PFAS Characteristics and Treatment for Landfill Leachate

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Abstract: Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are manmade chemicals which has been extensively used, resulting in great potential for human and environmental accumulation due to high persistence. PFASs can be divided to two parts, terminal and precursor compounds, based on how many fluorine atoms link to the carbon bonds. Many studies have examined the removal ability of method including activated carbon (AC), Foam fractionation (FF), chemical oxidation and boron-doped diamond anodes (BBD). This paper focus on introducing advanced remedial technology for PFAS treatment from landfill leachate. The details about PFASs are introduced, including the chemical structure, source, and properties. Then, different types of PFASs from landfill leachate are demonstrated. Some of PFAS, especially short-chain, are prone to stay as liquid phase. Activated carbon, a great model of adsorption, shows excellent performance on removal of PFOS and PFOA. In addition, boron-doped diamond anodes technique which belongs to the electrochemical anodic oxidation, have a great potential on landfill leachate in the future. Last but not least, the conclusion makes a summary and shows possible treatment to landfill in the future.

Keywords: PFAS, Landfill Leachate, Treatment, Activated Carbon, Oxidation.

1. Introduction

PFASs are synthetic compounds that have been extensively utilized, since 1940s, in a diverse range of commercial and technical purposes [1]. It is normal to found PFASs in the fields of textile, non-stick coating, electronics, fire extinguishing foam, and others [2]. PFASs have great potential to accumulate in human and environment due to high persistence. PFASs contain one or more carbon bonds, that every hydrogen element associated with carbon are substituted by fluorine bonds [3]. In addition, C-F bonds are the strongest among all bonds nowadays, which makes it extremely hard to degrade in the environment. Therefore, overuse of PFASs contribute to children bioaccumulation and toxicant exposure, which are also relative to immune system of children. There are many concerns on PFAS, and as a result, treatment exploring plays an essential role toward the environmental friendly society. PFASs are usually regarded as a group of chemical compounds, which include precursors and products after degrading. Chemical molecules with C-H bonds that have not yet been supplanted by fluorine elements are considered precursors. However, it still has possibility to transform to terminal PFAS. Furthermore, the intermediate substances of precursors and degradation compounds seem to have higher toxicity, compared to original sources [4].

The PFAS compounds from technical applications accumulate when the waste is disposed, including but not limited to groundwater, landfill leachate and wastewater treatment plant. When the industrial and household wastes are disposed to municipal solid waste landfill, the PFASs on the surface are potentially transforming to the leachate, due to the leachable peculiarity. Landfills play a significant role on separating the accumulated waste by using geotextile liners, and equally important, preventing the landfill leachate form polluting the groundwater through the gas and liquid phase collection. Moreover, the leachates are usually transferred to wastewater treatment plant after collection. Consequently, it is very crucial to explore the PFASs removal through some advanced remedial measures. Previous reviews have demonstrated it is very common to found perfluoroalkyl acids (PFAAs) in leachate from landfill [5]. Additionally, the majority of PFAAs are short-chain from C4 to C7, which is highly associated to its better fluidity compared with long-chain. There have been reviews focusing on summarizing the remediation of PFASs on groundwater [6]. However, the removal methods are usually unsuitable for cloudy solutions like wastewater and landfill leachate.

This paper aims to introduce the latest potential technology for PFAS treatment on landfill leachate, and discuss the possible method on landfill leachate.

2. PFAS characteristics in landfill

PFASs can be divided to two parts, terminal and precursor compounds, based on how many fluorine atoms link to the carbon bonds. Terminal PFASs normally include the species that all the atoms in alkyl are substituted by fluorine, with different functional group at the end of the compounds. Different types of terminal PFASs are classified by the tails ending of the compounds. Based on that, terminal PFASs include but not limited to Perfluoroalkyl carboxylates (PFCAs), Perfluoroalkane sulfonates (PFSAs), and Perfluorooctane sulfonamide, while PFCAs and PFSAs belong to PFAAs [7]. In addition, the length of compounds is also determined by the number of carbon atoms, which are classified as long chains and short chains. Long chains for PFCA are those with more carbons than octane, whereas long chains for PFSA are those with more carbons than heptane. However, there are also some species lacking fluorine on the alkyl, which means the carbon bonds are not fully substituted by fluorine, but more than two of carbon bonds are all supplied by fluorine. These species are regarded as precursors, additionally, potentially become terminal PFASs. This is because the C-F bonds are stronger than C-H bonds, which means the fluoride ions in the liquid phase prone to replace the elements attached to the carbon atoms.

A great amount of residential and industrial waste accumulates at landfill, which result in the PFAS stay in landfill. Some of PFAS, especially short-chain, are prone to stay as liquid phase. As PFAS-containing debris builds up on landfills, landfill leachate also serves as a substantial source of PFAS. Therefore, when the landfill leachate is discharged after being purified and treated, the PFAS treatments have a significant impact on the environment around. As a result, the leachates are required to remove PFASs before discharging to the surrounding environment. PFOA and PFOS are mainly detected in landfill. Consequently, most studies examine the treatment method from contaminated water by testing the concentration of PFOA and PFOS. The type of PFASs in landfill is decided by the amount of various sources. Therefore, the concentration of PFASs from landfill leachate varies with the solid waste in different country. Wei *et al*. reported the compounds value of global landfill leachate from more than 4 countries [7]. The treatment experiment should focus on removing short-chain of PFOA and PFOS, due to the higher concentration detected. This is main because the easily leachable and mobile characteristic of short-chain.

3. Removal technology for landfill leachate

3.1. Physical ways

Adsorption is a physical removal method for PFAS. The method for removing PFASs from water relies on hydrophobic contact. This approach works well and is simple to spread throughout water. Due to PFAS' non-degradability, the main challenges of this technology are the regeneration of materials and the disposal of materials that have absorbed PFAS [7]. Activated carbon is one of the most efficient ways to removal PFAS. Previous study shows the ability of PFOA removal by activated carbon in groundwater can reach 90% to 99% [8]. Therefore, the activated carbon has been regarded as an accepted method to treat the groundwater affected by perfluorooctane sulfonate (PFOS) and PFOA. The treatment of PFAS in landfill leachate by activated carbon occur because the similarity between landfill leachate and groundwater. However, landfill leachate contains a great amount of contaminant instead of groundwater impacted by PFAS. Due to its high porosity, activated carbon is renowned for its ability to absorb PFAS. The removal capacity of activated carbon depends on its source and various surface areas. Previous study shows activated carbon with Fe can remove more than 97% of PFOA from leachate [9]. The experiments are bench-scale, which prove the possibility of using carbon for the treatment of leachate. The result shows the activated carbon have high preference to PFOA compared to dissolved organic matter. Besides, the study in full scale experiments also evaluate the adsorption capacity of activated carbon to PFAS from landfill leachate [10]. Result also proves the high efficiency to remove PFCA and PFSA using activated carbon. Nevertheless, the practical use of activated carbon requires good management, because PFAS substances from landfill adsorbed by activated carbon have the possibility to be re-landfilled. In 2017, researchers examined the factor affected the adsorption of powered activated carbon (PAC) to PFOS, including phosphate competition and temperature [11]. They found the PAC absorbed more PFOS as the temperature increase. As inputting the phosphate, PAC show a stronger hydrophilicity, which makes the removal ability to PFOS weaker. However, different type of activated carbon show different removal behavior. The main factor is the surface charge, which show the strong correlation between PFAS removal ability and electrostatic interactions. There is no doubt that the high efficiency of activated carbon absorbing PFAS substances has been accepted. However, physical treatment like adsorption could not degrade the PFAS. It is very necessary to cleanup and recycle the activated carbon. The cost of activated carbon usually is lower, compared to the highly cost of recycle the activated carbon absorbed PFAS with methanol. Therefore, it is also crucial for the disposal of activated carbon after adsorbing PFAS, whether it is extraction with methanol or high-temperature degradation in the incineration site. However, Liu *et al*. examine the PFAS concentration changing of landfill leachate from inflow to outflow, with a powder AC system installed to the landfill [12]. They observed 26 types of PFASs and found the concentration of PFASs from outflow is higher than inflow. This is mainly because some long-chain substance broken during the process. The actual implementation through AC is quiet challenging, therefore, it is prospective to explore the suitable value and right installed place on landfill.

Foam fractionation (FF) is a possible alternative physical method for PFAS removal. An aqueous solution containing amphiphilic species can be eliminated using the adsorptive bubble separation process known as foam fractionation [13]. In 2017, study evaluated the efficiency of PFAS removal with FF [14]. Lee et al. compared the removal ability of FF adding different metallic activators. They found the highest efficiency from all metallic catalysts, up to 99%, happened when 11.5 mM of ferric iron was added in specific time. Besides, the efficiency of ferric iron can still improve when the PH become lower. The result shows that it is environmental friendly and possible to implement FF with ferric iron in PFAS removal on landfill leachate. In 2021, a field trial study on groundwater proved that the FF remove the PFAS successfully, only less than 0.5% of PFAS left [13]. The air bubbles from FF shows similar function with granular activated carbon, both belong to adsorption material. Foam fractionation, as an environmentally friendly and sustainable treatment method, has proved its ability to remove PFAS in groundwater field cases. This may also apply to landfill leachate, but it has to take into account that leachate has a lot of dissolved organics that may affect the effect of FF. The study evaluated the FF ability of PFAS from landfill leachate [15]. More than 10 type of PFAS were analyzed, with high removal (at least 90%) of terminal PFAS, and low removal (less than 80%) of PFAS precursors. The removal method of FF relies on the bubbles generation. It is also important to make sure the bubble keep stable for PFAS removal.

3.2. Chemical ways

The PFAS degradation is very essential because the landfill leachate need to be processed and purified on wastewater treatment, before discharging to the surrounding environment. One of the common degradation methods is chemical oxidation. The mechanism of oxidation is mainly achieving the C-F bonds broken as well as the broken of functional group. The study tested the ability of PFOA degradation with a highly reactive oxidant species called gallium oxide [16]. The result shows perfect behaviours of PFOA removal at 100% within 1.5 hours. The comfortable environment for the gallium oxide is pH at 3, which improves the PFOA destroy ability in acidic conditions. Besides, gallium oxide also exhibits excellent performance of PFOA degradation in wastewater treatment. Another advanced oxidation method, electrochemical oxidation, also has great potentiality on PFOA in full scale. The study proved the excellent capability in PFAS removal, with removing 96% of PFOA and almost 99% of PFOS respectively [17]. Besides, they also examined the treatment in high concentration carbon solution. Almost 96% of PFOS degrades, compared with closed to 80% of POFA removal [6]. The possibility of PFAS treatment by electrochemical oxidation was explored, the further study showed the PFASs with long-chain were easier to clean-up, with the addition of zinc anode [18].

Sharma *et al*. reported that the boron-doped diamond anodes (BBD) have high possibility to achieve PFAS free due to the result showing excellent performance [19]. There is no doubt that the PFAS removal experimental results play an important role on choosing better oxidation method. Bench scale testing, however, is crucial for determining the likelihood of PFAS removal and testing the removal impact of various materials for PFAS. It is essential to explore and examine the effect on complex liquid like landfill leachate as well as wastewater. Pierpaoli *et al*. first demonstrated that BBD have a promising future on landfill leachate [20]. They found BBD almost 80% of PFOA and PFOS in leachate because of the unique feature, not easily corroded and durable. Nevertheless, the study focused on the treatment of terminal PFAS, it should be encouraged to analysis the precursor and short-chain PFASs.

4. Conclusion

Recently the issue of PFAS toxicity has been brought to public attention owing to the extensive use. There is potential risk in waste disposal like landfill. Therefore, PFAS removal methods on wastewater and landfill leachate play a vital role in decreasing the PFAS exposure when people drink water. The main compounds detected from landfill are PFOA and PFOS due to the source of residential and technical waste. The excellent adsorption property of activated carbon shows a promising performance on removing PFOA from landfill leachate. Most studies demonstrated the activated carbon from different sources on bench scale. In addition, BBD and oxidation method also has been observed successfully in leachate. The single remediation is hard to meet the requirement, so the combination of multiple treatments should be encouraged. For example, installing adsorption process in landfill leachate and degrading the PFASs after pumping the leachate through BBD would be a feasible solution. The design of treatment should be considered environmentally friendly, economically.

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