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Related progress of graphene oxide composites in the adsorption and killing of Escherichia coli

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Abstract:

Escherichia coli (E. coli) is one of the most common pathogenic bacteria in modern society, and its resistance to antibiotics has led to the consideration of the abuse of antibiotics. It is necessary to find alternative bactericidal substances to reduce the use of antibiotics and kill E. coli efficiently. Graphene oxide (GO) composites have great potential in killing bacteria and other aspects. This study reviews the progress of graphene oxide composites in the adsorption and killing of E. coli, and concluded that graphene oxide/silver ion (GO-Ag) is the best choice for killing E. coli and reducing the spread of drug resistance genes.

Keywords: graphene, graphene oxide composites, Escherichia coli, antibacterial property

1. Introduction

As an opportunistic pathogen, Escherichia coli (E. coli) can exist in the natural environment, daily food and the intestinal tract of humans and animals. Since the discovery of penicillin by the British bacteriologist Fleming in 1928, human beings have been relying on antibiotics to destroy E. coli. However, its overuse makes the drug resistance of E. coli uncontrollable, and its drug resistance has become an aspect of environmental health that cannot be ignored. Song H J et al.[1]found that more than 90% of E. coli strains isolated from edible animals in Korea were highly resistant to quinolones and cephalosporins between 2011 and 2020; In 2017, the World Health Organization (WHO) published an article indicating that bacterial antibiotic resistance has become a global public health problem. In recent years, the research on graphene oxide composites provides a new idea for killing E. coli.

Graphene is the two-dimensional embodiment of graphite, in which carbon atoms are connected to each other by sp^2 hybridization forming strong σ bond and π bond, forming a number of stable regular plane hexagonal structures[2]. As a new type of two-dimensional material discovered in recent years, it has good conductivity and barrier properties, and is one of the strongest materials in the world. However, graphene itself has many defects, such as weak electrochemical activity, easy agglomeration, and difficulty in processing and molding. Therefore, most of the modern research directions are to functionally modify it or make derivatives such as graphene oxide (GO) and reduced graphene oxide (rGO) to meet the needs of various scenarios.

2. Preparation of GO and its bactericidal mechanism

2.1 Preparation method of graphene oxide

One of the preparation methods of graphene is the physical peeling method. Novoselov K S et al. [2] used the physical peeling method to produce graphene. As one of the derivative products of graphene, the preparation principle of graphene oxide is also about peeling and breaking. There are many preparation methods for graphite oxide, such as Brodie method, Staudemnaier method and Hummers method, etc. [4]. The principle is that by reacting graphite with strong oxidizing agents, the conjugate structure of graphite is destroyed while it is oxidized to form a large number of oxygen-containing groups. Due to the advantages of easy access to materials and less pollution, the mainstream preparation method of graphene oxide is the Hummers method. The improved Hummers method [5] is as follows: First, a certain amount and proportion of strong oxidants such as concentrated H_2SO_4, H_3PO_4 and $KMnO_4$ were used to react with scale graphite in a strong acid environment, and then after stirring in an ice water bath for 1h, insulation reaction at 50℃ for 12h, and 5% volume fraction of dilute hydrochloric acid and deionized water to fully rinse and dry, graphite oxide was made. It was then chemically stripped down to single or few layers of GO.

2.2 Bactericidal mechanism of GO

The bactericidal mechanisms of traditional antibacterial antibiotics can be divided into several types: inhibition of bacterial cell wall synthesis, dysfunction of cytoplasmic membrane resulting in cytoplasmic leakage, inhibition of mitosis, obstruction of DNA replication, and inhibition of RNA polymerase action. The carbon atoms on graphene oxide can be divided into two types according to different hybridization methods. One is the unoxidized sp^2 hybrid region, and the other is the oxidized sp^3 hybrid region, which is composed of a six-membered aliphatic ring. After its functionalized modification and grafting with other molecular groups with positive surface charges, its own surface is positively charged, and it can effectively adsorb E. coli with negative surface charges, which can effectively avoid the emergence of drug resistance. Nowadays, due to the discovery and widespread use of antibiotics, bacteria have gradually evolved stronger drug tolerance and drug resistance [6]. Due to different mechanisms of action, graphene oxide composite materials can effectively adsorb and kill E. coli while avoiding the further development of drug resistance, which is a time transgressive material.

3. Inhibitory effect of graphene oxide and its composites on E. coli

3.1 Graphene oxide

Song S et al. [7] prepared graphite oxide using the improved method of Marcano D C et al. [4], and prepared six kinds of GO with different transverse sizes under different ultrasonic times. Finally, after preliminary comparison, GO-0 with the best antibacterial property (average transverse size 2.5μm) was selected for subsequent experiments. After testing the killing of E. coli and other bacteria under a variety of GO concentrations, the results showed that the killing rate of 100 μg/mL GO to E. coli was close to 99%. Compared with 200 μg/mL GO, the killing rate of E. coli was improved but not obvious (Fig. 1).

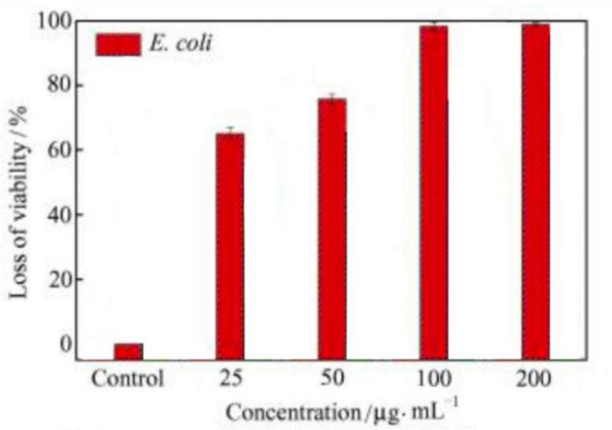

Figure 1 The killing rate of E. coli by different concentrations of GO in the experiment of Shuang Song et al

3.2 Binary graphene oxide composites

Pounraj S et al. [8] used graphene oxide nanosheets (GOns) and chitosan (ch) to prepare graphene oxide/chitosan (GOns-ch) and other composite materials. After testing, its inhibitory effect on the formation of E. coli membrane was not very ideal compared with other materials (Fig. 2). Chen Y. et al. [9] prepared graphene oxide (GO) -silver (GO-Ag) nanocomposites and tested their inhibitory effect on two types of E. coli containing different drug resistance genes in liquid and solid phases. The results showed that GO-Ag had excellent inhibitory effect on the growth of both types of E. coli, and the survival rate was less than 5% (Fig. 3). Under the action of photothermal therapy, 7 μg/mL GO-Ag could also completely kill E. coli in the material.

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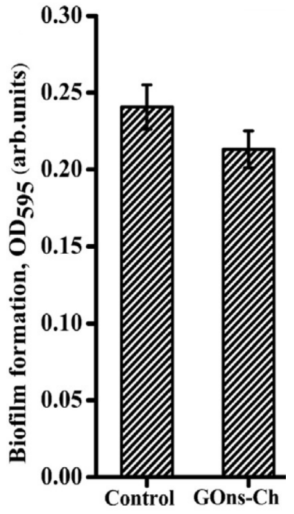

Figure 2 Effect of Pounraj S et al. experiments on inhibition of E. coli biofilms

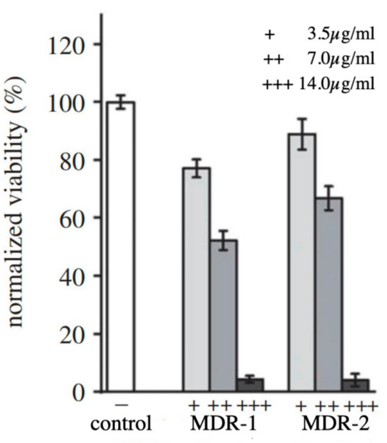

Figure 3 Survival rates of the two E. coli species in the experiments of Chen Y. et al

3.3 Multivariate graphene oxide composites

Wu Feng et al. [10] used dilution method and fluorescence quantitative gene amplification method to observe the minimum inhibitory concentration, antibacterial rate and the effect of removing drug resistance genes of GO-PEI-Ag nanocomposites with standard strains and drug-resistant strains of various bacteria as materials. The results showed that it had a good antibacterial property and the ability to remove drug resistance genes. The removal rate of E. coli was 100%. Khawaja H et al. [11] prepared GO, GO-CS, GO-Ag, and GO-CS-Ag with graphite powder, chitosan (CS), and Ag, and each material was prepared with a concentration of 1mg/mL and then tested against a variety of bacteria with 50μl. The minimum inhibitory concentration (MIC) value for Escherichia coli is shown in Table 1. It can be seen that GO-CS-Ag has the best antibacterial effect against E. coli at the same concentration.

4. Summary

According to Tab. 1, among the above GO materials, silver ion is the most effective composite raw material, and GO-Ag can be initially judged as the best E. coli killing material. Its advantage is that under the same killing rate, the material can be used in a small amount and save materials while killing E. coli efficiency is not reduced.

Table 1 Bactericidal rate and minimum inhibitory concentration of different GO composites against E. coli

Materials	Methods	Dosage/μg·mL^{-1}	Killing rate/%	MIC/μg·mL-1	Data sources
GO	Mix	100	99	35	[7]
GOns-ch	Mix	/	/	30	[8]
GO-Ag	Mix	14	95.75	4	[9]
	Photothermal therapy	7	100		
GO-PEI-Ag	Mix	10	100	4	[10]
GO-CS-Ag	Mix	/	/	8	[11]

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