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Graphene Photothermal Materials in Oncology Therapy

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Abstract:

Cancer, as one of the major causes of death worldwide, poses a serious threat to human health. Although traditional treatments such as surgery, chemotherapy and radiotherapy are effective, they have limitations such as high side effects and high recurrence rates. Graphene-based photothermal materials have become a hotspot for tumour therapy research due to their unique properties. This paper aims to analyse the therapeutic mechanism of graphene photothermal materials. This paper concludes that under near-infrared light irradiation, graphene and its derivatives can be efficiently converted photothermally to kill cancer cells, and can also be used as carriers for drug loading, and photothermal drug release can be achieved synergistically. Technical bottlenecks include low photothermal efficiency (narrow absorption, energy loss), shallow tumour penetration (only 1-2 cm of near-infrared light), easy agglomeration of the material, and biosafety to be verified, and the solutions include composite of other materials to broaden the absorption, the development of NIR-II responsive materials, and surface modification to improve dispersion, etc. The material can be used alone for photothermal therapy to kill cancer cells under near-infrared light irradiation. The material can be used for photothermal therapy alone, or in combination with chemotherapy (e.g. GO-loaded adriamycin), photodynamic (e.g. rGO-AuNP to produce heat and reactive oxygen species), immunotherapy, and some materials (e.g. NPGQDs) have low toxicity and a good tumour suppression effect in animal models. This study provides a reference for related fields and helps clinical translation.

Keywords: Graphene; photothermal materials; tumour therapy; combination therapy;

1. Introduction

Cancer is a serious threat to human health and is one of the leading causes of death worldwide. Traditional tumour treatments, such as surgery, chemotherapy and radiotherapy, although capable of controlling tumour development to a certain extent, all have their limitations. Surgical treatment is often difficult to completely remove some advanced or metastatic tumours; chemotherapeutic drugs kill tumour cells while causing damage to normal cells, resulting in serious side effects; and radiotherapy may lead to local tissue damage and has limited therapeutic effects on deep tumours. Therefore, the development of new, highly effective and low-toxicity tumour therapies is of great clinical significance and an urgent need.

Graphene, a two-dimensional material composed of carbon atoms, can be used in oncology therapy due to its unique structure and excellent physicochemical properties. For example, composites of polydopamine functionalized reduced graphene oxide loaded with AuPd bimetallic nanoparticles (AuPd-rGO/PDA) were prepared by the chemical reduction method by Punamshree Das et al. Under the irradiation of 915 nm near-infrared laser, the material can be warmed up to 51 ± 3 °C in 3 min at a concentration of 25 μg/mL for efficient cancer cell ablation with low toxicity to normal cells, and showed good biocompatibility in zebrafish embryo experiments, which provides a nano-platform combining both high efficiency and safety for photothermal therapy of cancer [1]. Nicolo Mauro et al. prepared folic acid-functionalized PEG-modified graphene oxide nanosheets (GO-PEG-Fol) by plasma etching, which can be efficiently loaded with adriamycin for targeted photothermal-chemotherapy synergistic killing of breast cancer cells under near-infrared light with low toxicity to normal cells [2]. Chunmei Wang et al. prepared polyphenol using Memecylon edule leaf extracts Chunmei Wang et al. prepared polyphenol-functionalized rGO from Memecylon edule leaf extract, which was highly effective in photothermal killing of lung cancer cells A549 under near-infrared light with good biocompatibility [3].

In this paper, we will systematically sort out the mechanism of graphene photothermal materials for tumour treatment, analyse the key technical bottlenecks and solutions, and provide systematic references for researchers in this field by integrating basic research and application technology, accelerating the clinical transformation of graphene photothermal materials, and ultimately providing safer and more efficient treatment options for tumour patients. This paper introduces the unique structure and properties of graphene and elaborates on its photothermal conversion principle and advantages. It discusses in detail the application results in photothermal therapy and combined therapy for tumours, analyses the challenges of photothermal efficiency, tumour penetration, dispersion and toxicity, and looks forward to the future development trends in clinical translation and interdisciplinary integration.

2. Properties of graphene and its derivatives

2.1 graphene

Graphene is a two-dimensional honeycomb lattice structure nanomaterial formed by sp^2 hybridisation of carbon atoms, with a variety of excellent properties. Mechanically, it is extremely strong, with a Young's modulus of 1,100 GPa and a breaking strength of 42 N/m, and at the same time flexible and stable, withstanding bending and stretching and maintaining structural stability. Electricity, carrier mobility up to 15000cm ^{2} / (V-s), low resistivity, the electron presents massless Dirac fermion characteristics (massless Dirac fermion characteristics refers to the Dirac fermion mass is close to zero, momentum and energy are linear) and room temperature quantum Hall effect (room temperature in a strong magnetic field in the two-dimensional electron gas Hall resistance presents the phenomenon of the quantum plateau), the specific conditions of the twisted 1.1° double-layer graphene will also appear superconducting phenomenon. Outstanding thermal performance, thermal conductivity of 5300W/(m-K), excellent heat dissipation efficiency. Optically, it is close to transparent at room temperature, with a nonlinear refractive index of $10^{-7}cm^2/W$ in the infrared region, suitable for optoelectronic devices. Chemically, the benzene ring skeleton is stable, the reactivity is concentrated at the boundary groups and defects, and derivatives such as graphene and graphene oxide can be generated through oxidation and reduction reactions, and their physicochemical properties change with the derivatisation process. These properties make graphene show important application potential in many fields [4].

2.2 GO and rGO

GO and rGO are important graphene derivatives with significant differences in properties. rGO consists of an isolated monomolecular graphite layer, with some of the carbon-carbon double bonds interrupted by oxygen-containing functional groups, resulting in some of the carbon atoms being converted from SP2 to SP3 hybridisation, and the surface is rich in -OH, -COOH, -O -OH, -COOH, -OOH, and other oxygen-containing functional groups on the surface. Its electrical properties can be adjusted by

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the oxygen-containing group coverage, type and arrangement, excellent optical transparency, thermal conductivity due to oxygen-containing functional group interference is less than graphene, with fluorescence and nonlinear optical properties, good chemical stability, large specific surface area, due to the strong charge and hydrophilicity, dispersed in aqueous solution and some organic solvents, poor electrical conductivity, insulating or semiconducting properties.

rGO is produced by removing part of the oxygen-containing functional groups on the surface of GO by chemical or physical methods, and its structure is between graphene and GO, retaining part of the sp^2 hybridised carbon skeleton with residual oxygen and structural defects. Its conductivity is greatly increased to 10^2~10^3 S/m, much higher than GO, and has high electron mobility, thermal conductivity and photocatalytic activity, light weight and soft, high Young's modulus. There are still some functional groups, such as hydroxyl and carboxyl groups on the surface, which are chemically active, with good physical and chemical stability and biocompatibility, and can be used in multiple fields through adsorption, doping, etc. to form composites with a variety of materials [5].

2.3 GQD

Graphene quantum dots (GQD) are carbon-based nanomaterials with both graphene sheet structure and quantum dot luminescence properties, which have significant potential for oncology applications. Optically, GQDs have strong absorption in the ultraviolet region, adjustable fluorescence, high quantum yield, good photostability, and upconversion luminescence; electrically, GQDs have strong ECL (electrochemiluminescence) stability, low onset potential, and N-doped electrocatalytic activity is close to that of Pt/C catalysts; the surface is rich in functional groups, which can regulate hydrophilicity and hydrophobicity, and is easy to be combined with drugs, etc.; thermally, they have high photo-thermal conversion efficiency, good thermal stability, and better biocompatibility than traditional quantum dots. Compatibility is better than traditional quantum dots and low toxicity. Based on these properties, GQDs can be used in photodynamic therapy (regulating ROS generation), photothermal therapy (compounding with other materials to generate heat), drug delivery (using the high permeability and retention effect of tumours to target drug release) and fluorescence imaging tracing (detecting circulating tumour cells), and their targeting and therapeutic efficiency in oncology diagnosis and treatment can be further enhanced by functional modification [6].

3. Graphene photothermal materials in oncology therapy

In this paper, we will focus on four applications of graphene photothermal materials in PTT, photothermal-chemotherapy, photothermal-photodynamic, and photothermal-photo-immunity.

3.1 Photothermal Therapy (PTT) for tumours

Graphene and graphene oxide (GO) are widely used in photothermal therapy (PTT), based on the principle that the material is irradiated by laser light to produce heat efficiently to kill cancer cells, and that reduced graphene oxide (rGO) is superior to GO due to stronger light absorption.

In the excitation mode, single photon excitation commonly used 808nm diode laser, PEGylated and other functionalized graphene nanosheets or rGO can be passively targeted to the tumor, such as PEGylated graphene nanosheets in the power density of 2W/cm² under the 808nm laser, the temperature of the tumour up to 50 ° C to achieve the ablation; and magnetic nanoparticles composite can be guided by magnetic resonance imaging treatment, and semiconductor quantum dots or Composite with magnetic nanoparticles can guide treatment by magnetic resonance imaging, and composite with semiconductor quantum dots or gold nanoparticles can achieve imaging guidance. After optimising the materials, nano rGO (nRGO-PEG) can be used for effective treatment at a low power of 0.15W/cm². Two-photon excitation using femtosecond pulsed laser, using the advantages of deep tissue penetration of near-infrared light and low background signal, GO is irradiated to rapidly reduce and release gas to form microbubbles, and its rupture produces microcavitation effect to enhance the mechanical damage of the cells, e.g., 4mW femtosecond laser irradiation of the cells labelled with GONs, the required power is reduced by more than 10 times compared to that of the cells without GONs.

When photothermal and chemotherapeutic synergistic treatment, GO or rGO is used as a carrier to load drugs such as adriamycin, and the laser generates heat to promote the release of drugs, and the two synergistically kill the cancer cells, and the in vitro experiments show that the combined treatment effect is the best with the increase of the drug concentration under the fixed laser intensity, and the in vivo experiments also confirm that the tumour can be ablated completely, such as the effect of the combined treatment of the PEGylated GO loaded with DOX under the laser of 2W/cm², is better than that of monotherapy. PEGylated GO loaded with DOX at 2W/cm² laser is better than monotherapy.

However, the technology faces some challenges; the non-biodegradable graphene-based material may lead to in vivo accumulation, the impact of microbubbles generated by GO under femtosecond laser on the in vivo environment needs to be assessed, and the clinical translation needs to be clarified in terms of long-term pharmacokinetics, biodistribution, and toxicity [7].

3.2 Combination therapy with chemotherapy

Breast cancer is a highly invasive tumour, often accompanied by distant metastases, which seriously threatens the life and health of patients [8]. In recent years, photothermal-chemotherapy combination therapy has received widespread attention in breast cancer treatment due to its synergistic effect. In the combined photothermal-chemotherapy treatment for breast cancer, the NGO-PEG-DOX system was modified by polyethylene glycol and loaded with adriamycin via graphene oxide nanoparticles. After 3 min of irradiation with 808 nm near-infrared laser (2 W/cm²), the tumour temperature increased from 26°C to 50°C, accelerating adriamycin release and enhancing cellular thermal sensitivity, and the in vitro inhibitory rate of EMT6 cells was better than that of free adriamycin and NGO-PEG. The results showed that tumours in 4 out of 5 mice treated were completely ablated after 1 day of treatment, and in the follow-up observation of The results showed that 4 of the 5 mice treated had complete ablation after 1 day of treatment, and none of the tumours recurred during the 40-day follow-up period.DTX-GO/CS temperature-sensitive hydrogel was formed by chitosan and graphene oxide loaded with docetaxel, and the 808 nm laser (2.5 W) irradiation produced heat to promote the release of docetaxel, which synergistically exerted the chemotherapeutic and photothermal effects. The inhibitory rate of the DTX-GO/CS temperature-sensitive hydrogel was higher than that of the no-laser group, and the volume and weight of tumours of the S180 loaded mice after 12 days of treatment were higher than those of the no-laser group. Tumour volume and weight were significantly reduced after 12 days of treatment. As for nanomaterial properties, the near-infrared absorption of nanoscale reduced graphene oxide at 808 nm was 3-4 times higher than that of graphene oxide nanoscale, and the tumour temperature was higher under low-power irradiation, with better photothermal efficiency. The temperature of the UDPs modified by polydopamine coating was significantly elevated in the near-infrared after the loading of adriamycin on the ultra-small graphene oxide nanoscale sheets, which confirms that the polydopamine enhances the photothermal effect and the reduced graphene oxide's NIR absorption is 3.2 times stronger than graphene oxide, and the photothermal conversion efficiency is higher. In other combined systems, GO-AuNP-Apt-DOX was combined with MUC1 aptamer-modified graphene oxide and loaded with adriamycin via gold nanospheres, and the release rate of adriamycin under NIR irradiation exceeded 80% for 2 h, which resulted in a better killing effect of MCF7 cells, and P-DOPA-rGO was obtained from polydopamine reduced graphene oxide, and cell survival rate only decreased by 36°C with the increase of NIR irradiation in a 3D tumour ball model. °C but cell survival only decreased to 30%, suggesting that parameters need to be optimised to enhance deep tumour penetration [5].

3.3 Combined treatment with photodynamic therapy

Graphene and its derivatives show significant application potential in photothermal-photodynamic therapy. Graphene-based materials such as rGO and GO can efficiently absorb near-infrared light and convert it into thermal energy for photothermal therapy (PTT), like BSA-modified rGO loaded with adriamycin that can elevate the temperature of the tumour and promote the release of the drug under the 808 nm laser irradiation, and folate-chitosan-modified GO that can be targeted to the tumour and inhibit the tumour by laser irradiation with a temperature rise of 57.6 °C. A composite of graphene and magnetic nanoparticles, such as FNPs/rGO-PEG, can be guided to the tumour site with the help of a magnetic field, and combined with the photothermal effect to enhance the anti-tumour effect. In photodynamic therapy (PDT), graphene can be used as a photosensitizer carrier, such as PEGylated graphene oxide nanoparticles wrapped with TPEred, to generate reactive oxygen species to kill tumour cells under light, and GO-loaded ICG combined with chemotherapeutic drugs to kill osteosarcoma cells with significant effect. Combined photothermal-photodynamic therapy, such as rGO-AuNP under NIR-II laser irradiation, simultaneously exerts a photothermal effect and induces ROS generation, which has better efficacy on deep-seated tumours, and graphene quantum dots under ultraviolet light can also generate reactive oxygen species to achieve efficient killing of cancer cells. However, this field still faces challenges such as biosafety and deep tumour penetration, and material properties need to be optimised in the future to promote clinical translation [4]. In addition, Liu et al. synthesized nitrogen-phosphorus co-doped graphene quantum dots (NPGQDs) with a diameter of about 4 nm by a one-pot hydrothermal method using murine erythrocyte membranes as a precursor, which efficiently catalyzed the decomposition of hydrogen peroxide to generate hydroxyl radicals under weakly acidic conditions in

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the tumour microenvironment and inhibited tumours by inducing apoptosis and iron death. In vivo experiments showed that intravenous and intratumoural injections of NPGQDs inhibited the tumour growth of triple-negative breast cancer mice by 77.71% and 93.22%, respectively, and were biocompatible, providing a new strategy for the use of metal-free nanoenzymes for cancer chemodynamic therapy [9].

3.4 Combination therapy with immunotherapy

Non-stoichiometric CoWO4-x nanoplatforms in combination with immunotherapy can enhance the tumour suppression effect of phototherapy by reducing HSP60- and NRF2-mediated immune resistance with high biosafety, providing a new idea for tumour treatment. The researchers designed a PEO-b-MAA-modified non-stoichiometric CoWO4-x nanoplatform, which can simultaneously perform photothermal and photodynamic therapeutic roles under 808 nm laser irradiation, and also act as a contrast agent for CT and photoacoustic imaging. The platform has a photothermal conversion efficiency of 72.75%, and the tumour temperature can be increased from 35°C to 61.7°C under laser irradiation with the generation of reactive oxygen species, with an in vitro inhibition rate of 88.2% against 4T1 cells, which is superior to monotherapy. In in vivo experiments, intravenous and intratumoural injections combined with laser irradiation resulted in tumour inhibition rates of 77.71% and 93.22%, respectively. Although photothermal and photodynamic therapy can induce immunogenic cell death and release HMGB1 and calreticulin to activate the immunity, it will promote the overexpression of HSP60 and NRF2 in the tumour microenvironment to form immune resistance, leading to tumour recurrence. In contrast, injection of etoposide, an HSP60 inhibitor, and ML385, an NRF2 inhibitor, weakened the immune resistance, significantly enhanced the therapeutic effect, inhibited tumour recurrence and prolonged the survival of mice. Body weight monitoring and H&E staining in mice indicated that the nanoplatform was biocompatible and had no significant toxicity. This study provides a new strategy to enhance tumour photothermal-immunotherapy by suppressing immune resistance, showing good potential for clinical application [10].

4. Bottlenecks and solutions

4.1 Bottlenecks in improving solar thermal efficiency

In the field of photothermal therapy for the treatment of tumours, there is an obvious bottleneck in photothermal efficiency. Traditional photothermal therapy needs to maintain a high temperature of over 50°C, which requires a long time of high-power laser irradiation, which can damage healthy tissues, and the defence mechanisms, such as heat shock protein expression and autophagy of cancer cells also greatly reduce the effect of photothermal therapy. At the same time, photothermal agents themselves also have shortcomings, such as poor biodegradability of inorganic materials, low photothermal conversion efficiency of organic materials, coupled with the limited depth of laser penetration, which have severely limited the effectiveness of photothermal therapy.

However, there are now some breakthrough strategies. For example, the use of photothermal agents that inhibit heat shock proteins, like the Mn - ICG@pHis - PEG/GA nanosystems; as well as methods to target organelles, like nuclear-targeted gold nanorods; and means of interfering with autophagy, like PDA - PEG/CQ nanoparticles. In addition, combination therapy strategies are also effective, combining photothermal therapy with ROS therapy, gas sensitisation, chemotherapy, etc., which can enhance the therapeutic effect. There is also the development of photothermal agents with high conversion efficiencies, like black phosphorus and V_2C quantum dots, as well as the use of near-infrared region II lasers to enhance the depth of penetration, which provides a new direction to break the bottleneck of photothermal efficiency [11].

4.2 Biosafety and toxicity issues

In photothermal therapy, biosafety and toxicity issues are of great concern. Graphene and its derivatives as a photothermal agent, its toxicity is affected by the size, surface charge, the number of layers and other factors, such as high concentrations of graphene will trigger cellular oxidative stress, destruction of cell membranes, long-term retention in the body may also lead to lung inflammation and organ damage, and inorganic materials with poor biodegradation also have potential risks.

However, there are some breakthroughs. Through surface functionalization modification, such as polymer coating with PEG, chitosan, etc., biocompatibility can be improved, toxicity can be reduced and cycle time can be prolonged; the development of biodegradable organic photothermal materials, which can reduce long-term toxicity; precise targeting design, like nuclear targeting or mitochondrial targeting, which can improve therapeutic efficiency, and reduce the damage of the normal tissues; combined treatment strategy, combining photothermal therapy with chemotherapy, immunotherapy, etc., can reduce the dose and side effects of monotherapy. In addition, the application of near-infrared region II laser can

improve the depth of tissue penetration and reduce the damage to superficial tissues. These measures provide a direction to solve the biosafety and toxicity problems of photothermal therapy [4].

5. Future Development Trends

In the field of cancer photothermal therapy, graphene-based nanomaterials show remarkable potential but still face many challenges. On the one hand, the biosafety and in vivo metabolism of the materials need to be urgently addressed, such as the toxicity and long-term retention risk of metal-based nanoenzymes, and the long-term biological effects of even metal-free N/P co-doped graphene quantum dots (NPGQDs) need to be thoroughly investigated [9]. On the other hand, the photothermal stability and deep tumour penetration of the materials are insufficient, like gold nanorods (AuNRs) are prone to morphological changes under prolonged laser irradiation, resulting in shifted light absorption properties, and the near-infrared one-region (NIR-I) light penetration is limited in depth, making it difficult to effectively act on deep tumours. In addition, the stability and targeting of materials in complex biological environments need to be improved, for example, graphene oxide (GO)-coated AuNRs may aggregate in physiological media, affecting the therapeutic effect, and breakthroughs are still needed in how to achieve precise targeting of tumours [12].

However, the field also shows great promise. The development of highly biocompatible and degradable graphene-based nanomaterials has become an important direction, such as the synthesis of NPGQDs using biofilm-derived materials, which provides a new idea for safe therapeutics [9]. The application of near-infrared two-region (NIR-II) light is expected to solve the problem of penetration depth, and the composites combining NIR-II-absorbing AuNRs with GO or graphene quantum dots (GQDs) can enhance the effect of deep tumour therapy. Optimising the catalytic activity and photothermal properties of the materials through heterogeneous atom doping (e.g., N and P co-doping) can enhance the responsiveness to the tumour microenvironment. In addition, the construction of multifunctional integrated nanoplatforms to combine photothermal therapy with chemotherapy and immunotherapy can achieve synergistic effects, such as the composite system of GO or GQDs with AuNRs, which shows potential in photothermal-induced drug release [12].

6. Conclusion

Graphene-based nanomaterials have unique advantages in cancer photothermal therapy.NPGQDs, as metal-free nanoenzymes, induce apoptosis and ferroptosis of tumour cells by catalysing the generation of hydroxyl radicals from hydrogen peroxide in the tumour microenvironment, significantly inhibiting the tumour growth without significant toxicity in animal models, providing a feasible pathway for drug-free catalytic therapy. The composites of AuNRs and composites of AuNRs with GO or GQDs, after surface functionalization, can improve the stability and photothermal properties, and show good photothermal killing effects in both cell experiments and animal models. Despite the current challenges of material design, biosafety, and clinical translation, graphene-based nanomaterials are expected to play an important role in precision cancer therapy through rational material engineering design and multidisciplinary cross-fertilisation, promoting photothermal therapy towards clinical applications. Future research should focus on the scale-up production of the materials, long-term safety assessment and optimisation of the targeted delivery system to accelerate its clinical translation.

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