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REVIEW

A review on graphite surface modification methods towards low carbon-containing refractories

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Abstract

The rapid development of iron and steel metallurgy technology has promoted the continuous innovation and iteration of carbon-containing refractories for clean steel smelting. To meet the high-quality requirements for clean steel production and full exploit the performance advantages of carbon-containing refractories in dynamic smelting environment, it is necessary to explore the role of graphite and modified graphite in carbon-containing refractories. Based on this, graphite surface modification methods, including surfactants, surface oxidation, and surface coating, and their applications in carbon-containing refractories are reviewed. The advantages and disadvantages of each method are analyzed for practical use. Furthermore, combined with the existing problems, the application prospect of improved graphite in carbon-containing refractories is discussed.

Keywords Graphite · Carbon-containing refractory · Clean steel · Surface modification · Service performance

1 Introduction

Over the past ten years, the steel industry has grown rapidly. At the same time, increasing demands have been put forward to improve the quality and performance of clean steel in industrial production. Carbon-containing refractories are the cornerstone to support and promote the development and progress of the steel industry [1–7]. Graphite is an important carbon source in carbon-containing refractories for clean steel smelting, and its inherent typical layered structure (Fig. 1) provides excellent chemical stability and a low coefficient of thermal expansion [8–11], which can effectively improve the thermal shock resistance and slag corrosion resistance of carbon-containing refractories [12, 13]. However, in the context of energy conservation, emission reduction and green development, traditional carbon-containing refractories have become low-carbon refractories. Moreover, a decrease in carbon content also leads to a decrease in the performance of carbon-containing refractories. In addition, graphite is highly susceptible to oxidation reactions in smelting environments and forms pores within the material, aggravating the erosion of carbon-containing refractory by slag, reducing service life and causing liquid steel pollution [13–15]. Adding antioxidants and modifying graphite are two common methods used to improve the performance of carbon-containing refractories [16–22]. The addition of antioxidants effectively alleviates the oxidation of graphite at high temperatures [23–32]. Table 1 lists commonly used antioxidants (metal element, carbide, boride, nitride, and oxide) [33–46]. Nevertheless, the addition of antioxidants to carbon-containing refractories causes defects such as hydration of by-products, liquid phase formation at high temperatures, and volume expansion. These problems adversely affect the mechanical performance and service life of carbon-containing refractories [47].

Graphite surface modification is an effective anti-oxidation technology, and with the development of science and technology, modification methods to improve the surface performance of graphite are also emerging [48, 49].

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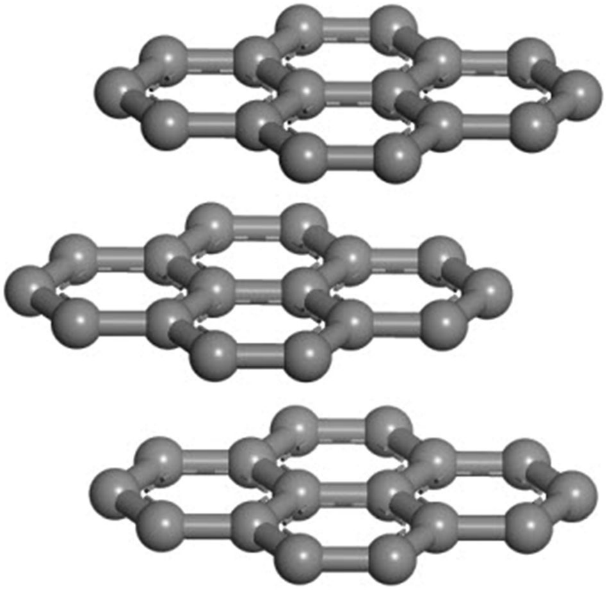

Fig. 1 Layered structure of graphite

Table 1 Commonly used antioxidant types of carbon-containing refractories [33–46]

Antioxidant type	Example	Reference
Metal element	Al, Si, Mg	[33–36]
Carbide	SiC, Al_4SiC_4, $Al_4Si_2C_5$	[37–39]
Boride	ZrB_2, AlB_2, MgB_2	[40–42]
Nitride	Si_3N_4, AlN	[43, 44]
Oxide	TiO_2, Y_2O_3	[45, 46]

Through surface modification and structure optimization of graphite to improve the performance of carbon-containing refractories, it is expected to further promote the development of clean steel smelting [50, 51].

2 Graphite surface modification methods

Graphite surface modification technology can effectively improve the oxidation and thermal shock resistance of carbon-containing refractories, which plays an important role in ensuring the cleanliness of molten steel. Researchers have proposed numerous ideas for surface modification. Modified graphite can solve its problems to a certain extent and broaden its scope of use through different modification methods [52]. Surfactants, surface oxidation, and surface coating are the more extensively implemented graphite modification methods, combined with the service conditions of carbon-containing refractories for clean steel smelting [53–57]. In this study, these three methods are summarized, and their advantages and disadvantages in improving the high-temperature performance of carbon-containing refractories are analyzed. In addition, there are other graphite modification methods such as plasma method, refining particle method, graphite granulation method and low temperature calcination method. The application of these methods (Fig. 2) has not been maturely used to improve the performance of carbon-containing refractories, and thus, they have not been analyzed in this study [58–61].

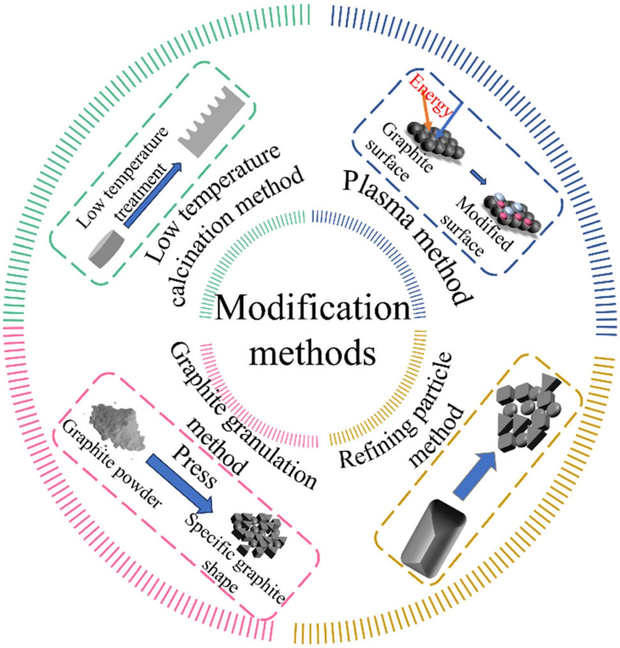

Fig. 2 Other graphite surface modification methods [58–61]

2.1 Surfactant method

Carbon-containing refractories have a low thermal expansion coefficient, good thermal shock resistance, excellent corrosion resistance, and have evident advantages in the field of clean steel smelting; however, the disadvantages including unsatisfactory oxidation resistance and poor fluidity have been the focus of researchers. Surfactants can improve the dispersibility and other performance of graphite materials without destroying their surface structure [62, 63]. According to the actual application environment of graphite, selecting suitable surfactants to improve surface performance is conducive to optimizing the service performance of carbon-containing refractories. Yoshida et al. [64] prepared lanthanum ultraphosphate (LaP_5O_{14}) crystals on the graphite surface. LaP_5O_{14} did not coat the graphite but improved its oxidation resistance by blocking the active sites on the graphite surface. LaP_5O_{14} crystals can significantly improve the anti-oxidation performance of graphite powder in oxidation experiments at 900 °C in air. Modified graphite has great potential for improving the oxidation resistance of carbon-containing refractories. Yi et al. [65] used different concentrations of dodecyltrimethylammonium bromide (DTAB) as an active agent to explore its effect on the coating wettability of graphite. The results show that the coating effect of different concentrations of DTAB on the graphite surface varied, and the coating rate of DTAB on the graphite surface significantly increased from 3.71% to 85.44% with the increase in the active agent concentration. DTAB was found to aggregate on the graphite surface in the form of streaks. In the wettability experiments, the hydrophilicity of DTAB-coated graphite was substantially improved, which effectively improved the performance of carbon-containing refractories.

Carbon nanotubes and graphene are important sources for carbon-containing refractories, and many researchers have performed surface treatment to enhance the service performance of carbon-containing refractories [66, 67]. Li et al. [68] prepared carbon nanotubes and magnesium oxide composite powders using nickel nitrate hexahydrate ($Ni(NO_3)_2 \cdot 6H_2O$) as an active agent and investigated their effects on the performance of low-carbon carbon-containing refractories. The results show that carbon nanotubes had a nodular hollow structure and could improve the mechanical performance and thermal shock resistance of the low-carbon alumina–carbon refractories. Khan and Mariatti [69] used graphene oxide as a carbon source and investigated the effect of natural surfactants, such as gum arabic, cellulose nanocrystals, and alkali lignin, on the performance of graphene oxide, which promoted the application of modified graphite in carbon-containing refractories. Comparing the contact angles of modified graphene without surfactant and with gum arabic, cellulose nanocrystals and alkali lignin as surfactants (Fig. 3), the contact angle of graphene without surfactant was approximately 76°, whereas the contact angle of graphene treated with alkali lignin was reduced to 38°. The wettability of the material significantly was improved, which increased the applicability of modified graphite in carbon-containing refractories.

Surfactants can increase the type and number of oxygen-containing functional groups on the graphite surface and improve wettability. However, this method has limited influence on the surface structure of graphite. The bonding strength between the surfactant and the graphite matrix is weak, which is prone to adsorption instability, accelerates the oxidation of graphite and reduces the resistance of carbon-containing refractories to slag erosion and thermal shock. These phenomena are not conducive to the production and preparation of clean steel. Therefore, it is necessary to select excellent active agents and optimize the modification process to enhance the bonding effect between the active agent and graphite matrix.

2.2 Surface oxidation method

Surface oxidation treatment of graphite can improve the surface energy of the material and the bonding effect between the material and other substances. Surface oxidation methods can be divided into gas-phase and liquid-phase oxidations. The gas-phase oxidation method has the advantages of short reaction time and continuous treatment, and researchers have applied it in the field of modified graphite [55, 70]. Xie et al. [71] preoxidized the graphite sample in air at 900 °C and then prepared coating on the surface of the sample. The results show that the surface of the sample had a porous and rough structure after oxidation (Fig. 4). This structure promoted the interlocking of the graphite material and coating, enhanced the bonding strength, and effectively improved the defects in the coating, which could be easily peeled off. In addition, the preoxidized sample could effectively mitigate crack extension and substantially improve its oxidation resistance. Jing et al. [72] preoxidized a carbon material in air and synthesized SiC nanotoughened buffer layers in situ on a specimen surface. The results show that a porous structure was formed on the surface of the sample after the preoxidation treatment, which provided a path for the formation of a buffer layer between the coating and material, enhanced the surface roughness of the material, and improved interfacial bonding. Moreover, the introduction of SiC nanowires inhibited the expansion of microcracks, resulting in excellent thermal shock resistance of composites.

The liquid-phase oxidation method results in a mild oxidation behavior of the material, which is less likely to

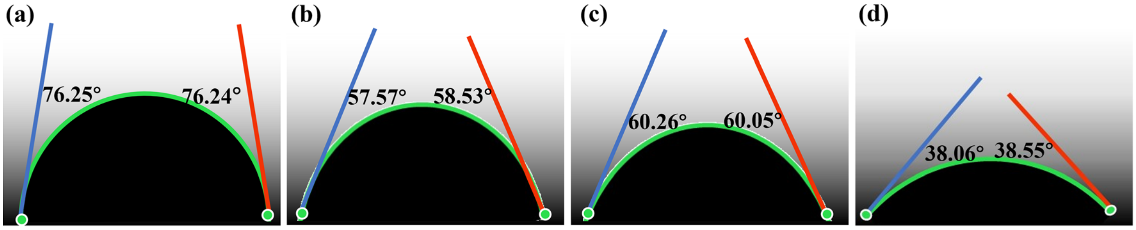

Fig. 3 Contact angle of graphene treated with different active agents. **a** Without surfactant; **b** with gum arabic; **c** with cellulose nanocrystals; **d** with alkali lignin [69]

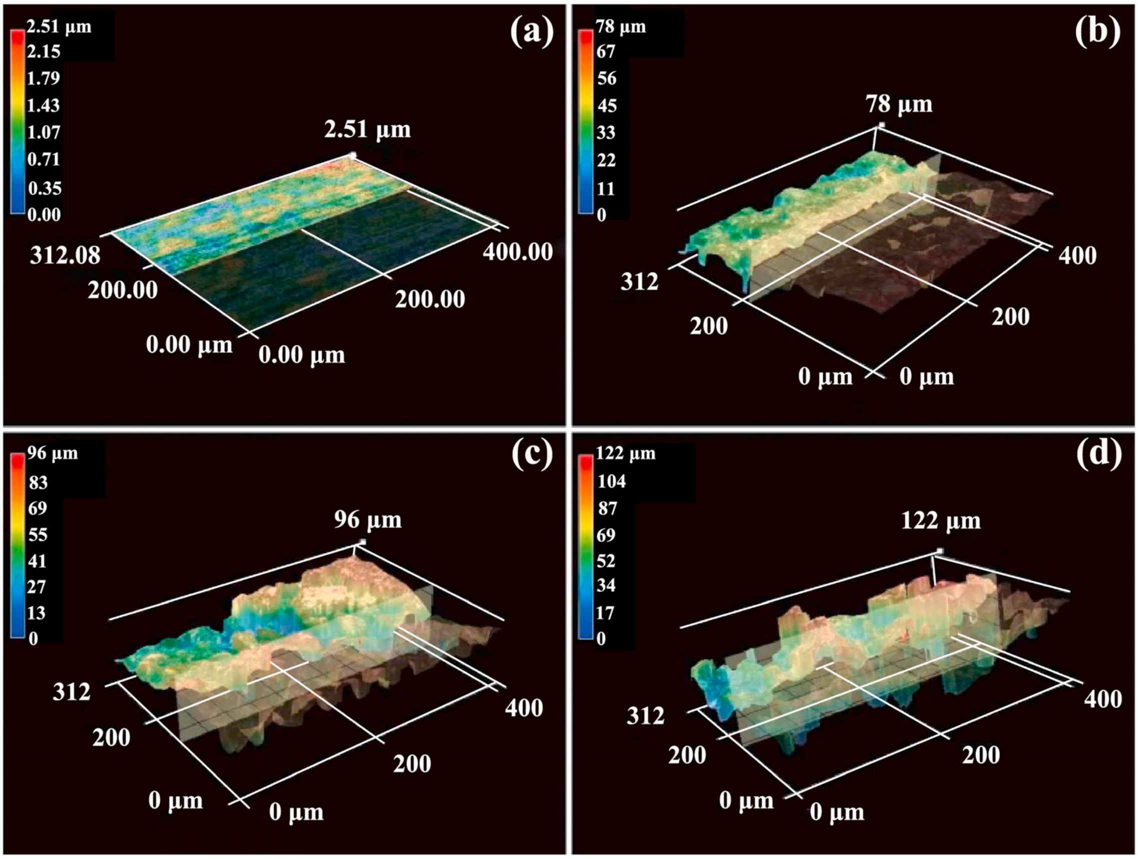

Fig. 4 Change of graphite surface with preoxidation time. **a** 0 min; **b** 5 min; **c** 10 min; **d** 20 min [71]

cause excessive etching of the material, and the application range is wide. Saberi et al. [73] used hydrogen peroxide (H_2O_2, 30%) and nitric acid (HNO_3, 65%) as oxidants to preoxidize flake graphite and prepared $MgAl_2O_4$ spinel to coat graphite. Water wettability and oxidation resistance experiments were carried out, and it was found that water contact angle of the original graphite was 88°. The water contact angle of preoxidized graphite could be reduced to 52°, and preoxidized graphite had a higher onset oxidation temperature than virgin graphite. Through the preoxidation treatment, the water wettability and oxidation resistance of graphite coated with $MgAl_2O_4$ spinel were substantially improved, and homogenization and densification of the coating were also promoted. Cho et al. [74] used a mixture of sulfuric and nitric acid to acidify the flake graphite and then modified the graphite with an aluminum nitrate solution to prepare modified graphite coated with Al precursor. By comparing the oxidation experiments of graphite before and after modification, the original graphite started to be oxidized at 700 °C, while the modified graphite started to undergo oxidation at 900 °C. The oxidation resistance of graphite after surface oxidation treatment was significantly improved.

Oxidation modification of graphite has the advantages of simple operation, low cost, and good bonding. However, oxidation modification method causes environmental pollution, and this method often needs to be combined with the coating method.

2.3 Surface coating method

Surface coating methods refer to the preparation of a layer of covering layer with good stability and protection on the surface of graphite material by physical, chemical, or other methods to modify the structure of surface of graphite materials. In recent years, surface coating methods have been widely used in graphite modification because of excellent protective effect. This method has a great effect in improving the oxidation resistance of graphite and the service performance of carbon-containing refractories [75–77]. Figure 5 shows that some researchers modified the surface of graphite by coating method and explored the change in water wettability of modified graphite and their effect on oxidation resistance and corrosion resistance of carbon-containing refractories [67, 78–85]. Mukhopadhyay et al. [86] prepared calcium aluminate ($CaAl_2O_4$) coating on graphite surface using the sol–gel method. The

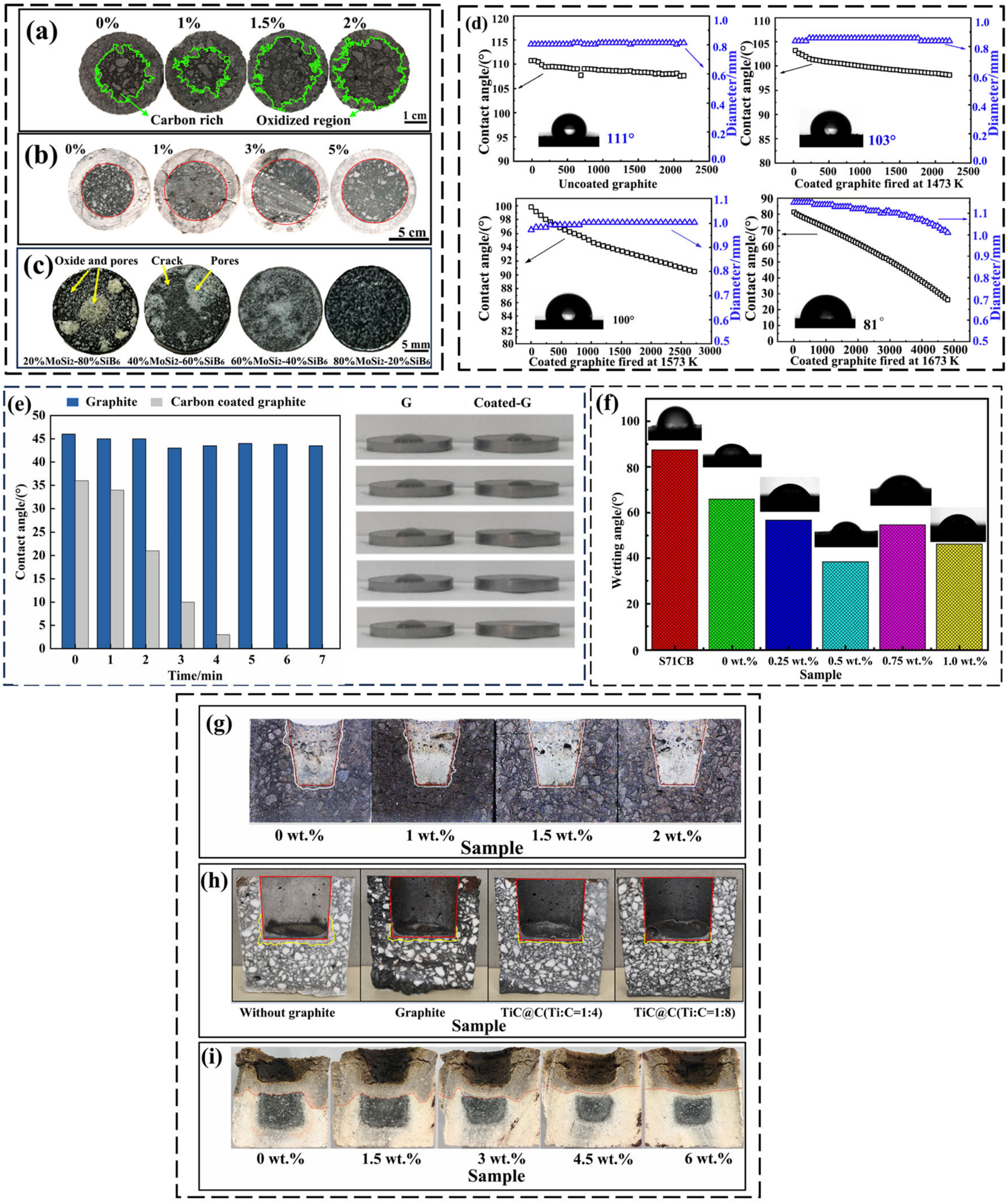

Fig. 5 Effect of coating method on water wettability of graphite and oxidation resistance and corrosion resistance of carbon-containing refractories containing coated graphite. **a** Silicon carbide whiskers coated graphite; **b** zirconium carbide coated graphite; **c** $MoSi_2$–SiB_6 coated graphite; **d** SiC/SiO_2 coated graphite; **e** carbon coated graphite; **f** S71C and CNFs@CAC with different contents of iron catalyst; **g** silicon carbide modified graphite with different contents; **h** TiC@C powders; **i** $C/MgAl_2O_4$ composite powders with different contents [67, 78–85]

hydrophilicity of graphite was improved by increasing hydrophilic groups. Additionally, the oxidation resistance and thermal stability of the modified graphite in an oxidation environment were better than those of the original graphite. Grashchenko et al. [87] prepared SiC coating on the surface of graphite materials and the resulting coating consisted of branched structures of dendritic SiC crystals extending into the interior of the graphite matrix. Special growth direction enhanced the bonding strength between the coating and graphite material. The composite coating significantly improved hardness of the material, and the composite material was 254 times harder than the original

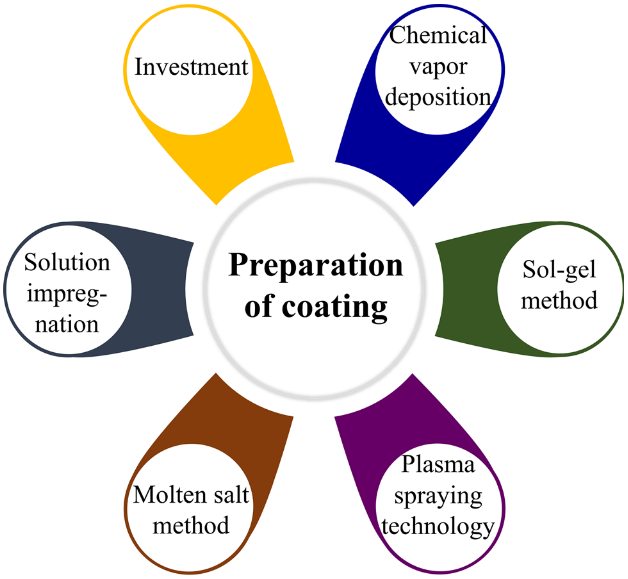

Fig. 6 Methods of preparing coatings on graphite surface [88–95]

graphite surface hardness. The anti-oxidation performance of the treated graphite materials was also improved. Mukhopadhyay [88] prepared mullite and spinel on graphite surface by using sol–gel method and introduced modified graphite into the alumina-based carbon-containing refractories. The coated graphite had good hydrophilicity and oxidation resistance and improved the corrosion resistance of carbon-containing refractories.

At present, there are plenty of methods of preparing graphite surface coating, and some of them have achieved good results and have played an important role in enhancing performance of carbon-containing refractories [88–95]. Figure 6 shows some common methods used to prepare coatings. Liu et al. [80] and Chen et al. [96] prepared $MoSi_2$–SiB_6 and $MoSi_2$–SiC coatings on the surface of graphite materials, respectively. Both coatings have the characteristics of being dense and crack-free, and after high-temperature oxidation, the surface of coatings generates SiO_2 film that reduces the diffusion of oxygen effectively and improves the anti-oxidation performance of modified graphite. Yang et al. [97] prepared TiB_2 coatings on the surface of graphite, and the coatings and the internal materials crossed each other, enhancing the bonding between them. There are no cracks on the sample surfaces through water quenching and thermal shock test. The modified carbon material has better high-temperature corrosion resistance.

The surface coating methods are used to modify graphite, which often only changes the surface of graphite matrix material without destroying the overall performance and structure. Therefore, modifying graphite by coating method does not have a significant negative impact on the comprehensive performance of carbon-containing refractories. Strong bonding effect exists between the coating and the substrate that is not easy to fall off. However, the discrepancy in the thermal expansion coefficient between the coating and substrate led to poor toughness of the material interface and brittle fracture under thermal shock [76]. When preparing coated graphite, the coating material with a suitable thermal expansion coefficient should be selected to better exert the excellent oxidation resistance and thermal shock resistance of carbon-containing refractories.

In summary, using an active agent to modify graphite effectively improves its surface characteristics, but poor high-temperature stability causes graphite material to be oxidized and aggravates the erosion between carbon-containing refractories and molten steel. Graphite treated by oxidation has a better surface energy, but this method is often combined with the coating method when used in carbon-containing refractories, which increases the workload to some extent. Coating is an ideal method of graphite modification that has made great progress in improving oxidation resistance of carbon-containing refractories. Therefore, the surface coating methods of modified graphite are mainly analyzed and summarized, and their influence on the performance of carbon-containing refractories is discussed.

3 Types of graphite surface coating

The coating protection method refers to coating a layer of compound on the surface of graphite. This method effectively prevents the chemical reaction between water vapour, oxides, and other environmental substances with graphite materials. It significantly slows down the oxidation and erosion of graphite and improves the water wettability of graphite to improve the service performance of carbon-containing refractories in clean steel smelting. Table 2 shows the effect of diverse coatings on the performance of carbon-containing refractories for clean steel smelting [84, 98–103]. Figure 7 shows the coatings according to different classifications [104–110]. At present, the coatings used to modify graphite mainly include oxide coatings, nitride coatings, and carbide coatings, and these modified graphite coatings are mainly summarized.

3.1 Oxide coating

Oxide coatings significantly enhance the flowability of castable refractories due to their superior water wettability. Oxides exhibit high wear resistance, excellent anti-oxidation and corrosion resistance. They are widely used in the manufacturing of metallic materials, petrochemical equipment, wear-resistant machinery, and other related fields in the metallurgical industry [111, 112]. Numerous studies

Table 2 Effect of different coatings modified graphite on carbon-containing refractories [84, 98–103]

Coating preparation method	Resistance to oxidation	Wettability	Reference
Preparation of SiC whisker coatings by molten salt method	At 1200 °C, modified graphite had no obvious mass loss in air	Contact angle reduced from 90° to 10°	[84]
Preparation of $MgAl_2O_4$ on graphite surface by sol–gel method coating	Starting oxidation temperature increased from 700 to 850 °C, and oxidation rate decreased from 0.057 to 0.048 g/°C	Contact angle reduced from 88° to 40°	[98]
Preparation of Cr_3C_2 coating by molten salt method	–	Contact angle reduced from 101° to 75°	[99]
Preparation of Al_2O_3/SiO_2 coating by sol–gel method	Oxidation of modified graphite reduced from 70% to 39% at 1300 °C	–	[100]
Preparation of $TiSi_2$–Si–SiC/SiC bilayer coatings by two-step filler cementation method	Oxidized in air at 1500 °C for 200 h, coated graphite spheres only gained 0.96 wt.% mass	–	[101]
Preparation of nano-SiC/SiC bilayer coatings by electrophoretic deposition	After oxidation at 1600 °C for 28 h, mass loss value of double-layer coating decreased from 29 to 2.4 wt.% compared to single-layer coating	–	[102]
Preparation of mullite and spinel coatings by sol–gel method	At 1200 °C, oxidation resistance of spinel and mullite-coated graphite increased by 16.0% and 8.3%, respectively	–	[103]

have shown that oxide coatings such as ZrO_2, TiO_2, and Al_2O_3 effectively improve the oxidation resistance and wettability of graphite, thus preventing the erosion and oxidation of carbon-containing refractories in steelmaking operations, which has a positive effect on clean steel smelting [113–115]. Sunwoo et al. [114] first synthesized a uniformly textured ZrO_2 precursor layer using zirconium chloride and polyvinyl alcohol as raw materials. The ZrO_2-coated graphite powder was subsequently prepared by hydrolysis for investigating the oxidation resistance of carbon-containing refractories for clean steel. The improvement in oxidation resistance of modified graphite plays an important role in the oxidation resistance of carbon-containing refractories. Lin et al. [116] prepared ZrO_2 (–Y_2O_3)-coated graphite with a core–shell structure, which improved the wear resistance and oxidation resistance of graphite due to the unique three-dimensional structure of the coating layer and the good physical and chemical performance of the ZrO_2 ceramic phase. Kim et al. [117] used graphite as a substrate and enhanced its surface roughness by coating it with SiO_2 powder through chemical vapor deposition and thermal decomposition processes. Then, Y_2O_3 and 8 mol% Y_2O_3–st. ZrO_2 were sprayed on the substrate by plasma spraying method. The elevated temperature performance of two variants of modified graphite materials was subsequently analysed using U-10 wt.% Zr alloy. The results indicate that after five thermal cycles, the graphite substrate coated with an 8 mol% Y_2O_3–st. ZrO_2 coating displayed fractures, whereas the graphite substrate treated with Y_2O_3 coating was denser with no fractures. Y_2O_3 coating made the modified graphite composite material have better high-temperature resistance and oxidation resistance. Dutta et al. [118] prepared calcium–aluminate coated graphite and analyzed the effect of the introduction of coated graphite on the oxidation resistance of carbon-containing refractories. The results show that the oxidation resistance of carbon-containing refractories with 5.0% coated graphite significantly improved.

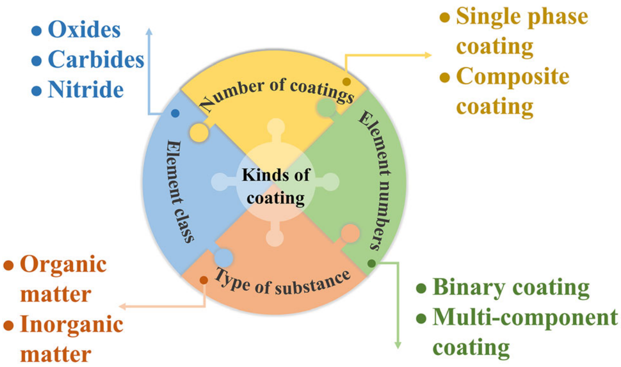

Fig. 7 Coatings according to different classifications [104–110]

Al_2O_3 has been widely used as a coating material due to its remarkable chemical stability and compatibility [119, 120]. Zhang et al. [121] prepared cubic γ-Al_2O_3 coatings via non-electrode plasma electrolysis to modify graphite powder. By performing wettability and anti-oxidation tests on the powder before and after modification, the results show that the wettability of modified graphite

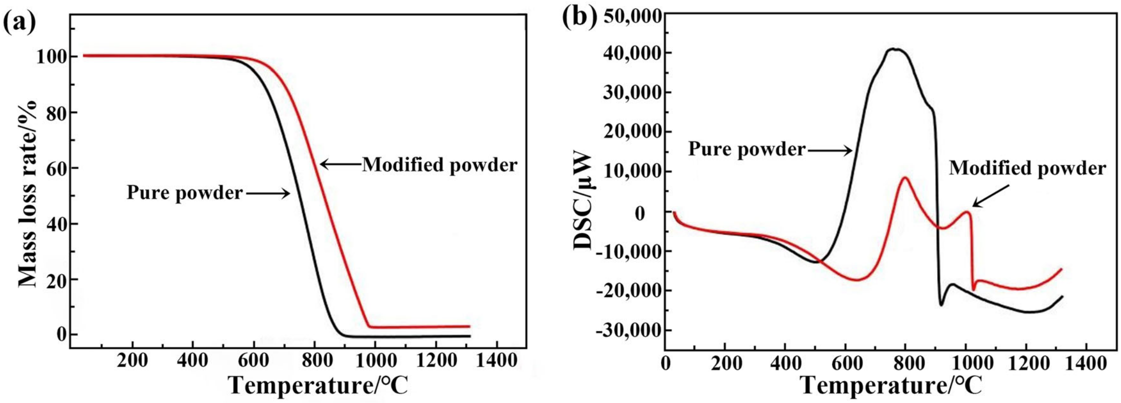

Fig. 8 TG–DSC (thermogravimetric–differential scanning calorimetry) curves of pure powder and modified powder. **a** TG curves; **b** DSC curves [121]

powder was significantly improved by approximately 53% compared to the pre-modified graphite powder. Figure 8 shows the oxidation of the modified graphite in comparison to the original graphite. The modified graphite demonstrated an increase in both the starting oxidation temperature and the complete oxidation temperature, along with a notable decrease in the oxidation rate. As a result, the treated graphite powder exhibited improved resistance to oxidation. Furthermore, in a high-temperature environment, the graphite surface coating demonstrated no evidence of defects such as cracks or spallation. The coating had an excellent protective performance for graphite powder and enhanced the practicability of graphite in carbon-containing refractories. Li et al. [122] used Al_2O_3 cladding of expanded graphite (EG) to react between the O atoms in Al_2O_3 and the C atoms on the surface of EG to produce a good cladding layer. The EG capped by Al_2O_3 showed higher hydrophilicity. In the study to improve the wettability of graphite and its application in carbon-containing refractories, Yilmaz et al. [123] prepared Al_2O_3-coated graphite via the sol–gel method and found that due to its non-wettability, Al_2O_3 coating failed to fully cover graphite. Nevertheless, the coated graphite exhibited excellent oxidation resistance in comparison to pristine graphite, demonstrating its promise for steelmaking.

Oxide coatings show excellent performance in modifying graphite, but their coefficients of thermal expansion are different from those of graphite materials, decreasing the protective ability of the coating during use [124]. Therefore, it is crucial to select coating materials with appropriate coefficients of thermal expansion, develop new composite coatings, and enhance the bonding strength between the coating and substrate materials to widen the range of applications and enhance the performance of oxide coatings.

3.2 Nitride coating

Because of their great hardness, outstanding oxidation resistance, and thermal stability, transition metal nitrides are frequently employed as coating materials [125, 126]. Tao et al. [127] employed plasma nitriding technique to nitride TiO_2 on the surface of graphite, resulting in the formation of a uniform and densely structured TiN coating. The coating had a high hardness and low coefficient of friction, which enhanced the wear resistance of the coated graphite material and significantly improved the corrosion resistance of the coated graphite compared to uncoated graphite. Zhong et al. [128] used sodium borohydride and ammonium formate as starting materials to prepare hexagonal boron nitride nanocrystals (h-BNNC) on the surface of graphite flakes by wet-chemistry coating method. As a result, the coated graphite flakes have no obvious aggregation phenomenon and good dispersion.

Titanium nitride materials have good specific strength and corrosion resistance, making them a promising candidate for a variety of applications [129, 130]. Ranjan et al. [131] used a reactive plasma spraying technique to prepare TiN coating on graphene nanosheets. The coating had high hardness, good wear resistance, and fracture toughness, which ensured that the coated graphene still maintains its structural integrity under high-temperature environments. The significance of its potential impact on carbon-containing refractories warrants further study in the future. Ding et al. [132] synthesized titanium nitride whiskers with various morphologies on the surface of graphite by molten salt method under different reaction conditions. During the oxidation experiments, the untreated graphite had been completely oxidized at 1000 °C, while only a small fraction of TiN on the surface of modified graphite was oxidized to generate TiO_2. The presence of whiskers increases the specific surface area of graphite on the one hand, enhances

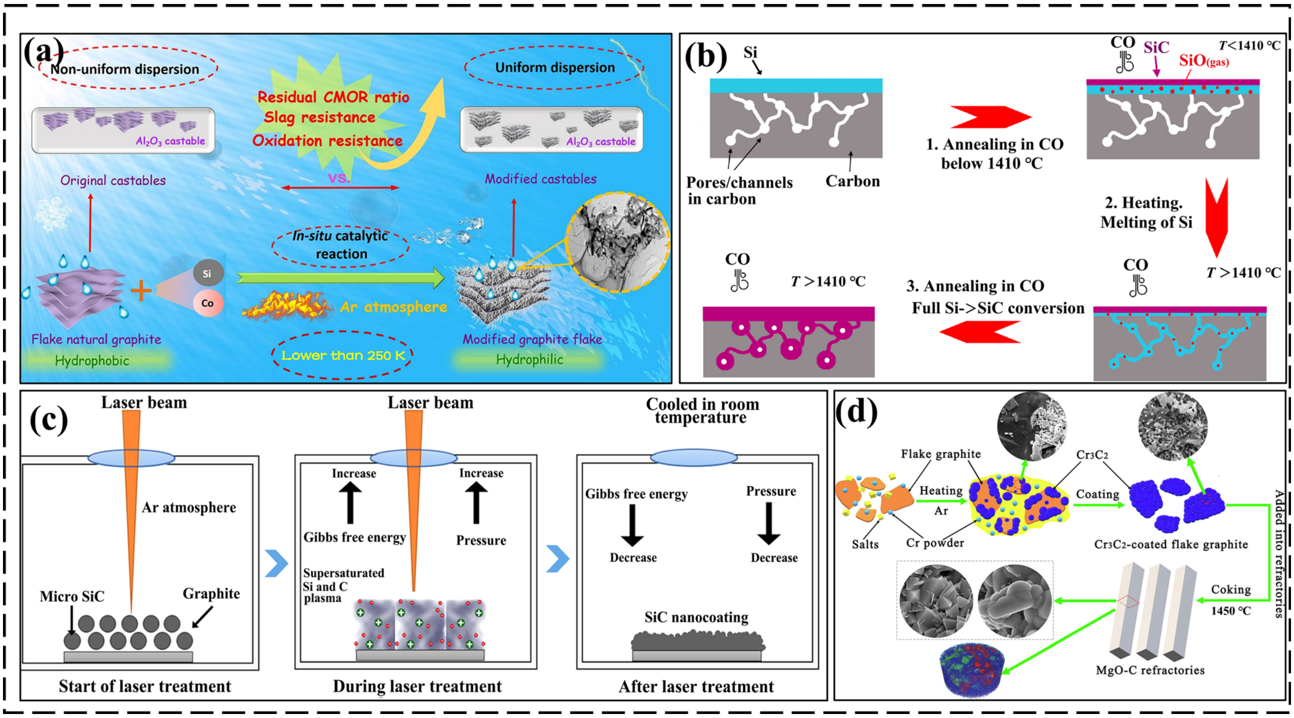

Fig. 9 Mechanism process of synthetic carbide coating. **a** In-situ synthesis process of catalyst; **b** chemical vapor deposition; **c** laser treatment; **d** molten salt method [53, 87, 141, 142]. *T*—Temperature

oxidation resistance of graphite and lowers rate of oxidation on the other hand, and both of them are crucial to improve the oxidation resistance of carbon-containing refractories.

Nitride coatings have excellent thermal stability and corrosion resistance and show potential for improving the oxidation and erosion resistance of carbon-containing refractories. However, it cannot be ignored that nitride coatings have low toughness and brittle fracture. The subsequent studies should concentrate on these inadequacies to elevate the performance of nitride coatings [133, 134].

3.3 Carbide coating

Carbide coatings have good wear resistance, corrosion resistance, excellent high-temperature resistance, and oxidation resistance, which make them widely used in protective coatings for carbon materials [135–140]. With the deepening of research, the preparation methods of carbide coatings are also increasing. Figure 9 shows some of the mechanism processes of preparing carbide coatings [53, 87, 141, 142]. At present, the main carbide coatings used are SiC, TiC, Cr_3C_2 and WC. Figure 10 summarizes the morphology of some different carbide coating coated graphite [142–148]. Li et al. [142] coated flake graphite with Cr_3C_2 and introduced modified graphite into MgO–C refractories to study its effect on material performance. They found that the oxidation resistance of coated flake graphite was significantly improved compared to that of untreated graphite, and the introduction of the modified graphite promoted the densification of MgO–C refractories. By using the molten salt method, our team prepared nanocrystalline ZrC-coated graphite and added it to low-carbon alumina–carbon refractories [149, 150]. The results show that capping graphite with zirconium significantly enhanced its oxidation resistance, and adding modified graphite to low-carbon alumina–carbon refractories improved the mechanical performance, oxidation resistance, and thermal shock resistance of the materials.

SiC has a high melting point, excellent corrosion resistance, and chemical stability. Several researchers have studied SiC coating materials [151–153]. Liu et al. [154] investigated the impact of modified graphite on the performance of low-carbon alumina–carbon refractories by forming SiC whisker coatings on the graphite surface. The results show that SiC whisker-coated graphite demonstrated improved water wettability compared with unmodified graphite. Furthermore, the low-carbon alumina–carbon refractories using the modified graphite exhibited excellent oxidation resistance and slag resistance. Yang et al. [155] prepared HfC–ZrC–SiC composite coatings on the surface of carbon material covered with a SiC transition layer. The resulting bonding between the composite coatings and the carbon material was excellent, and the composite coatings were uniformly compact in texture. By performing ablation experiments on the coating material, it was observed that as the temperature increased to 1927 °C, the mass ablation rate

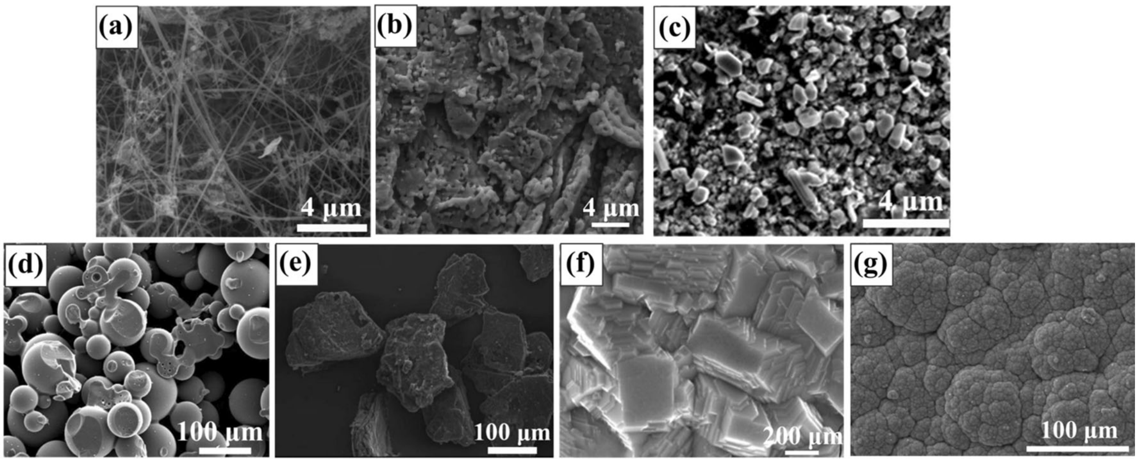

Fig. 10 Scanning electron microscopy images of different carbide coatings. **a** SiC; **b** Cr_3C_2; **c** WC; **d** SiC; **e** TiC–Ti_3AlC composites; **f** TiC; **g** TaC [142–148]

of the material surface gradually tended to 0, meaning that the carbide coating began to oxidize to produce an oxide coating. The reaction stopped at around 2 min, indicating that the generated oxide coating effectively prevents the material from further oxidation. Additionally, the newly formed oxide coating has a dense structure that effectively prevents crack extension, thus significantly boosting the resistance of the carbon material against oxidation and erosion. In the study of carbon-containing refractories bricks for steelmaking furnaces, Ye et al. [156] synthesized SiC-coated carbon black by the molten salt method using carbon black (CB) and silica powder as raw materials. More silanal groups were present on the surface of SiC-coated CB particles compared to uncoated CB particles. In addition, the Si–C bond can interact with water to form carboxylate groups, resulting in improved dispersibility and flowability of SiC-coated CB.

Analysis shows that the carbide coating effectively bonds with the graphite substrate, facilitating the maintenance of excellent performance in high-temperature environments. This reduces the potential for high-temperature oxidation and slag erosion of carbon-containing refractories and helps maintain the quality and performance of clean steel [157]. However, carbides have the disadvantages of higher synthesis temperature and depletion of matrix material during preparation. Using carbide coating should take into account the performance of material, ensuring that the initial structure and performance of material are not destroyed to the maximum extent.

4 Conclusions and prospect

Carbon-containing refractories inevitably come in contact with oxygen and oxides in the environment during their service. Oxygen and other substances enter the interior of the material and react with carbon, resulting in oxidation, erosion and molten steel pollution of carbon-containing refractories. To improve the oxidation resistance and other performance of graphite used in carbon-containing refractories, the main methods of graphite surface modification are reviewed, and their influence on the use of carbon-containing refractories in high-temperature smelting environment and the advantages and disadvantages of different modification methods are summarized. Currently, with the continuous improvement in high-performance steel material requirements, clean steel smelting with carbon-containing refractories performance needs to follow the pace of the times, innovation, and development. Graphite, one of the essential components of carbonaceous refractories for improving their performance, still needs to be further explored, regarding the following aspects:

1. New surface engineering techniques to improve the surface activation energy, water wettability, and bonding ability of graphite without destroying its surface structure and properties.
2. Adjustment of the expansion coefficient to improve the bonding strength between the composites according to the bonding mechanism between the coating and graphite.
3. Development of composite coatings with different structures to combine the advantages of different types of coatings and maximize the performance of the coating and graphite.

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Declarations

Conflict of interest Bei-yue Ma is a youth editorial board member for *Journal of Iron and Steel Research International* and was not involved in the editorial review or the decision to publish this article. The authors state that they have no known competing financial interests or personal relationships.

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