A review about microwave regeneration technology of waste activated carbon

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Abstract. Activated carbon (AC) can concentrate and transfer pollutants using its own adsorption capacity. Waste AC is a kind of solid waste, its adsorption capacity of pollutants is greatly reduced after adsorption saturation. If handled improperly, it will cause secondary pollution to the environment. Therefore, it is necessary to regenerate the waste AC to restore its adsorption performance and reduce environmental pollution and resource waste. Microwave regeneration technology is of great research value because of its low energy consumption, time saving and high efficiency. Based on the characteristics of microwave heating and regeneration mechanism, this paper summarizes the advantages of microwave regeneration compared with conventional heating regeneration, and the application status of microwave technology in the regeneration of waste AC was reviewed. The influence of different types of pollutants adsorbed by AC, the change of control conditions for microwave regeneration of AC and the characteristics of AC itself on microwave regeneration were briefly described. And the development direction of microwave regeneration of AC in the future is also prospected.

1. Introduction
Activated carbon (AC) is a kind of recycling and economic carbon adsorption material because of its developed pore structure, large specific surface area and rich surface functional groups, and has a strong adsorption performance. AC is widely used in liquid and gas phase environment for the treatment of various organic and inorganic pollutants [1]. In the adsorption process, pollutants continue to accumulate on the surface of AC, the surface functional groups and internal pore structure of AC have changed, and its specific surface area has decreased. As a result, the activity of AC has reduced, AC no longer has adsorption capacity, and finally it loses its adsorption and becomes waste AC. If the waste AC is not properly treated, it will not only bring additional environmental pollution, but also waste a lot of reusable resources [2]. Therefore, in order to alleviate the resource waste and environmental pollution caused by the waste AC, it is necessary to regenerate the saturated AC after adsorption, so as to restore its adsorption performance and reuse. At present, the most widely used regeneration method is the thermal regeneration method. However, after the conventional heating and regeneration treatment of AC, the loss rate of AC is high, and the intensity of AC structure is reduced. At the same time, in the process of regeneration, and the energy consumption of regeneration and the operating costs are high. Microwave regeneration technology, can make up for the shortage of conventional heating regeneration, achieve a short time rapid regeneration, and relatively low energy consumption, the properties of AC regeneration recovered well after regeneration, so in recent years has been widely concerned.
2. Mechanism and characteristics of microwave heating regeneration

2.1. Mechanism of microwave heating
Microwave regeneration technology is to make the waste AC rapidly heating under microwave radiation, in order to break its adsorption balance, the adsorption mass desorption or decomposition, so as to achieve the regeneration of waste AC. Microwave is a kind of electromagnetic wave with a frequency between 300MHz and 300GHz. When it acts on different materials, it will show different characteristics [3]. In simple terms, a conductor (e.g. metal) can reflect microwaves; for insulators (e.g. glass & ceramics), microwave penetration is almost possible; the dielectric material (such as water & food) can absorbs the microwave and achieves the effect of heating. Substances in nature are made up of molecules (or dipoles) with different electrical charges at both ends, called The Dielectric Material. In the natural state, the dipoles in the medium show random location distribution and chaotic motion. The dielectric material is composed of polar and non-polar molecules. When the dielectric material is under the action of electromagnetic field $E$, such as microwave radiation, the ion shifts rapidly in a manner similar to rotation, and the dipoles in the dielectric material will rearrange themselves in the electromagnetic field $E$ in accordance with the principle of opposite-pole attraction [4]. This will turn the jumbled dipole into a regular array of polarized molecules. Figure 1 shows the effect of an electromagnetic field $E$ on the dipoles of a polar molecule [5]. As a result of these rotational or translational movements within the polar medium, internal friction occurs, which causes heating of the medium [6]. The more polarised the dielectric, the more energy is stored in the material. This is because the energy conversion of dielectric materials in microwave field mainly depends on ionic conduction and dipole rotation [7]. The former is the current formed by the directional movement of dissociated ions under the action of microwave field, and the dielectric material hinders the movement of ions and generates heat energy. The latter is the positive and negative dipoles that constitute the dielectric material in the magnetic field formed by the AC voltage, will rotate with the conversion of the voltage, generate friction between each other, and generate heat energy [8].

![Figure 1. The effect of an electromagnetic field $E$ on the dipoles of a polar molecule.](image)

2.2. Characteristics of microwave heating

2.2.1. High frequency and penetration. Different from conventional heating method, in which heat radiates from the outside to the inside of the material, forming a temperature gradient of high outside and low inside. When microwave acts on the dielectric material, it directly acts on the molecules of the dielectric material under the high-frequency vibration of microwave, making the molecules move violently, leading to the overall heating of the dielectric material and greatly shortening the heat conduction time. In addition, due to the heat dissipation of the material surface, the outer temperature of the material is often lower than the internal temperature, which is conducive to the outer diffusion of the material, that is, the outward diffusion of the material has a beneficial effect [9].

2.2.2. Selective heating. Whether a material absorbs microwaves is mainly determined by the "tangent of loss" of the dielectric material. The dielectric loss tan$\delta$ is a parameter that characterizes the dielectric loss. The tangent of dielectric loss tan$\delta$ is an indicator parameter to characterize dielectric loss. The larger the value of tan$\delta$ is, the stronger the microwave absorption capacity of the material is [10]. Microwave heating is selective and relatively more energy efficient because only microwave absorbing materials can be heated.
2.2.3. Small heat inertia. Under the action of microwave, the dielectric molecules move violently in a short time, making the dielectric material heat up rapidly and be heated instantly. When the microwave radiation is stopped, the dielectric molecules do not move, the heating stops, and no heating is carried out. At the same time, there will be no slow heating reaction and waste heat phenomenon, which makes the process easier to control [11].

2.2.4. Nonionizing solvent. Microwave quantum energy is not large enough, nor is it enough to change the internal molecular structure of matter, can only partially weaken or destroy the bond energy between different substances [12].

2.3. Effect of microwave on regeneration of AC
The regeneration of AC in microwave field can roughly include the following three aspects: When the polar substance is adsorbed by AC, the rapid rotation of the dipole will be produced under the action of microwave, resulting in heat energy. When the heat energy is accumulated to a certain extent, the phenomenon of desorption of the substances adsorbed by AC occurs. Further heating will cause the adsorbent which is not easily desorbed to decompose or carbonize. At the same time, the space charge polarization of AC will occur under the action of microwave field, and will make the AC have polarity, so the AC can absorb microwave energy. The active center is formed at the adsorption position to promote the degradation of organic matter [13]. In addition, in the process of regeneration, some activators, such as CO$_2$ and water vapor, are usually added. Due to the effect of microwave, both carbon and activator are at a higher temperature. At this time, the activator will play an etching role on the carbon wall surface and restore part of the pore structure reduced by the collapse and ablation of the carbon wall surface at high temperature [14].

**Table 1.** Table of comparison between microwave and conventional heating regeneration.

| Items                        | Traditional heating regeneration                                                                 | Microwave regeneration                                                                 |
|------------------------------|--------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------|
| Regeneration efficiency      | The residue after pyrolysis of organic pollutants blocked the pores of AC, and the regeneration efficiency of AC decreased. | The micropores of AC regenerated by microwave are more developed, and the adsorption performance of AC is even better than that of the original carbon. |
| Heating rate and energy consumption | The heating process of AC needs a long reaction time; High energy consumption.                      | High speed, only 1/100~1/10 of the time of traditional heating regeneration; Energy conservation and efficient. |
| Strength of AC after regeneration | AC loss rate is larger, generally 5%~10%; The strength of AC machinery decreases after regeneration. | By controlling microwave irradiation time and heating rate, the carbon loss rate can be greatly reduced on the premise of ensuring the strength of regenerated carbon. |
| Environmental protection    | The desorbed pollutants will cause secondary pollution to the environment.                          | The adsorbed pollutants are decomposed into small molecules and discharged without polluting the environment. |
| Cost                         | Complex equipment; High operating cost; High energy consumption cost.                              | Save energy consumption; Equipment miniaturization Investment costs are relatively low. |

2.4. Advantages of microwave regeneration of waste AC
Traditional heating regeneration of waste AC by providing the necessary heat to decompose the pollutants adsorbed by AC. However, the traditional heating regeneration takes a relatively long time and consumes relatively high energy. In addition, it is worth noting that the pore structure of AC deteriorates more and more obviously with higher regeneration time and temperature, which reduces the adsorption capacity and regeneration efficiency of AC after regeneration. While the waste AC in
low temperature desorption stage, the discharge of organic pollutants, will cause pollution to the atmosphere [15].

As an efficient and clean regeneration method, microwave regeneration has advantages over conventional heating regeneration. Microwave regeneration has the advantages of higher heating rate, which means that microwave heating technology can achieve AC regeneration in a relatively short time and save energy. Microwave makes organic pollutants overcome Van der Waals force attraction and achieve desorption. With the accumulation of microwave energy, parts of organic pollutants are pyrolyzed into small molecules under the combined action of thermal and non-thermal effects, which are removed in the form of CO₂, CO and H₂O. Charred residue and refractory organic pollutants are carbonized at high temperatures. This reduces environmental contamination by desorbed contaminants [16]. The application of waste AC to microwave regeneration has shown good results (Table 1).

3. Application of microwave regeneration of AC

For the regeneration of AC by microwave heating, the main influencing factors are the type of AC adsorption substances and the change of control conditions and the characteristics of AC itself. In view of these characteristics, domestic and foreign scholars have done a lot of research, their research results show that the microwave regeneration of waste AC is feasible.

3.1. Effect of different adsorbent types on regeneration

AC is widely used in water treatment and waste gas treatment, so adsorbent types are mainly divided into gas phase and liquid phase. Scholars have conducted a lot of research on the regeneration of waste AC loaded with different adsorbents under the action of microwave:

Through the experiment of microwave desorption of loaded toluene AC, Gu et al. [17] found that in the process of microwave heating, microwave energy is absorbed by AC, generating a large amount of heat, and toluene is driven out of the pore channels of AC. Under different microwave power levels, a peak value of toluene desorption rate of AC appears. With the increase of microwave power, the peak time moves forward slightly and the peak time increases. During the desorption process under different microwave radiation power, almost all the toluene was desorbed from the AC in only 20 minutes, which is 1.5 to 3.5 times more efficient than the traditional heating regeneration process. This result indicates that the microwave desorption is far more effective than the traditional heating desorption process. After three cycles of regeneration, the adsorption penetration time of AC was reduced by about 22%

Erica et al. [18] through the microwave radiation regeneration experimental study of granular ACs (GACs) loaded with Poly- and perfluoroalkyl substances (PFAS) found that GACs had a strong ability to transform the adsorption microwave power into a rapid temperature rise, which is the key to the use of microwave regeneration. The enhancement in the Regeneration Efficiency (RE) may not be exclusively attributed to the complete desorption of PFAS, but an increase in the overall uptake was attributed to the changes in GACs porous structure (e.g. the development of mesopores), as demonstrated by the variation in pore volume. After 5 consecutive adsorption/regeneration cycles, the weight loss of AC <7%, indicating that microwave irradiation regeneration of waste AC has a certain potential applicability.

Zhang et al. [19] conducted regeneration experiments on AC after treating ester containing wastewater, and found that the regeneration efficiency began to decrease gradually after the microwave power was 420W. This is because the greater the power, the more energy absorbed inside the AC, excessive power will make the AC inside at high temperature burning oxidation and other phenomena, resulting in more serious loss of carbon. And too much power can destroy the pore structure of AC, leading to the adsorption capacity of AC decreased, the reduction of regeneration efficiency. In addition, through the observation of the absorption peak of the infrared spectrum of AC before and after regeneration, it is found that, compared with the AC before regeneration, the position of C=C double bond in the regenerated AC has a large shift to the lower wave-number, and the anti-symmetric stretching vibration peak of C≡C is enhanced, indicating that the surface unsaturation of
AC increases. In addition, the C–OH absorption peak becomes wider, indicating that the surface functional groups of AC have changed after microwave regeneration, which promotes the adsorption of pollutants on AC.

Kong et al. [20] used microwave to regenerate the AC adsorbed with NOx. The results showed that the points occupied by NOx on the AC were more likely to absorb microwave energy and could become hot spots in a short time. Degradation of NOx occurred in these hot spots. Under the action of microwave, more than 90% of NOx was degraded, and the final products were CO and CO2.

### 3.2. Effect of microwave control conditions on regeneration

The microwave regeneration control conditions affecting the regeneration effect of AC mainly include microwave power irradiation time and the presence or absence of carrier gas. Therefore, it is important to study the control conditions of microwave regeneration for AC regeneration.

Yang [21] conducted microwave regeneration experiments on AC adsorbed with radioactive gas 222Rn. The effects of microwave power, carrier gas flow rate, water content of AC and different particle size on the desorption rate and carbon loss rate of AC after microwave regeneration were studied. The results show that the higher the microwave power, the smaller the carrier gas flow, the smaller the water content, and the use of small particle size of wood or coconut shell AC, the faster the heating rate of AC, the higher the maximum temperature, the maximum desorption rate can reach 97.6%, but the corresponding loss rate of carbon is also greater.

Chen [22] studied the regeneration of saturated AC adsorbed phenol wastewater by microwave method, and compared and analyzed the influence of different regeneration conditions on the recovery of AC properties. The effects of microwave power irradiation time, energy density, AC amount and reactor diameter on regeneration were investigated. The results show that the regeneration of AC by microwave is feasible. The higher the microwave power, the longer the irradiation time, the higher the energy density, the less the amount of AC, the smaller the diameter of the regeneration reactor, the more beneficial to the improvement of AC regeneration efficiency. The regeneration effect of microwave method can reach 100% or even higher. The study on the regeneration mechanism of AC by microwave regeneration showed that most of the phenol adsorbed on AC was decomposed into other organic compounds during the regeneration process. The alkaline groups on the surface of AC increased while the acidic groups decreased, which were beneficial to the adsorption of phenol on the AC.

Shi et al. [23] used microwave heat treatment method to regenerate the granular AC fully adsorbed SO2 in air & N2 atmosphere respectively. The experimental results show that the desulfurization performance of AC after microwave regeneration in air atmosphere is less than half that of the original carbon. In N2 atmosphere, the desulfurization performance of the regenerated AC increased to 97% of that of the original carbon. Compared with the traditional heating regeneration method, the microwave heating treatment regeneration method in N2 atmosphere reduces the regeneration temperature and greatly shortens the regeneration time, and the remote energy efficiency index of the former is higher than that of the latter, reflecting the advantages of high efficiency and energy saving. Under the condition of isolation of air, it can prevent the excessive oxidation of AC, and it is conducive to carrying out the substance desorbed at high temperature. However, adsorbents cannot be completely decomposed into CO2 and H2O, and may even lead to the production of intermediate products. While in the atmosphere of air, the adsorption is decomposed more thoroughly.

### 3.3. Effect of characteristics of AC on regeneration

The surface structure and surface chemistry of AC materials have different effects on the regeneration effect after microwave treatment, so a lot of research has been carried out in this field.

Zou [24] used microwave technology to physically modify the AC to improve the adsorption capacity of 2,4-DCP by changing the pore structure of the AC, and then microwave regeneration experiments were carried out on the modified saturated AC. The experimental results showed that the maximum adsorption capacity of modified AC increased by 12.5% compared with that of unmodified
AC after microwave regeneration. This is because the modification by microwave makes the pore structure of AC enlarged, the graphitization degree of carbon structure increased, the content of C=O bond increased, and the alkalinity enhanced, which are conducive to the adsorption of 2,4-DCP.

Stevy [25] separately investigated the microwave regeneration of AC loaded with nickel & copper after saturated adsorption of phenol. The AC modified by nickel can absorb more microwave energy than that modified by copper, so the regeneration ability of AC modified by nickel is higher than that of AC loaded with copper. Because when AC loaded Ni\textsuperscript{2+} modified, the surface of Ni\textsuperscript{2+} is a strong absorbing substance, which can form local hot spots under the microwave field, so that the microwave decomposition of phenol more thoroughly. On the contrary, the surface of copper-loaded AC will reflect the microwave and affect the absorption of microwave, resulting in slow temperature rise of the bed and incomplete decomposition of phenol.

4. Conclusions and future perspectives

Through studying the practical application and scientific research results of microwave regeneration of waste AC, it is found that: In the process of microwave regeneration of waste AC loaded with different adsorbates, the mechanism of action on the pore structure and surface chemical properties of AC is different. In both gas and liquid phase adsorbents, although there is a phenomenon of carbon structure ablation at high temperature, the overall effect of microwave regeneration of waste AC is significantly better than traditional thermal desorption. The control conditions of microwave regeneration technology have different effects on the recovery of AC adsorption performance, so the regeneration effect and economy should be weighed to find the optimal process scheme. By modifying the surface structure and surface chemical properties of AC, the microwave regeneration effect of AC can be improved. Compared with direct microwave regeneration, the adsorption performance of AC can be restored to a greater extent.

Compared with traditional thermal regeneration methods, microwave regeneration technology of waste AC has the advantages of uniform heating, high efficiency and energy saving as well as reducing pollutant emissions and the operating investment cost is low, which is a very promising and advantageous AC regeneration technology. However, at present, the microwave regeneration technology still stays in the laboratory stage, and most of the microwave reactors used are self-modified. Both the equipment parameters and the scale are not enough to be applied to the practical engineering. In the future, in order to achieve successful commercial application of microwave regeneration technology, research and development should include the exploration of suitable microwave heating rate and reaction temperature, in addition, the amplification of microwave reactor must be done. Secondly, the mechanism of microwave regeneration needs to be further studied, to master the essential principle, to establish mathematical model, and to perfect and supplement the theoretical database. Finally, multi-effect regeneration technology can be studied to improve the regeneration efficiency, and finally promote the comprehensive development of waste AC regeneration field.

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