

INNOVATIVE TECHNOLOGIES OF OIL AND GAS

INNOVATIVE SOLUTIONS TOWARDS ACHIEVING COMPREHENSIVE RESTORATION OF PETROLEUM-CONTAMINATED SOILS

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Composting is an effective and cost-efficient engineering technique used to treat agricultural waste. It involves the conversion of organic materials into stable compounds and the rapid degradation of organic matter through microorganisms found in feces. The resulting high-quality fertilizer can improve soil physical, chemical, and biological properties. However, the excessive use of heavy metals in livestock breeding can restrict the use of livestock manure for composting. Long-term application of compost products containing heavy metals can cause irreversible damage to farmland soil environments. This paper summarizes several important factors that affect the detoxification of heavy metals in composting and discusses the passivation effect of typical heavy metal passivators. The detoxification mechanism of heavy metals in compost is summarized from two perspectives: the humification effect of heavy metals and the environmental interface effects of microorganisms. This paper provides a foundation for improving the agronomic use value of avian manure aerobic composting products and for studying heavy metal passivation in compost. The application of aerobic composting in the remediation of petroleum-contaminated soil exhibits a dual impact, primarily focusing on the synergistic effects on petroleum hydrocarbon degradation and soil improvement. Such research endeavors are poised to offer innovative solutions towards achieving comprehensive restoration of petroleum-contaminated soils.

Keywords: composting, heavy metal, passivation mechanism, detoxification.

1. Heavy metal pollution current status during aerobic composting

As the global population continues to grow and living conditions improve, the demand for meat and dairy products has sharply risen, leading to a near doubling of livestock farming compared to 40 years ago [1, 2]. Improper management of agricultural waste, particularly from large-scale intensive livestock farming, can exceed the environment's natural carrying capacity, resulting in environmental pollution and ecological damage [3, 4]. In 2014, China's agricultural organic waste emissions reached a critical level. Agricultural production produced 8.19 million tons of straw and 5.51 million tons of fertilizer (dry weight basis), of which 179.7 tons of organic fertilizer (32.9% of total organic fertilizer) and 212.9 tons of crop residue (26% of total residue) can be used as compost. About 38% of pig and dairy farm manure can be directly used in farmland [5]. In the process of poultry breeding,

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farmers add a large amount of heavy metals to enhance animals' bio-resistance and disease-resistance. Only a small part of these heavy metals can be absorbed and utilized, and most of the heavy metals that are not used will enter the farmland with the feces [6]. Due to the long toxic time of heavy metals and the environmental storage capacity, they can migrate along the food chain. It is difficult to detect after pollution and challenging to remove from the environment, increasing the environmental risk of heavy metal pollution [7].

Aerobic composting is a technical measure that uses poultry manure, a certain amount of straw, leaves, and other plant stems and leaves, and additives to mix and decompose [8-10]. This process can achieve organic degradation and removal of toxic and hazardous substances [11]. Composting involves complex environmental interface effects and biochemical cycle processes with hydrolysis, ammoniation, nitrification, carbonization, humification, etc. Through these complex biochemical processes, the morphology, valence, and migration of heavy metals can be changed [12]. The addition of heavy metals in poultry intensive farming increases the physiological resistance of livestock, thereby improving their growth performance. However, only a small amount of heavy metals can be absorbed and utilized by animals, and most of them are excreted with feces [13]. Heavy metals in poultry manure can be divided into exchangeable, organic combined, carbonate bound, iron-manganese oxidation, and residual states [14]. Among these, exchangeable heavy metal ions can be directly absorbed by plants, and the carbonate binding state has a potential release risk. The organic binding state and the iron-manganese oxide binding state are difficult to be absorbed by plants, and the residual state cannot be absorbed by plants [15]. There are significant differences in the total content of heavy metals in different types of livestock manure. Cu and Zn are mainly used in pig manure and cow manure, while Cr, Cu, Ni, and Zn in chicken manure and cow manure are mainly used. Zn is dominant, and the difference in the forms of different heavy metals is significant. The Zn in pig manure is mainly in the form of iron-manganese oxide-bound state, Cu is mainly in the form of organic bound state, Mn is mainly oxidized by iron and manganese substances, carbonate bound state, and residual state form [16]. Hsu et al. found that the total amount of heavy metals generally increased due to the relative concentration effect. The morphological characteristics of different heavy metals also showed different transformation trends. Copper was mainly the organic component, manganese was mainly the solid component, and zinc was enriched in the organic complex fractions in the process of manure composting [17]. Studies have shown that the form of a heavy metal is at greater risk of leaching due to its morphological toxicity than the total amount of the metal [18]. Therefore, it is of great significance to strengthen the study on the morphology of heavy metals and reduce the toxicity of heavy metals.

2. The critical factors affecting the passivation of heavy metals

Heavy metals in compost can cause harm to human and animal health, as well as plants, due to their biological fluidity and environmental closure [19]. However, the activity of heavy metals is often affected by various factors, such as redox potential, organic matter, silica content, calcium and magnesium oxidation conjugates, etc., which can affect the release of heavy metal toxicity [20, 21]. Among these factors, pH, ionic strength, initial C/N ratio, and heavy metal deactivator are crucial factors affecting the passivation of heavy metal ions.

2.1. Effect of pH on passivation of heavy metals during aerobic composting

The pH of the composting substrate changes throughout the composting process. The typical composting pH curve is mainly divided into three stages:

- 1) In the initial stage of composting, the pH briefly declines over time due to the release of humic acid and organic acid.
- 2) As humus is further degraded, the nitrification process strengthens the release of NH_3 , raising the pH.
- 3) NH_3 volatilization and nitrification release H^+ .

The pH drops again and stabilizes between 7 and 8 [22, 23]. In this process, pH value is an important factor affecting the saturated adsorption flux of heavy metals due to the existence of point adsorption competition between H^+ and adsorptive cations. Heavy metals have higher bioavailability in acidic environments [24, 25].

As the pH of the heap increases, the ability of various functional groups to dissociate H^+ ions is enhanced, thereby increasing the affinity of the cation. The functional groups on the surface of the substrate are negatively charged due to the dissociation of H, and the heavy metal cations are united with the surface functions and removed from the soil solution [26]. Acid or alkaline

modifiers can be used to adjust the pH of the heap to improve the material structure, chemical properties, and heavy metal adsorption capacity of the heap [27]. At low pH, the adsorption surface is mainly occupied by the hydrated hydrogen ions, and the metal ions are rejected by the generated electrostatic force. As the pH increases, the hydroxide neutralizes the hydronium ions, resulting in more negative charge sites to adsorb heavy metal ions [28].

The process of composting heavy metals plays an important role in the process of organic matter humification. Due to the electrostatic repulsion between the negative charges of humic acid, the humic molecule configuration changes. At high pH, the molecular configuration becomes a more open structure, allowing for faster cation diffusion, which quickly chelates to more negative binding sites. On the one hand, specific binding sites with higher pH have more deprotonated carboxylates and phenol groups, which promote the formation of strong binding complexes of metal humic acids. On the other hand, there will be more surface conjugate formation. By lowering the pH, the positive charge increases. For example, the low adsorption of copper at low pH may be due to the protonation of acidic functional groups on the surface of the HA and the competition between hydrogen ions and copper ions. As the pH increases, the increase in adsorption can be attributed to the complexation of the deprotonated functional groups of humus and the competition with hydrogen ions [29]. Due to the fluctuation of pH, the binding site of humic acid also appears as a dynamic competition between hydrogen ions and metal ions. Therefore, the passivation of heavy metals also needs to be stabilized in a relatively constant pH environment to be more effective.

2.2. Effect of ionic strength change on adsorption and resolution of heavy metals

The composting system is a complex ion dispersion system with abundant cations and anions. During the composting process, feces are generally negatively charged, and metal cations are adsorbed in the diffused ion group of the double-electrode diffusion layer on the surface of the colloid, forming the ion composition configuration in the diffusion layer. According to Coulomb's law, the more negative charges on the surface of the colloid, the more heavy metal cations are adsorbed. The higher the charge density on the surface of the stack, the stronger the cation's charge carrying capacity, and the stronger the heavy metal ions adsorbed [30]. Based on the characteristics of ions, high-valent heavy metal cations have a strong affinity with the surface of the compost colloid, while low-weight heavy metal cations have weak affinity. The heavy metal ions with the same valence state are mainly determined by the water and radius of the ions. The smaller the water and radius, the stronger the adsorption capacity of the charge. In compost, cations exchange between the cation adsorbed by the colloid and the heavy metal ion. H^+ , K^+ , Na^+ , Ca^{2+} , and plasma are adsorbed in livestock manure, while heavy metals participate in the exchange of cations under the effect of composting humification. Heavy metal ions displace the cations in the heap, so adding beneficial cations during the composting process can change the exchange direction of the metal cations, thereby improving the passivation ability [31]. The influence of other cations on the passivation is also reflected in competitive adsorption, and there is competition between coexisting heavy metal cations at the adsorption site of the same adsorbent. However, when a trivalent metal is added to the solution, it competitively replaces the divalent ions previously adsorbed on the vermiculite pure clay mineral, causing the metals to be desorbed into the solution [32]. This enhances the activity of the target heavy metal ions. Anions dispersed in the heap can be affected by negative adsorption and obligate adsorption. The main reason is that it will form a complex with metal ions and hinder the biosorption of metal ions by microbial colloids. The greater the complex equilibrium coefficient, the more obvious the adsorption effect [33, 34]. The specific adsorption of anions is achieved by entering the atomic coordination shell of the metal surface oxide, re-participating in the coordination of the hydroxyl hydration groups in the coordination shell, and directly adsorbing the double colloid in the compost colloid by covalent bonding and coordination on the inner layer of the layer of electrons.

2.3. Effects of initial C/N on heavy metals in aerobic compost

The initial C/N ratio is a key factor in determining the quality of composting products in aerobic composting of poultry manure. A suitable ratio of 20-30 is generally believed to be appropriate [35, 36]. However, some studies have found that a low C/N ratio (16-20) can increase nitrogen loss rate, increase the mineralization degree of N, shorten the thermophilic period, and lengthen the period of maturity, although it reduces the amount of C source material used [37, 38]. On the other hand, some scholars have shown that low C/N ratio treatment can also produce high-quality fertilizers. During the 12-month pig manure straw composting process, low C/N ratio treatment at the beginning of composting leads to rapid degradation of fibers (hemicellulose:

50-80, cellulose: 40-60), while a higher initial C/N ratio leads to slow degradation of hemicellulose and cellulose (10-20). Low C/N ratio microbial biomass and respiration are significantly higher than high C/N ratio, which is related to the fact that the initial warming phase of low C/N treatment is longer than high C/N ratio processing [39]. The appropriate C/N ratio plays a key role in the toxicity release of the heap [40]. The effect of C/N ratio on heavy metals in compost mainly manifests in the ability to directly affect the C and N content, which affects the ratio of functional groups such as hydroxyl groups and carboxyl groups, thereby affecting the adsorption sites of heavy metal ions. At the same time, the C/N ratio can also indirectly affect the microbial community structure, the decomposition rate of the mesophilic period and macromolecular substances (cellulose, hemicellulose, sugars, lipids), and affect the binding status of heavy metal ions through the transfer effect [41].

3. The critical detoxification process of heavy metals through aerobic composting

3.1. Mechanism of chemical passivation

In the aerobic composting process of poultry manure, organic matter rapidly humifies to form humus, which is a multi-phase macromolecular organic compound with abundant carboxyl, hydroxyl, sulfhydryl, aldehyde, methoxy, and amino groups [42]. The humic acid and fulvic acid in the humus in the compost belong to this type of surface configuration. Humus contains a large number of acidic functional groups, and the acidic functional groups of humic substances can adsorb heavy metal ions. Studies have shown that the total acidic functional group content and carboxyl group content are significantly correlated with the adsorption capacity of heavy metals, especially acid-containing carboxyl groups, which may lead to higher adsorption flux. This high adsorption flux and rapid adsorption process are mainly due to the effective surface of humus. The point content is high, and the dissociation of the humic acid functional group, especially the carboxyl group, may be the main source of effective adsorption sites. In addition, groups such as COOH, phenol, or NH₃ in humus split into COO⁻, phenol, and NH₂, which also create favorable conditions for electrostatic adsorption (Frutos et al., 2016). The complexation of heavy metals mainly forms stable complexes with humic acids through the strong humification process of composting, reducing the bioavailability of heavy metals [43]. The participation of agricultural waste in the composting process effectively increases the content of organic matter, strengthens the humification effect, increases the amount of surface charge, and contributes to the adsorption of complex substances [44]. The complexation process of metal by agricultural waste composting may be through electrostatic attraction and complexation of cellulose with Pb, complexation of carboxyl groups with Cd, and complexation of n-alkyl groups with Cu [45]. At the same time, due to the electrostatic repulsion between the negative charge of humic acid, the shape of the molecule changes, and the more open structure at high pH allows for faster cation diffusion, thereby rapidly sequestering to more binding negative sites [46]. In addition, humic or inorganic phosphate compounds react with heavy metal ions to form insoluble humus compounds or form phosphate metal precipitates. These precipitates maintain a steady-state precipitation state and are not easily hydrolyzed or chemically dissociated, thereby achieving the purpose of fixing heavy metal ions [47, 48]. The heavy metal ion combines with the electron coordination to form a coordination complex. In the complexation reaction, a complex having more than one ligand of the coordination group and the metal ion forms a metal chelate compound. Generally, the more the number of ion chelate rings in the metal in the compost, the more stable the complex, and the risk of release of heavy metals is less.

3.2. Mechanism of microbial passivation

The biochemical behavior of microbial passivation of heavy metals in aerobic composting mainly includes biosorption (metal ions are adsorbed on the cell surface), methylation of metal ions, biomineralization (by forming insoluble sulfides and macromolecules), intracellular accumulation and redox catalytic processes [49]. The tolerance of microorganisms to heavy metals depends on the ability of cell surface functional groups to bind metal ions. Bacteria and fungi have different levels of adsorption on heavy metals, with fungi being more tolerant than bacteria due to their larger specific surface area. According to Fomina et al. 2018 [45], fungi are more tolerant than bacteria and have a larger specific surface area. During composting, the adsorption of metal by fungi is mainly manifested by the complexation of cell wall chitin with other polysaccharides and glycoproteins, as well as the synthesis of metallic thiopurines and other functional groups containing metallic thiols.

Cell surface adsorption mainly involves extracellular polymers produced by extracellular walls with chelated or complexed heavy metals and chemical groups with cell walls capable of adsorbing heavy metal ions. When studying the effect of extracellular polymer on the biosorption of Pb^{2+} by the metal ion-resistant fungus *Aspergillus*, it was found that the minimum inhibitory concentration of fungal extracellular polymeric substances (EPS) to Pb can reach 72 mm, which is due to the deprotonation of functional groups in EPS. Mohapatra et al. also studied the interaction mechanism between mercury (Hg^{2+}) and *Bacillus thuringiensis* PW-05 extracellular polymer (EPS), and found that the heavy metal ions bind to the polymer actively. In general, the surface cells of microorganisms are mainly composed of polysaccharides, proteins, and lipids, which contain functional groups that can combine with metal ions such as hydroxyl group, carboxyl group, phosphoryl group, amino group, sulfate group, and amide. The complexation reaction mainly increases the metal by surface adsorption or formation of a stable complex with humic acid by adding a heavy metal immobilizing conditioning agent to form a more stable complex or as an adsorbent for adsorbing metal ions [50]. However, the exact mechanisms involved in ion exchange, coordination complexation, physical adsorption, and inorganic microprecipitation are still unclear. In general, we can say that it all comes down to the adsorption of microbial cell surface.

Microbial effective Cells Transfer ionic heavy metals to the interior of the cell for adsorption, precipitation, and transformation through metabolism [51]. This detoxification effect involves the transport pathway of heavy metal ions inside the cell and the functional regionalized detoxification effect within the cell [52-53]. When the heavy metal ions in the heap system exist in an appropriate amount, the speed of transmission can be accelerated, but in excess, the effective cells can reduce the accumulation in the cells by reducing transport, impeding, or even shutting down the transport transfer pathway to reduce heavy metal ions to reduce the damage of heavy metal ions to their intracellular system. For example, copper can act as an enzymatic factor to increase the activity of cellular enzymes, but excess copper reduces the activity of enzymes [54]. High concentrations of uranium do not affect biosorption capacity, but inhibit biomineralization, indicating high concentrations of heavy metals may inhibit the enzymatic activity involved. The distribution of heavy metal ions in different regions of the cell is also different, and the passivation conditions are also very different. After entering the cell, part of the heavy metal ions can be enclosed in the organelle, such as Cd, which is easily enriched on the chloroplast [55]. Heavy metal ions can also interact with intracellular protein species to form more stable and low-toxic compounds. Many studies have shown that metal ions can be combined with proteinaceous materials to transform into low-toxic forms. Metallothionein contains a large amount of sulfhydryl groups, and this special group structure gives it a chelated heavy metal ion and detoxification function [56]. In addition, intracellular thiol can also store, regulate, and detoxify heavy metal ions in cells, and its ability has been proved to varying degrees. Metal ions can be transported through the membrane and enter the cell body, but metal ions cannot be immobilized in the vacuoles of the cells or on the keratinocytes by the cells participating as nutrients in the intracellular material circulation [57]. Microbial immobilization of heavy metal ions is mainly achieved by epidermal adsorption, intracellular regionalization, and chemical transformation [58].

4. Research progress on the application of aerobic composting products in petroleum contaminated soil

The application of aerobic composting in the remediation of petroleum-contaminated soil exhibits a dual impact, primarily focusing on the synergistic effects on petroleum hydrocarbon degradation and soil improvement. In terms of petroleum hydrocarbon degradation, studies have unveiled mechanisms by which aerobic composting promotes soil microbial activity, leading to more efficient petroleum degradation. By introducing oxygen and organic matter, aerobic composting creates a conducive environment for soil microbes, accelerating the degradation of various petroleum hydrocarbons, including complex structures like polycyclic aromatic hydrocarbons and polyalkanes [59]. Concerning soil improvement, the application of aerobic composting results in alterations in soil microbial community structure and physicochemical properties. This encompasses improvements in soil structure and increased porosity, as well as an enhancement of microbial diversity and functional potential in the soil [59, 60]. Such micro-level regulation positively influences soil ecosystem functions, including the inhibition of plant pathogens and organic matter decomposition [61]. However, challenges and unresolved issues persist in this field. Firstly, a deeper understanding of the dynamic changes in microbial community structure and function following the introduction of aerobic composting is required to more accurately predict its long-term impacts on petroleum hydrocarbon degradation and soil improvement [62, 63]. Secondly, researchers need to conduct detailed studies on the applicability of aerobic composting in different soil types and environmental

conditions to formulate more targeted remediation strategies. Additionally, potential negative effects of aerobic composting, such as soil acidification and organic matter overload, need comprehensive assessment and management [64, 65].

In future research, in-depth exploration of the interactions of aerobic composting in different soil environments, fine-tuning of microbial metabolic pathways, and its long-term impacts on soil ecosystem functions and stability will provide crucial insights and guidance for more effective and sustainable petroleum-contaminated soil remediation. Such research endeavors are poised to offer innovative solutions towards achieving comprehensive restoration of petroleum-contaminated soils.

5. Conclusions

The passivation mechanism for reducing heavy metals during aerobic composting is complex and involves changes in the micro-ecological community of microorganisms. Although omics technology has advanced our ability to identify key microbial species, quantifying complex microbial chemical processes remains challenging. For instance, the extent of heavy metal degradation by microbial groups at different temperatures and the rate of influence on different heavy metal forms are not well understood. Therefore, further research is needed to strengthen our understanding of these relevant aspects. By fostering a holistic understanding of the intricate interplay between aerobic composting and soil ecosystems, these research efforts aim to contribute to the broader field of environmental science and remediation practices. The outcomes of such endeavors have the potential to revolutionize our approach to petroleum-contaminated soil management and significantly advance the prospects for achieving holistic and enduring soil restoration.

Acknowledgements

This research was supported by The Humanities and Social Science Research Project of Hebei Education Department, grant number SD192007; Hebei Province Natural Science Foundation for Youths, grant number D2022105002.

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