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Study on the adsorption properties of humic acid biochar on naphthalene in soil

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Abstract

A new environmentally friendly soil remediation material based on wood vinegar modified sodium humate loaded with biochar powder (BC-NaHA/WV) was prepared, and the effects of the composition, addition amount and aging time of this material on the remediation effect of naphthalene-contaminated soil were studied. The results showed that the solidification rate of naphthalene by wood vinegar modified sodium humate-loaded biochar powder reached 60.20%, which was 10.08% higher than that of biochar powder loaded with sodium humate; the optimum application rate of 1kg naphthalene-contaminated soil was 30g; The rate is proportional to the aging time. The longer the contact time of naphthalene and the remediation material in the soil, the lower the mobility of naphthalene in the soil.

Keywords

Naphthalene-contaminated soil; humic acid; biochar; adsorption performance.

1. Introduction

With the rapid development of industry, the burning of a large number of fossil fuels, the random discharge of industrial sewage, the leakage of oil and other factors have led to the continuous increase of organic pollutants in the soil (Li et al, 2018). Organic pollutants will change the physical and chemical properties and biological properties of soil, affect the quality and quality of crops, and indirectly cause fatal harm to human health (Zhang et al, 2017). Soil polycyclic aromatic hydrocarbons (PAHs) pollutants, usually represented by naphthalene, have been remediated.

Humic acid is a natural organic matter that exists widely in soil. It is often used in the remediation and treatment of organically polluted soil due to its advantages such as low price, wide source, and environmental protection (Aeschbache et al, 2012). The oxygen-containing functional groups in humic acid interact with organic matter to remediate soil organic pollution (Li, 2020; Gunasekara et al, 2003) and oil pollution (Pellegrino et al, 2008) through adsorption and solidification. Humic acid has good adsorption and degradation ability to medium and low ring aromatic hydrocarbon PAHs such as naphthalene (Gu et al, 2011).

The composite remediation material based on humic acid is the main trend of soil remediation and remediation in the future (Yu et al, 2012). Humic acid (HA) exhibits a strong ability to adsorb cationic and hydrophobic organic pollutants, and the mechanism of action of humic acid with hydrophobic organics is hydrophobic effect, π-π interaction and hydrogen bonding. Activated humic acid can significantly improve the adsorption and complexation capacity, and acidification is an effective method for activating humic acid (De et al, 2012). Wood vinegar is a waste liquid derived from charcoal production. It is rich in organic acids, easily degraded, easily accepted by the soil, and has a particularly low price. It has great application prospects in soil improvement (Lu et al, 2017). Humic acid to enhance repair performance.

Biochar is a solid biomass waste such as manure, sawdust, straw, etc., which is prepared by high temperature pyrolysis under anaerobic or oxygen-limited conditions. Biochar has high internal

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porosity, large specific surface area and strong adsorption capacity, and has a certain removal effect on various pollutants in water and soil (Sun et al, 2021). In order to further improve the ability of biochar in soil pollution control, it is usually modified to prepare biochar-based soil remediation materials with high adsorption activity, which are widely used in soil pollution remediation (Hu et al, 2020).

In this paper, biochar powder loaded with humic acid and wood vinegar were used to synergistically remediate naphthalene-contaminated soil. Biochar powder provides a carrier for humic acid, and wood vinegar liquid activates humic acid to effectively reduce the toxicity of excess humic acid in the soil, improve its adsorption performance, and have a better curing effect on organic pollutants in the soil.

2. Materials and Methods

2.1. Experimental reagents

Sodium humate: industrial product, weathered coal humic acid, black-brown solid, free value is 55%, solubility >82 g, aqueous solution is brown, pH value is 11.10, density is 0.736 kg/L; wood vinegar: industrial product; naphthalene , n-hexane is of analytical grade; biochar powder: industrial product.

2.2. Experimental Materials

The tested biochar powder (BC) is an industrial biochar powder that has been pulverized, ground, and passed through a 100-mesh sieve. The tested wood vinegar (WV) is the waste liquid produced by the charcoal manufacturing process and left standing for 7 days, filtered, distilled and collected at 120~150℃. This fraction is light yellow clear liquid, acetic acid accounts for 80.40% (mass fraction, the same below), phenol 8.03%, ketones and alcohols 10.58%, other 0.99%, pH value is 2.45, specific gravity is 1.0181, density is 0.998 kg/L .

Preparation of wood vinegar liquid modified biochar powder loaded humic acid remediation materials: various remediation materials are fully mixed and evenly passed through a 100-mesh sieve after natural air-drying, and the addition amount of naphthalene-contaminated soil is 30g/kg. Restoration material with a mass ratio of biochar powder, sodium humate and wood vinegar of 1:1:1 (BC-NaHA/WV); a restoration material with a mass ratio of sodium humate and wood vinegar of 1:1 (NaHA/WV); restoration material with a mass ratio of biochar powder and wood vinegar liquid of 1:1 (BC-WV); restoration material with a mass ratio of biochar powder and sodium humate of 1:1 (BC-WV) -NaHA); biochar remediation material (BC), in which biochar is 100%; blank group (CK) is without any remediation material, and the rest of the condition steps are the same as the experimental group (adding remediation material).

Preparation of naphthalene-contaminated simulated soil: Take the soil from the surface layer of about 0 to 20 cm in the residential area, pass it through a 100-mesh sieve, remove grass roots, stones and other debris, and place it to air dry naturally. Referring to "Soil Environmental Quality Construction Land Soil Pollution Risk Control Standards (Trial)" (GB36600-2018), weigh 1.00 g of naphthalene and dissolve it in 500mL of n-hexane solution, and then fully stir and mix with 1.00 Kg of soil sample, and put it into a fume hood After stirring with a glass rod for 1 h, place it in an open place in the dark. After the n-hexane solvent is evaporated to dryness, wrap the container with aluminum foil and age at 4°C for one month. Finally, the soil particles are ground with a mortar and freeze-dried for storage.

2.3. Determination of Naphthalene Contaminants in Soil

Measurement of naphthalene content in contaminated soil: Weigh 10 g of contaminated soil into a 100 mL conical flask, add a certain amount of repair material and 20 mL of n-hexane solvent, put the mixture in a constant temperature oscillator at room temperature (25℃) and shake for 2h, and after ultrasonic extraction for 1h , centrifuge at 5000r/min for 20min, separate the supernatant after purification by column chromatography, transfer it to a dry round-bottom flask, use a rotary evaporator to concentrate the supernatant to 1~2 mL, and then use n-hexane Dilute to 20 mL, pass through a 0.22 μm filter membrane, measure the naphthalene content in the supernatant with gas-mass spectrometry GC-MS, and calculate the solidification rate. Set the blank group as no repair material, and the rest of the operation steps are the same as above. The adsorption effect is measured by the solidification rate, and its value is expressed by the ratio of the solidified amount (the content of naphthalene that is solidified in the soil and cannot migrate with water or can be absorbed by plants) to the initial pollution amount.

Solidification rate

$$(\text{wt\%})=(m_1-m_2)/m_1\times 100\text{wt\%} \quad (1)$$

In formula (1), m_1 is the initial contamination amount (mg), and m_2 is the dissolution amount (mg).

Determination of the standard curve of naphthalene: use GC-MS to measure the naphthalene solution with gradients of 0, 5, 10, 20, 40, 60 μg/ml, and obtain the regression curve equation y=43350x (y-peak area, x-concentration) . Measure the supernatant of the blank group (contaminated soil without remediation material) and the experimental group (contaminated soil with remediation material) by GC-MS, get the peak area and bring it into the regression equation to get the concentration of naphthalene in the solution, The initial contamination and dissolution of naphthalene can be obtained by multiplying the volume.

3. Results and analysis

3.1. The effect of different remediation materials on the solidification rate of naphthalene in soil

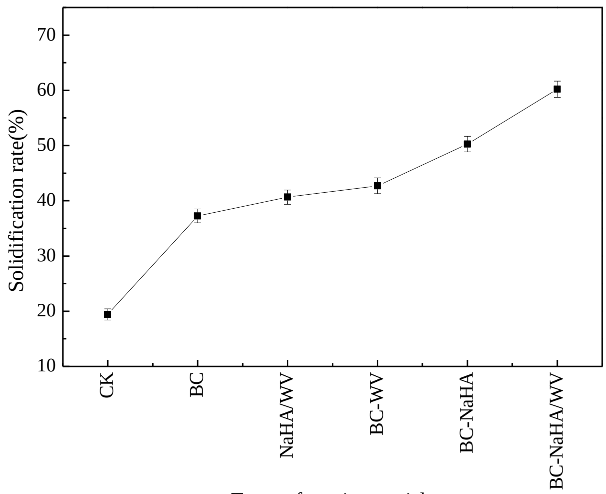

Fig.1 Effects of different remediation materials on the curing rate of naphthalene

It can be seen from Figure 1 that, compared with CK, BC, NaHA, BC-WV, BC-NaHA, and BC-NaHA/WV all have a curing effect on naphthalene in polluted soil. Among them, the solidification efficiency of BC to naphthalene in soil is the lowest, the solidification rate is 37.26%; the solidification rate of NaHA/WV to naphthalene in soil is 40.66%; the solidification rate of BC-WV to naphthalene in soil is 42.71%; BC The solidification rate of -NaHA to naphthalene in soil is 50.28%; BC-NaHA/WV has the highest solidification efficiency of naphthalene in soil, with a solidification rate of 60.2%, which is 10.08% higher than that of BC-

NaHA to naphthalene. The results showed that the addition of wood vinegar promoted the activation of sodium humate into humic acid, and the number of oxygen-containing functional groups increased. After loading with humic acid and biochar, the adsorption and solidification of naphthalene was enhanced (Lu et al, 2017).

3.2. Influence of the addition amount of BC-NaHA/WV on the curing rate of naphthalene

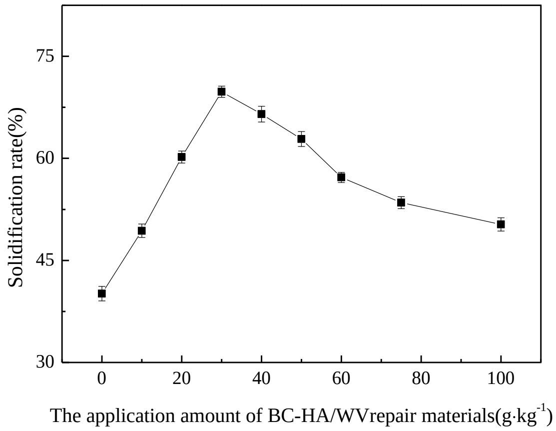

Fig.2 Effect of BC-HA/WV repairing material application rate on the solidification rate of naphthalene

It can be seen from Figure 2 that the low concentration of biochar powder-humic acid/wood vinegar remediation material (BC-NaHA/WV) promoted the adsorption of naphthalene in the soil, while the high concentration of the remediation material reduced the adsorption capacity of naphthalene in the soil. In 1 kg of naphthalene-contaminated soil, the application rate of remediation materials was in the range of 0-30 g, and the solidification rate of naphthalene increased gradually. When the application amount of BC-NaHA/WV is 10 g, the minimum curing rate is 49.36%. At 30 g, the adsorption reached the highest value, and the curing rate reached 69.8%. Secondly, when the application amount is 20 g, the curing rate is 60.20%. The application amount was in the range of 30-60 g, and the curing rate decreased with the continuous increase of the amount of repair material. When the application amount is 40 g, the curing rate is 66.50%. In the range of 60~100 g, the curing speed gradually stabilized. The results showed that the optimal application rate of BC-NaHA/WV material to naphthalene-contaminated soil was 30 g.

3.3. Effect of aging time on curing rate of naphthalene

When the temperature is 25℃, the same batch of naphthalene simulated contaminated soil and placed for different times (7d, 14d, 28d, 42d, 56d) were taken, and 30 g/kg of biochar powder-humic acid/wood vinegar remediation materials were added respectively. (BC-NaHA/WV), take the quantitative test respectively, and obtain the solidification rate of naphthalene in the simulated soil with different aging times. It can be seen from Figure 3 that the aging time is proportional to the solidification rate. The longer the naphthalene is in contact with the soil, the lower the efficiency of its migration in the soil. When the aging time is 28d, the adsorption rate is the fastest, and in the process of 28d~42d, the adsorption rate gradually slows down, and the adsorption gradually reaches equilibrium at 56d.

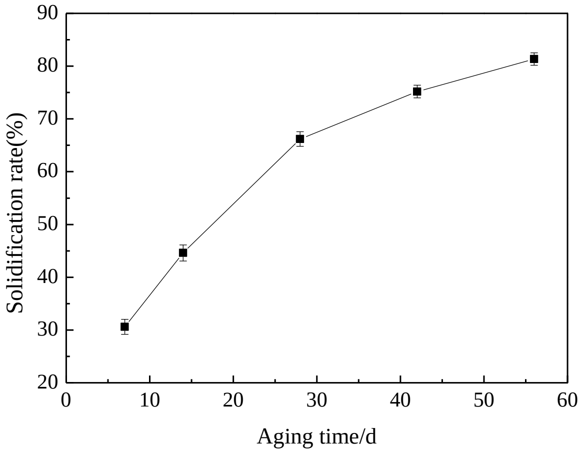

Fig.3 Effect of aging time on the solidification rate of naphthalene

4. Conclusion

In this paper, a novel soil remediation material based on wood vinegar modified sodium humate loaded with biochar powder (BC-NaHA/WV) was prepared. By studying the effect of different remediation materials on the solidification effect of naphthalene-contaminated soil, the optimal remediation material was determined to be BC-NaHA/WV. By exploring the influence of factors such as optimal material addition amount and aging time, the results show that the solidification rate of naphthalene by wood vinegar modified sodium humate loaded biochar reaches 69.20%, which is 18.92% higher than that of biochar loaded with sodium humate. ; The optimal dosage of 1kg naphthalene-contaminated soil is 30 g; the curing rate is proportional to the aging time, the longer the contact time between naphthalene and the remediation material in the soil, the lower the mobility of naphthalene in the soil.

Acknowledgements

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