pesticide	[58,59]
Bromates	Municipal waste incinerators, chemical industry, disinfection by-product from ozonated water	[60–62]
Fluorides	Naturally occurring element, founds naturally in mineral, water, foods and soil. Also originate from industry and volcanic glass	[63–65]
Sulfate	Naturally occurring minerals; used as coagulant in water treatment; industrial wastes discharged	[66,67]
Arsenic	Widely distributed in earth's crust, mineral and ores, industrial effluent	[68–71]

2.3. Organic pollutants

The term 'organic pollutants' are comprised of carbon in its backbone structure in which it is emitted into water sources [52]. It is one of the most common types of pollutants that constitutes the wastewater. The wastewater from textile industries, for instance, are highly comprised of dyes which are 50% azoic dyes [54]. These dyes exhibit hazardous impact and removal of them is challenging owing to their complex structure [55]. An azo dye, Congo red are claimed to be carcinogenic and cause toxicity to aquatic life [56–59]. These dyes are discharged from the industrial's effluent to nearby surface water leading to the potential transfer of this pollutant. The fact of the presence of these pollutants in wastewater reflects the inadequacy of a water treatment system or the lack of efficiency of water treatment in the industry itself. Organic pollutants from different sources are summarized in Table 3.

3. Palladium

Heterogeneous catalysis using palladium-based catalyst is commonly applied in various industries owing to its

Table 3
Organic pollutants in wastewater

Pollutant	Origin	Reference(s)
Dyes	Textile industries	[78]
Polychlorinated biphenyls (PCBs)	Plasticizers; coolants; used in capacitors and transformers	[79,80]
Polybrominated biphenyls (PBBs)	Flame retardants in electronic products	[80]
Polyaromatic hydrocarbon (PAH)	Petroleum coke; sediments	[81,82]
Trihalomethanes	By-product of drinking water disinfection	[59]
Trichloroethylene (TCE)	Industrial solvent	[83–85]
Perchloroethene (PCE)	Industrial solvent; used during reforming of hydrocarbons in petroleum refinery; Insulating fluid in electric transformers	[85–87]
Organochlorine pesticide (Aldrin, lindane, methoxychlor, dieldrin)	Pesticide	[82,88]
Phenolic substances	Disinfectant, petroleum refining; insecticides; herbicides	[82,89–93]
Endocrine disrupting chemicals (estrogens)	Animal manure, oral contraceptives, wastewater discharged from drinking water treatment plants	[94,95]

easiness in recycling and recovery of the catalytic material [60,61]. This transition metal resembles platinum in its chemical properties and is often applied as an alternative for platinum in heterogeneous catalysis [62]. Most notable application of palladium was in catalytic converters for automobiles [63]. Palladium's significant feature lies at its hydrogen adsorption properties which makes it an ideal catalyst for reduction process [64]. This remarkable characteristic is primarily due to its ability to form a strong affinity with hydrogen and also corrosion resistance property [65–67].

In catalysis, palladium has been extensively studied in multiple aspects to fully utilize its potential and optimize catalytic performance with great concern is paid on understanding its surface morphology [68,69]. Palladium is often customized to smaller particle size to achieve a high distribution of metal particles [70,71]. In recent years, development of highly branched catalyst has received enormous attention as it offered relatively high surface area [72]. Watt et al. [73], highlighted the development of highly branched palladium nanostructures for hydrogenation application of nitrobenzene to aniline. In the study, they had successfully synthesized extensive branches of palladium nanostructures by varying the nature of organic stabilizer system to control the surface morphology.

Watt et al. [73] stated that formation of highly branched nanostructures have resulted from the ultrafast growth of tripod-shaped intermediate in indefinite crystallographic direction. Results of the study showed palladium nanostructures exhibit good catalytic activity in the hydrogenation of nitrobenzene to aniline in 20 minutes reaction. The impact of this study has provided better insight on manipulation of palladium surface morphology to achieve higher catalytic activity and created an opportunity to explore deeper understanding on the effect of morphological changes to palladium's catalytic performance.

In another perspective, palladium also has been studied in the aspect of its interaction with a support system. Different utilization of support material, preparation procedures, and varying catalyst loading resulted in a different degree of enhancement in catalytic activity [74,75].

4. Palladium catalyst in water treatment

Catalysis is an excellent destructive technology that served as a very promising alternative for degradation of pollutants in wastewater by eliminating the toxic pollutants and transfer them into benign end-product [19]. With numerous development of ideal catalysts nowadays, palladium remains the preferred catalytic material employed in a diverse scope of applications ranging from industrial chemical production, oil refining, environmental protection as in automobile and air emission control as well as water purification system [76–81]. Using palladium as a catalyst is not a new idea as over the past decades, palladium has received immense interest among scientists to study its effects in abatement of water contaminants [82–92].

Among reactions subjected to this noble metal, palladium catalyst has been most favored in reductive reaction, particularly reaction involving hydrogen as the reductant [93–95]. As first reported by Conrad et al. [96], palladium has the ability to activate the hydrogen by easily dissociates the H_2 molecules into adsorbed surface hydrogen. The adsorption of hydrogen at room temperature and atmospheric pressure forms palladium hydride (PdH_x) [64]. Its superior properties to form hydride under ambient conditions are related to small or no activation energy barrier during dissociation of H_2 molecules on Pd surface [97]. This property is different from other metals where high pressures or elevated temperatures are applied to supply energy input in order to overcome the activation energy barrier. Furthermore, the surface of the catalyst is stable that interaction between hydrogen and Pd on the surface does not weaken the Pd-Pd bond [98]. All these features made palladium a promising catalyst for an efficient reduction of water contaminants.

The use of Pd on its own exhibits a good catalytic activity, however, the performance can be enhanced by immobilizing the metal onto the support material. In this context, the support is categorized into two types.

4.1. Inorganic support

In addition to the active metal phase, the choice of support material also plays a key role in optimizing the catalytic performance to maximum activity. Generally, the activity of supported Pd catalysts, specifically in water treatment, is influenced by the nature of the metal phase, preparation method properties of the support material, Pd precursor and also the nature of water matrix [99–101]. The support provides a high surface area, influencing dispersion of active phase, improving selectivity, stability and increasing resistance toward deactivation [102]. The inorganic support such as Al_2O_3, SiO_2, TiO_2, and zeolite are extensively used to assist Pd catalyst. Recently, Śrębowata et al. [103] developed catalyst by impregnating the Pd with microporous zeolite as support material for the removal of trichloroethylene from drinking water. The importance of the support material has been emphasized where the acidic and textural properties of zeolites are found to influence the Pd dispersion and Pd^0/Pd^{2+} ratio, hence impacted on the performance of the catalyst. The removal of trichloroethylene (TCE) has shown to be significantly effective in the presence of uniformly dispersed Pd species. The higher amount catalyst used contributed to 90% removal of TCE in 10 min reaction.

In another study using faujasite zeolite as a support, Soares et al. [98] studied the catalytic reduction of bromate over various monometallic and bimetallic catalysts under hydrogen at room temperature and atmospheric pressure. The results showed that all the bromate was completely removed from the water in less 10 min and are enhanced by the presence of metal in the zeolite structure. The study also confirmed that Pd is the best metal choice, owing to its properties of high H_2 adsorption capacity, with Cu-Pd catalyst emerged as the most promising catalyst. Despite the encouraging performance of the catalyst, the author observed a pronounced decrease in the specific surface area of the parent zeolites after metals are incorporated into the support. This phenomenon was attributed to the blockage of the zeolite's micropores by the metals [101].

Shin et al. [104] utilized Pd catalyst supported on TiO_2 for the treatment of wastewater contaminated by nitrate. The Pd/TiO_2 catalyst had shown to influence the selectivity toward the production of harmless N_2 production (78.6%–93.6% and 57.8%–90.9%) over harmful NH_3 when there was both a decrease in H_2 flow rate and an increase in NO_2^- concentration. In a similar study using Al_3O_2 as support, Zhao et al. [105] have recently reported the selectivity toward ammonium formation increased greatly (28 µmol L^{-1} after exhaustion of <5 µmol L^{-1} nitrite) when approaching complete conversion of nitrite. Pd/Al_2O_3 catalyst not only has been employed in nitrite reduction process but has been applied in catalytic reduction of bromate [106]. The study reported an increase in Pd loading amount (0.48–4.95 wt.%) would enhance the increase of initial bromate reduction rate from 1.6 to 25.1 mM g Cat^{-1} h^{-1}. The pH of bromate was also affected the bromate reduction where the higher catalytic reduction was achieved at pH 5.6.

In another study, Wang et al. [107] synthesized a core-shell support material consisting of nonporous silica core decorated with Pd nanoparticles and further encapsulated with mesoporous silica (SiO_2@Pd@mSiO_2) in the reduction of bromate (BrO_3^-), a drinking water contaminant produced during the disinfection process. The catalyst was found to be promising for application of water purification where the support developed significantly enhanced Pd catalytic activity compared to unsupported Pd nanoparticles and SiO_2@Pd.

4.2. Organic support

Organic materials such as resin, carbon, graphene, chitosan and various polymeric substances are several common organic supports used with Pd catalyst for water purification [108–111]. Due to its high catalytic activity, excellent stability and flexibility and also extensive supplies of raw materials for the production of the support, the use of carbon are recognized as a promising catalyst support for Pd [112]. The catalytic performance of Pd loaded on activated carbon catalyst (Pd/AC) was investigated by Díaz et al. [81] in the removal of phenol for the treatment of phenolic wastewaters by using two different catalysts (commercial and home-made Pd/AC). The catalyst achieved its highest phenol conversion values when using $PdCl_2$ as catalyst precursor. Furthermore, the study had found that the application this catalytic system increases the selectivity toward the formation of cyclohexanol, a less harmful reaction product.

The usage of graphene as support for palladium is accounted to its remarkable properties such as high surface area, excellent thermal, chemical and mechanical stability as well as flexibility [113]. Owing to these properties, Chang et al. [108] had developed a graphene-based Pd catalyst (Pd/G) for catalytic hydrodechlorination of chlorophenols via reduction of Pd(II)/Graphene Oxide (Pd(II)/GO) to Pd/G by microwave irradiation. The catalyst was found to show highest catalytic performance than Pd/Y (Pd supported on zeolite) and Pd/MCM–41 (Pd supported on mesoporous material) with the complete decomposition of all chlorophenols within 2 h in 3% Pd/G catalyst. In addition, the catalyst could be recycled several times without obvious loss of catalytic activity, hence implied its efficiency for remediation of chlorophenols. Li et al. [114] utilized nitrogen-doped graphene to support Pd nanoparticles in the removal of halogenated emerging contaminants, TBBPA (tetrabromobisphenol A). The study showed that the Pd supported on nitrogen-doped graphene (Pd/NG) demonstrated better catalytic performance than Pd supported on carbon and graphene (Pd/C and Pd/G) for removal of TBBPA. The Pd/NG catalyst showed the higher rate TBBPA dehalogenation (26.7% and 39.0%) compared to Pd/C and Pd/G.

Vincent et al. [115] had synthesized chitosan sorbent as a catalyst support (Pd/Chitosan) for 2-chlorophenol dehalogenation. The study revealed that the stability of chitosan in acidic solution enhanced by cross-linking the chitosan using glutaraldehyde. However, the catalytic activity of Pd/Chitosan was found to appear lower than Pd/C or mineral materials and thus, required optimization process of chitosan.

5. Future prospect

The discussion of this review indicates significant advancement in palladium catalyst used for water treatment with respect to the removal of inorganic and organic pollutants. The extensive applications of palladium in the heterogeneous catalytic process have shown its capability to be an ideal catalyst for water treatment where it provides greater efficiency in degradation of water pollutants, simple recovery of catalytic material and also cost-effective with the only small quantity of palladium is required for the reaction. However, palladium catalyst intended for wastewater treatment is susceptible to a vast range of complex contaminant matrixes that may exert a synergistic effect and interfere with catalytic activity. Considering of the ever-increasing amount of pollutant with complex composition entering a water body, new studies must be developed to contribute on the enhancement of palladium's structural stability and ensure a good catalytic performance even at the minute amount. Although the central focus of this review is specifically on the degradation of inorganic and organic pollutant, it is also nearly as crucial to study in depth on the adverse effect of different classes of pollutants such as emerging contaminants (pharmaceuticals, endocrine disruptor contaminants).

Besides, further, improvement must be made to increase palladium resistance towards sintering which reduces the surface area available for catalytic reaction. Hence, lowers the catalytic reaction of palladium. Although sintering is inevitable for certain processes such as calcination at elevated temperature, it can be suppressed by supporting the palladium with suitable support material. It is worth to denote that the choice of support materials also play its key role in assisting a catalytic reaction. A support material can be tailored accounting to the target material to be degraded. Studies on understanding the interaction between palladium and its support are necessary to provide a better insight on their influence in the reaction and increase selectivity toward target pollutant.

6. Conclusion

The severe water scarcity demands immediate action be taken to alleviate the issue. Wastewater treatment serves as an alternative measure to provide clean water either for potable or non-potable uses by means of purification method. Catalytic treatment method through utilization of palladium as an ideal catalyst have received enormous attention as this technology can efficiently degrade the pollutant without producing harmful by-product. The palladium catalyst can be tailored with various support materials to enhance the performance and increase the catalytic efficiency. The catalyst, as shown from previous researchers demonstrated an excellent reduction of water pollutants and thus, has outstanding potential for application in water treatment.

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