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Innovative technology for improving the yield of large-flake graphite based on enhanced gravity concentration

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Abstract: Improving the yield of large-flake graphite is the research focus in the field of crystalline graphite separation. However, graphite flakes would be seriously damaged by gangues with high hardness during grinding in the conventional multi-stage grinding and multi-stage separation because flake graphite and gangues were always together. Based on this, this paper put forward a new technology and technology for separating gangues and graphite as early as possible by enhanced gravity concentration (EGC), and separating large-flake graphite from fine-flake graphite separately at the same time, which reduced the damage of gangues to graphite flakes during grinding. The experimental results showed that the yield of large flake graphite increased to 4.74% after adopting this new technology. Furthermore, with the help of the atomic force microscopy (AFM) detection technology, the large-flake graphite concentrate obtained by the latest technology was conformed with a lower roughness and more superior performance.

***Keywords*:** liberation degree, large-flake graphite, hydrophobicity, enhance gravity separation, atomic force microscopy

1. Introduction

As we all know, it is a hot and challenging spot to effectively protect the flake structure to improve the large-flake graphite yield for flake graphite beneficiation, which is also the notable difference between flake graphite beneficiation and other minerals beneficiation (Baiju et al., 2025; Ma et al., 2021). There are many hard gangue minerals in graphite ore, such as quartz (Mohs hardness of 7) and feldspar (Mohs hardness of 6-6.5), whose hardness is much higher than that of graphite (Mohs hardness of 1-2) (Cheng et al., 2017; He et al., 2017; Cen et al., 2018; Zhou and Cheng, 2016; Liu et al., 2022). The root reason that graphite flake is easily damaged is that the existence of a large number of hard gangues will not only increase the grinding time and stages, but also cause severe destruction to the flake structure during the grinding process (Liu et al., 2014; Feng, 2015). Therefore, graphite beneficiation generally adopts a special method of multi-stage grinding and multi-stage flotation, that is, the dissociated gangues are separated using flotation in time after grinding, and then the concentrate is fed into the subsequent grinding (Ren, 2021, You et al., 2020).

In recent years, some scholars have continuously optimized the traditional stage grinding-stage flotation process, and successively put forward improved processes such as the stage grinding-stage flotation-pre-cataloging process and stage grinding-concentration process (Lao et al., 2014; Liu et al., 2015). Some scholars have put forward the flotation-shaking table-flotation process, high-pressure roller-stirred grinding mill-nanobubble column flotation process, and particle bed breakage-grading separation process by introducing new equipment or technology (Ma et al., 2021; Niu et al., 2018; Ma and Tao, 2022; Zhang et al., 2024). In addition, some new auxiliary flotation methods (such as ultrasonic-assisted flotation and high-shear exfoliation) have been proposed to protect large-flake graphite (Barma et al., 2019; Ni et al., 2022; Zhou et al., 2023). By optimizing the flotation or grinding process, the

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destruction of graphite flakes by gangue minerals in the grinding process can be weakened. However, it is necessary to introduce more effective measures before flake graphite grinding and flotation, that is, it is a more effective method to prevent large flake graphite from being destroyed by separating large flake graphite from gangue as early as possible to reduce the grinding times with gangue minerals.

It is well known that flake graphite has good natural hydrophobicity, which will spontaneously form agglomeration in water and even partially float on the water surface (Qui, 2016; Vilaverde et al., 2015). In addition, the hydrophobicity of graphite can be enhanced by hydrophobic reagents (Zhu et al., 1999). On the contrary, the main gangue quartz and feldspar have good hydrophilicity (Ding, 2010). Therefore, the differences in hydrophobicity and agglomeration between graphite and gangues will inevitably lead to a tremendous difference in sedimentation velocity in water, and then aggravate the difference of movement trajectory and distribution in the gravity separation process, thus realizing the effective separation of large-flake graphite and gangues (Wang et al., 2022). Based on this, the author innovatively introduces the enhanced gravity separation process before flotation (Qiu et al., 2016; Xia et al., 2022; Ning et al., 2012; Shi et al., 2011; Yang et al., 2009). Different flotation processes were carried out for other products of enhanced gravity separation to realize precise and effective protection of graphite flake structure and significantly improve the yield of large-flake graphite.

This paper describes the novel enhanced gravity separation-precise flotation process, focusing on the product characteristic analysis of enhanced gravity separation. It confirmed that the novel method can effectively separate large-flake graphite from gangues and produce two different products based on the characteristic analysis. Finally, by comparing the large-flake graphite content of the novel and conventional processes, it can be concluded that the novel enhanced gravity separation-precise flotation process could effectively protect +0.15 mm large-flake graphite. This study provides an alternative and practical new idea for solving the problem of safeguarding large flake graphite in the actual production of flake graphite ore, which has important practical significance.

2. Experimental

2.1. Minerals and reagents

The flake graphite ore samples used in the experiments were obtained from a particular area of Heilongjiang Province in Northeast China. The samples were comminuted to less than 1.0 mm by a laboratory-scale high-pressure grinding roller (HPGR) (Shenyang Shengshi Wuhuan Science and Technology Co., Ltd, China). The flake graphite ore contained 14.91% fixed C, and the other main ingredients were 43.34% SiO_2, 8.41% Al_2O_3, 14.34% CaO, 3.19% MgO, and 3.64% TFe, respectively. The mineralogical composition was analyzed using X-ray diffraction (XRD) analysis. As shown in Fig. 1, in addition to the valuable graphite peaks, other intense diffraction peaks are observed, resulting from gangues such as feldspar and quartz.

Fig. 2 visually shows various liberation degrees of graphite in different grain sizes using mineral liberation analysis. Only about 30% of graphite in −1.0+0.30 mm grain size distributes as monomer

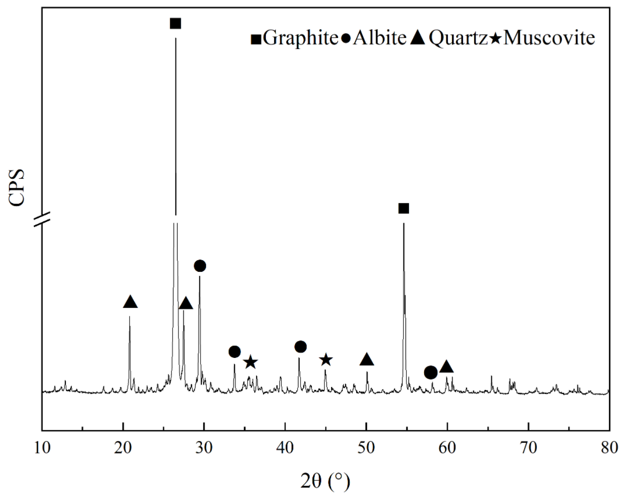

Fig. 1. XRD patterns of the raw ore

particles, that is, most graphite particles connect with gangues. About 60% and 75% of graphite in −0.30+0.18 and −0.18+0.15 mm grain sizes distribute as monomer particles, and some coenobiums of graphite and gangues are still visible. More than 95% of graphite in −0.15+0.074 mm grain size has dissociated, and the liberation degree is reasonably sufficient. Therefore, the liberation degree of graphite increases with the particle size decreasing, which indicates that the graphite ore must be ground to achieve the necessary liberation degree. However, it should be noted that the liberation degrees of −0.18+0.15 mm grain sizes is nearly 75%, indicating that the separation of large-flake graphite can be achieved at a coarser feed size. This has a positive effect on protecting and increasing the yield of large-flake graphite.

Kerosene (Dongying Yuxing Chemical Co., Ltd, LP) and 2# oil (Tieling Flotation Reagents Co., Ltd, LP) were not only used as hydrophobic reagents in enhanced gravity separation, but also as the collector and frother in flotation, respectively. Sodium silicate (Tianjin Zhiyuan Reagent Co., Ltd, AR) was used as the depressor of gangues.

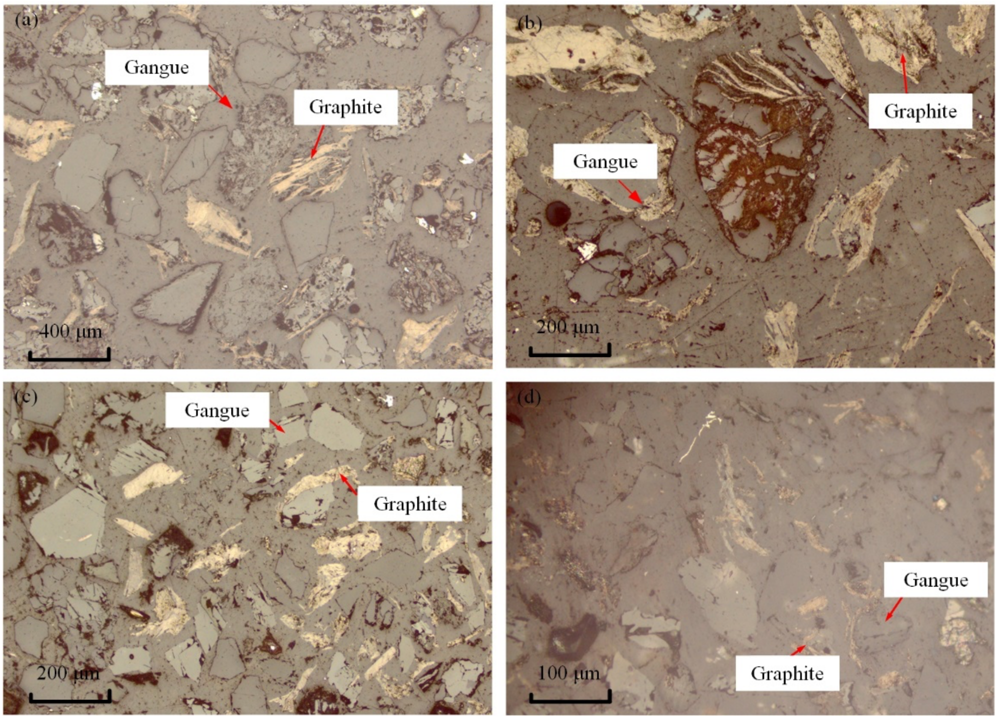

Fig. 2 Liberation degrees of graphite (a: −1.0+0.30 mm; b: −0.30+0.18 mm; c: −0.18+0.15 mm; d: −0.15+0.074 mm)

2.2. Methods

2.2.1. Enhanced gravity concentration-precise flotation experiments

The enhanced gravity separation experiments were conducted using a shaking table (LYN(S) 1100×500, Wuhan Exploration Machinery Co., Ltd). The 1.0 kg graphite ores were stirred with hydrophobic reagents (kerosene and 2# oil) in a 3.0 L XFD_{IV} flotation machine for 3.0 min to enhance the surface hydrophobicity before gravity concentration.

The flotation experiments were performed in the XFD_{IV} flotation machine with volumes of 3.0, 1.5, and 0.5 L. The sodium silicate, kerosene, and 2# oil were added in sequence with 3.0 min stirring.

2.2.2. Contact angle measurement

The natural contact angles and contact angles strengthened by kerosene of graphite, quartz, and feldspar were detected using the XG-CMAB contact angle measuring instrument. A 0.5 g single mineral and 1.0 μL kerosene were added into a stirred tank containing 40.0 mL deionized water stirring for 3.0 min. After filtering and drying, the sample was pressed into a cylindrical, dense, and smooth tablet with a thickness of 2.0 mm and a diameter of 10.0 mm using an electric-meter tablet press. The tablet is put on the glass slide, and the ionized water droplets drip slowly. The photos of the contact moment between the droplet and tablet are obtained by a real-time camera and measured by the five-point method. Each sample is measured three times to take the average value.

2.2.3. SEM and EDS analysis

The morphology of enhanced gravity concentration products was directly scanned by a Gemini 300 scanning electron microscopy (SEM) detector (Zeiss, Germany) equipped with energy-dispersive X-ray spectroscopy (EDS). After scanning at different multiples, the distribution of the chemical elements was detected by EDS analysis.

2.2.4. Atomic force microscopy analysis

Atomic force microscopy (AFM) analysis was operated using a MultiMode 8 microscope (Bruker, Germany) in the Peak Force Tapping mode. Before AFM imaging, the mineral samples were dispersed in a 10.0 mL measuring glass with deionized water for 10 min using an ultrasonic cleaning device. A drop of the upper suspended liquid with mineral particles was dropped onto the mica substrate cleaned with adhesive tape. After drying at a low temperature, the images were captured by SNL-10 probes at a scan size of 1.0 μm. Typically, 5 to 10 individual particles of each sample were randomly chosen for pictures, and the representative picture was selected according to the Nanoscope Analysis v1.8 software.

3. Results and discussions

3.1. Wettability difference between graphite and gangue minerals before and after enhancement with kerosene

According to the above ore properties based on the XRD and mineral liberation analysis results, a number of large-flake graphite are embedded in the ore, which are likely to be seriously damaged if ground together with high-hardness gangues (quartz and feldspar). Therefore, it is necessary to separate graphite from gangues before grinding so as to protect large-flake graphite effectively. Graphite is naturally hydrophobic, while quartz and feldspar are hydrophilic (Shalamov et al., 1981; Wakamatsu and Numata, 1991). The wettability difference between graphite and gangue minerals can be enhanced by changing the interaction between the solid surface and liquid molecules with hydrophobic reagents (Liu, 2021). Considering that kerosene is used as the collector of graphite in the later flotation stage, which has good hydrophobicity, it is selected as the hydrophobic reagent to enlarge the difference in wettability between graphite and gangue minerals. Thus, the large flake graphite can be separated from gangue such as quartz or feldspar as early as possible by gravity concentration.

The wettability of minerals is generally characterized by contact angles, the larger the contact angle, the stronger the hydrophobicity. To prove the influence of kerosene on the wettability of graphite, quartz, and feldspar, the natural contact angles and the contact angles after enhancing with kerosene were measured, respectively, and the results are shown in Fig. 3.

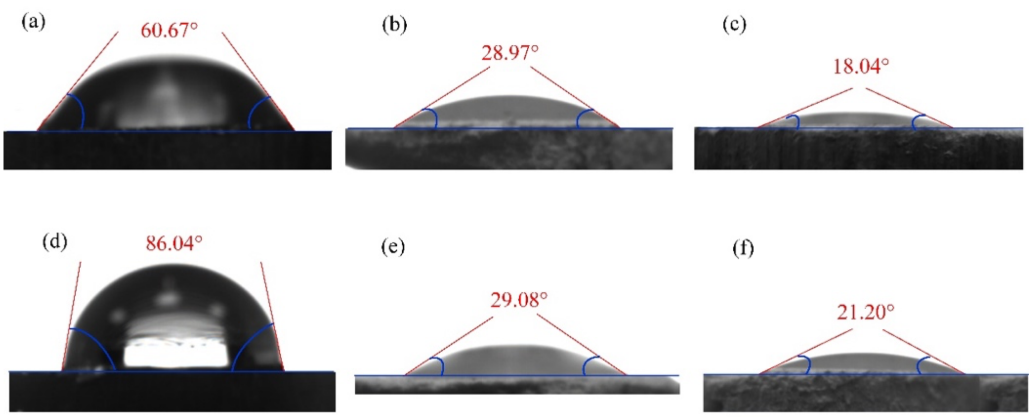

Fig. 3. Contact angles of graphite, quartz, and feldspar before and after enhancing (a-c: natural contact angles; d-f: contact angles after enhancing with kerosene)

It can be seen from Fig. 3 (a)-(c) that the natural contact angle of graphite is 60.67°, which has good natural hydrophobicity. In comparison, the natural contact angles of quartz and feldspar are 18.04° and 28.97°, which are pretty different from the natural contact angle of graphite, indicating that they are more hydrophilic. Therefore, there are noticeable natural wettability difference between graphite and gangue minerals, which provides a favorable basis for gravity concentration of graphite, especially large flake graphite particles and gangue minerals particles by relying on the wettability difference.

As shown in Fig. 3 (d)-(f), the contact angle of graphite increases greatly after kerosene treatment, which shows that kerosene adsorbed on the graphite's surfaces can effectively improve its hydrophobicity. This is mainly because kerosene droplets can closely arrange on the graphite surface and then spread slowly, and finally gradually form an oil film to wrap graphite, thus significantly enhancing the hydrophobicity of graphite (Ren, 2018). However, the contact angles of quartz and feldspar almost have no change after adding kerosene, which shows that kerosene does not change the wettability of quartz and feldspar because they belong to the polar minerals, which makes it difficult to be adsorbed by the nonpolar oils. Therefore, kerosene can remarkably enhance the wettability difference between graphite and gangue minerals. In this case, the separation of large flake graphite from gangue can better realized by gravity concentration after adding kerosene, which was confirmed by the following gravity concentration experiment.

3.2. Comparison of gravity concentration with and without kerosene

Based on the wettability differences between graphite and gangues before and after enhanced by kerosene, direct gravity concentration (DGC) and enhanced gravity concentration (EGC) was conducted, that is, graphite was firstly enhanced hydrophobicity with kerosene and 2# oil before gravity concentration using shaking tables. The comparison of the output position schematic diagram and separation indexes between DGC and EGC is shown in Fig. 4.

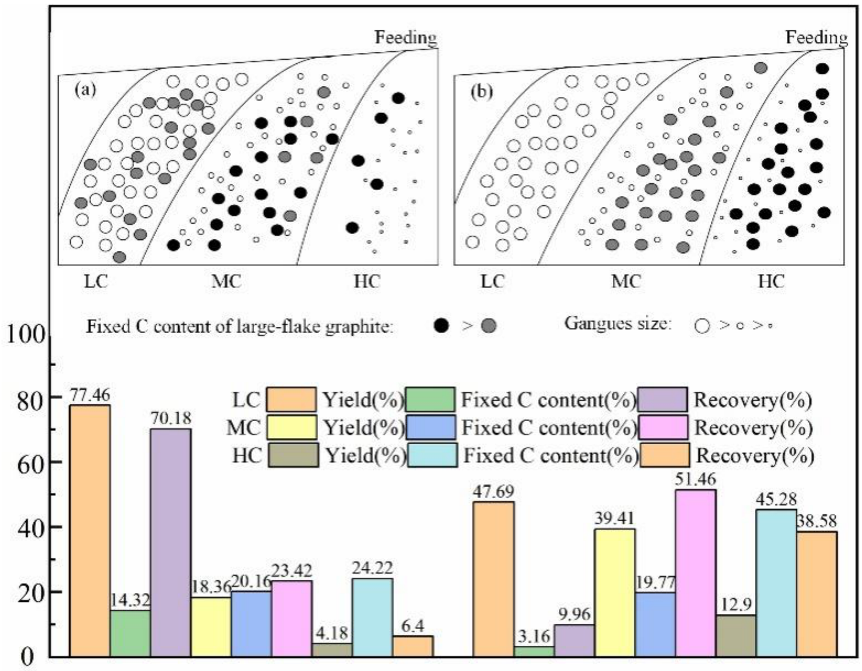

Fig. 4. Contrast of output position schematic diagram and separation indexes (a: DGC; b: EGC)

As shown in Fig. 4, the yield, fixed C content, and recovery of low carbon product (LC) obtained from the EGC were much less than those of the DGC. However, the yield, fixed carbon content, and recovery of medium carbon product (MC) and high carbon product (HC) obtained from the EGC substantially increased compared to the DGC. In addition, Fig. 4 (a) and (b) intuitively depicted that the presence of kerosene and 2# oil essentially enhanced the hydrophobicity of graphite particles resulting in the large-flake graphite skewed to MC even HC. At the same time, the output position of gangues did not changed on the shaking table used for gravity concentration.

Thus, the large-flake graphite was more conducive to being enriched into MC and HC through the EGC, which resulted in effective separation of larger flake graphite from coarse gangues.

3.3. Characteristic analyses of the EGC products

3.3.1. Distribution rate of the +0.15 mm large-flake graphite

The purpose of this novel process is to protect the large-flake graphite (+0.15 mm) from gangue as much as possible, so the distribution rate of +0.15 mm large-flake graphite for the three products (LC, MC, and HC) from EGC was detected, and the results are shown in Table 1.

Table 1. Distribution rate of +0.15 mm large-flake graphite for LC, MC, and HC

Size (mm)	LC			MC			HC		
	Yield (%)	Fixed C (%)	Distribution Rate (%)	Yield (%)	Fixed C (%)	Distribution Rate (%)	Yield (%)	Fixed C (%)	Distribution Rate (%)
+0.15	24.37	5.04	7.74	8.85	34.53	19.24	8.64	53.90	29.36
−0.15	25.57	2.01	3.25	20.84	13.84	18.19	11.73	30.04	22.22
Total	49.94	3.49	10.99	29.69	19.92	37.43	20.37	40.02	51.58

As shown in Table 1, the distribution rate of +0.15 mm large-flake graphite for LC, MC, and HC was 7.74%, 19.24%, and 29.36%, respectively. The total distribution rate of +0.15 mm large-flake graphite in the raw ore is 56.35%, in other words, the theoretical maximum recovery of large-flake graphite was 56.35%. In addition, the distribution rate in +0.15 mm MC and +0.15 mm HC were 34.14% and 52.10% of the total, respectively. The fixed C content of +0.15 mm LC was similar to that of −0.15 mm MC and HC, however, it was much lower than that of +0.15 mm MC (34.53%) and +0.15 mm HC (53.90%), which indicates that the large-flake graphite all selectively enriched into MC and HC. So most large-flake graphite was selectively and successfully separated from gangues by the EGC.

3.3.2. Morphologies and elements distribution

Based on the distribution analyses, the products from EGC were divided into five products, including LC, −0.15 mm MC, −0.15 mm HC, +0.15 mm MC, and +0.15 mm HC. To detect the morphological characteristics of the five products, SEM images are captured and shown in Fig. 5. The elements mapping and EDS spectrum of +0.15 mm MC and HC are further studied and demonstrated in Fig. 6.

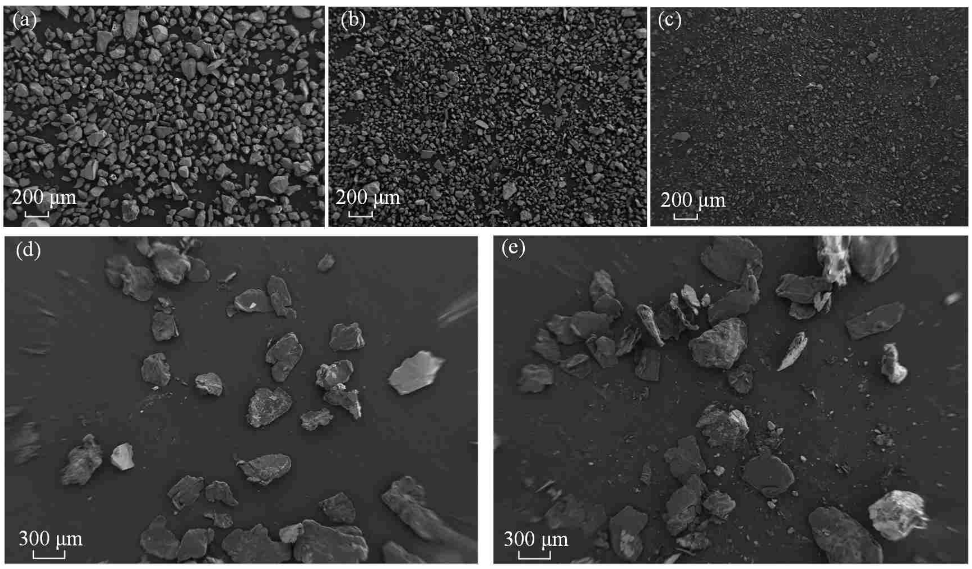

Fig. 5. SEM images of enhanced gravity separation products (a: LC; b: −0.15 mm MC; c: −0.15 mm HC; d: +0.15 mm MC; e: +0.15 mm HC)

It can be seen from the SEM images that LC (in Fig. 5 (a)), −0.15 mm MC (in Fig. 5 (b)), and −0.15 mm HC (in Fig. 5 (c)) were all irregular granules, but the granularity was decreasing at the same 40X magnification. The three products were main gangues hardly containing the large-flake graphite. On the contrary, +0.15 mm MC and HC were both flakes, showing that the two products were dominated by the large-flake graphite.

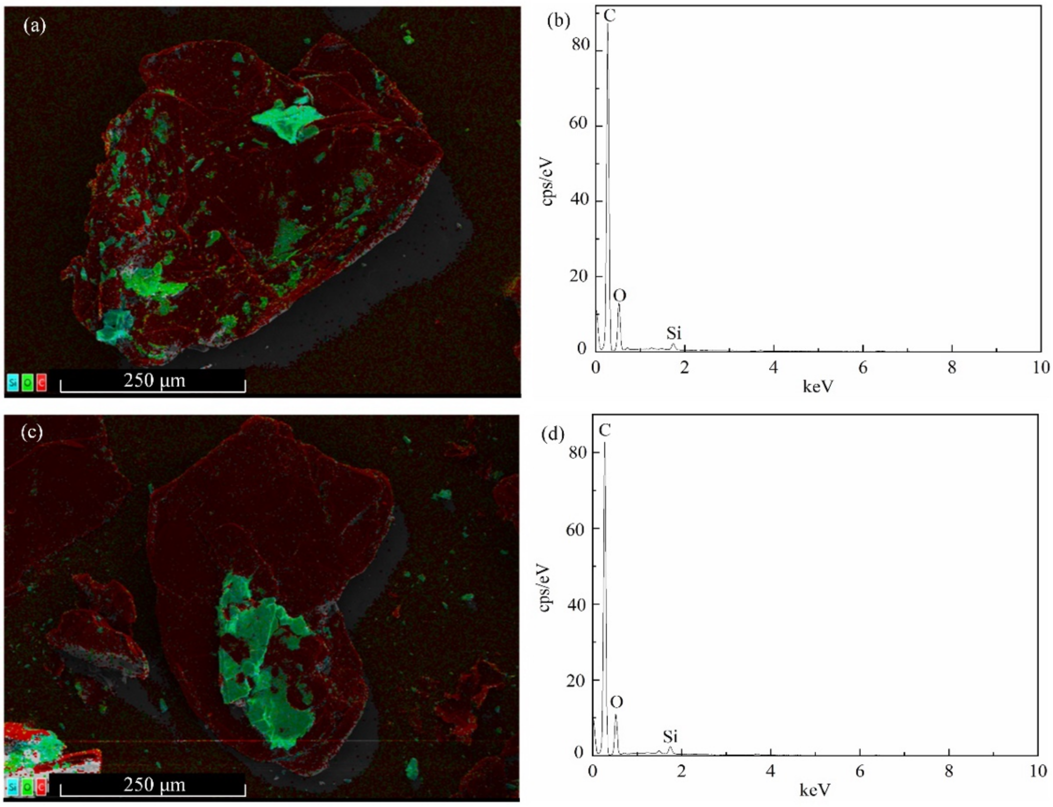

Fig. 6. SEM elements mapping and EDS spectrum of +0.15mm MC and +0.15mm HC (a, b: +0.15 mm MC; c, d: +0.15 mm HC)

The elements mapping images in Fig. 6 (a) and (c) showed that the two products both contained three elements including C, O, and Si, illustrating that the large-flake was graphite and the gangues on the graphite surface were silicate minerals such as quartz. In addition, the EDS spectrum drawn according to elements mapping further indicated that the main content of the two products was both C, which was further confirmed to be graphite.

In a word, LC, −0.15 mm MC, and −0.15 mm HC were combined into a low-quality product (LQ) dominated by gangues. On the contrary, +0.15 mm MC and +0.15 mm HC both mainly contained the large-flake graphite according to the similar morphology and chemical elements distribution, so the two products could be combined into a high-quality product (HQ) dominated by the large-flake graphite. The two products with different characteristics as feeders for subsequent flotation should be adopted different flotation and grinding processes to precisely protect the large-flake graphite from being destroyed, which was the essence of separating large flakes graphite as soon as possible by the EGC.

3.4. Results of the enhanced gravity concentration-precise flotation closed-circuit process

According to Table 1, 86.25% of large-flake graphite was enriched into HQ (from 14.91% to 44.10%) with a yield of 17.49% by the EGC. Thus, it was equivalent to discard a large amount of tailings, which would inevitably shorten the following flotation and grinding stages of HQ and be extremely favorable for the protection of the large-flake graphite.

After EGC, HQ was determined to adopt three stages grinding-three stages screening-three stages flotation process with the final fineness of 10% passing 0.15 mm, while LQ was determined to adopt three stages grinding-seven stages flotation process with the final fineness of 95% passing 0.075 mm.

The results of concentrates and tailings are shown in Table 2, and the quantity-quality of the novel enhanced gravity concentration-precise flotation closed-circuit flowsheet is presented in Fig. 7.

Table 2 showed that the concentrate 1 separated from LQ and the concentrate 2 separated from HQ were both certified graphite concentrate products (over 95% of fixed C). At the same time, the total recovery of Concentrate 1 and Concentrate 2 can be calculated to be 95.83%, indicating that the graphite concentrate has been effectively recovered. In contract, the +0.15 mm large-flake graphite proportion of the concentrate 2 was 100.00% far higher than that of concentrate 1 (1.85%). Compared to the total recovery of +0.15 mm large-flake graphite of 56.35%, the recovery of +0.15 mm large-flake graphite in the two concentrates was 30.73%, which manifested that more than half of +0.15 mm large-flake graphite flake had been effectively protected after the novel technology. In addition, the novel technology could effectively recover graphite with a fixed C grade and recovery of tailings of 0.72% and 4.17% in tailings.

Table 2. Results of the closed-circuit flowsheet

Products	Yield (%)	Fixed C (%)	Recovery (%)	+0.15 mm large-flake graphite		
				Proportion(%)	Yield(%)	Recovery(%)
Concentrate 1	10.26	95.14	66.33	1.85	0.19	1.23
Concentrate 2	4.55	95.37	29.50	100.00	4.55	29.50
Tailings	85.19	0.72	4.17	-	-	-
Raw ore	100.00	14.71	100.00	-	-	-

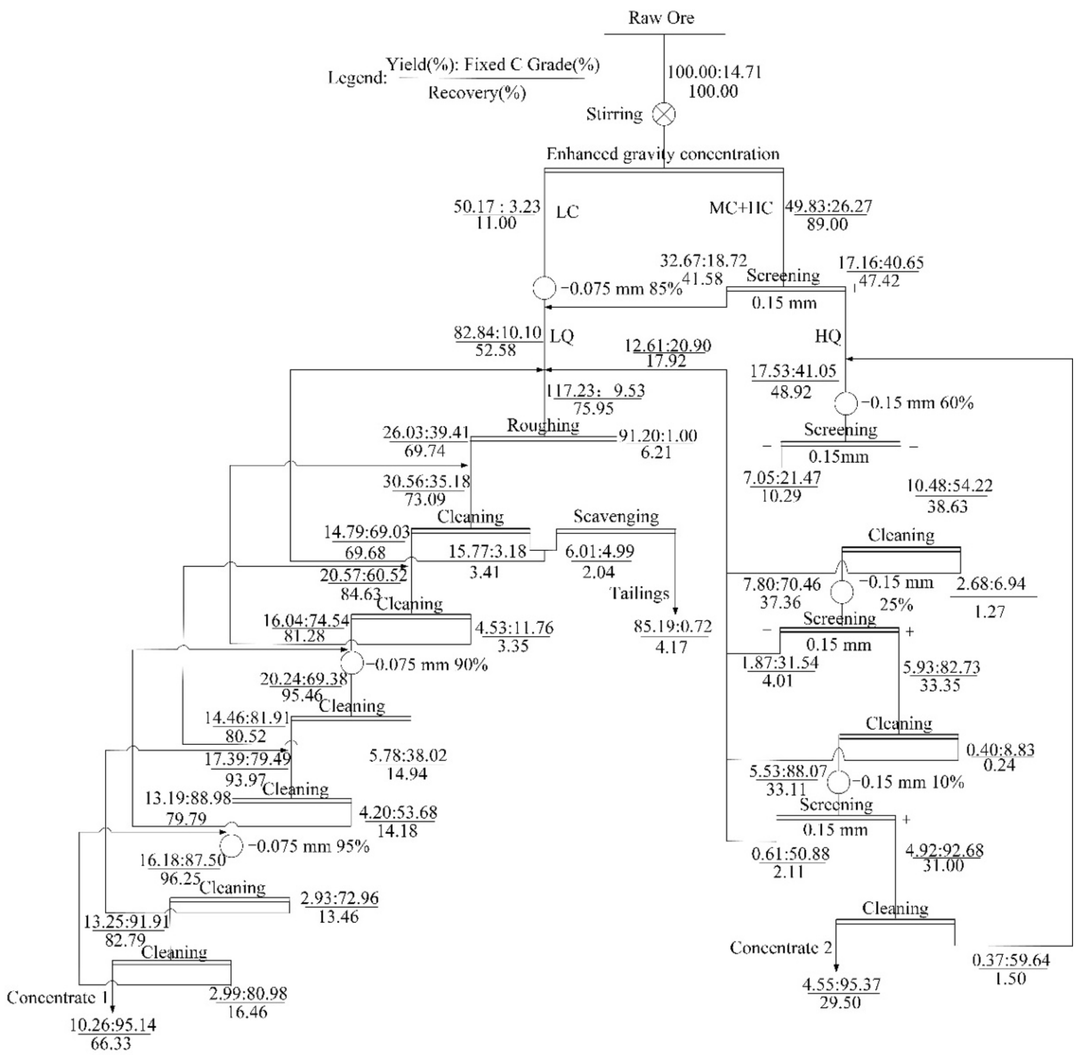

Fig. 7. Quantity-quality of the closed-circuit flowsheet

As shown in Fig. 7, concentrate 2 was obtained from HQ after three stages grinding, screening, and flotation process, which mainly focuses on ensuring the effective protection of large-flake graphite. In contract, concentrate 2 was obtained from LQ and −0.15 mm product form HQ after screening, which mainly focuses on maximizing the recovery of graphite. Thus, the novel enhanced gravity concentration-precise flotation not only effectively protect the large-flake graphite, but also improved the recovery of the graphite.

3.5. Comparison between the innovative and conventional process technology

In order to protect the large flake graphite from being destroyed in the separation of crystalline graphite ores, the process flowsheet of multi-stage grinding and multi-stage flotation is usually adopted. Therefore, in order to prove that the innovative technology proposed in this paper can effectively protect the large flake graphite than that of the conventional process, that is, it can improve the yield of large flake graphite mineral. After experimental study, the conventional separation process with three-stage grinding and eight-stage flotation was at least determined as a suitable process, which obtained the graphite concentrate with of fixed carbon grade and 87.84% of recovery and tailings with 2.06% of fixed carbon grade and 12.16% of recovery. Compared with the experimental results of innovative technology shown in Fig.7 and Table 2, the results amply proved that the novel enhanced gravity concentration-precise flotation process dramatically improves the recovery from 87.84% to 95.83%. More importantly, the yield of large-flake graphite in two concentrates was different, the comparison of concentrates between the two processes is shown in Fig. 8.

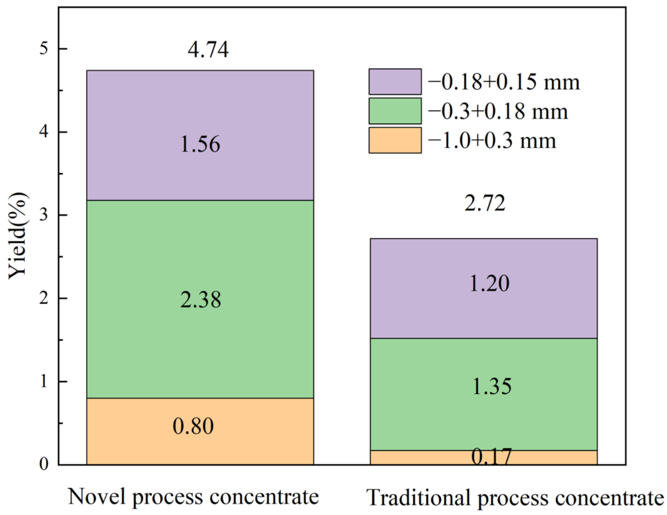

Fig. 8. Contrast between the novel process and the conventional process concentrates

As shown in Fig. 8, the +0.15 mm large-flake graphite yield from the novel process concentrate with 4.74% was much higher than that of the conventional process concentrate with 2.72%. To be specific, the −1.0+0.3 mm graphite yield of the former (0.80%) was more than four times of the latter (0.17%). In addition, the −0.3+0.18 mm graphite yield of the former (2.38%) was also much higher than that of the latter (1.35%). The results persuasively indicated that the novel enhanced gravity concentration-precise flotation process could effectively reduce the large-flake graphite from being destroyed, especially for −1.0+0.3 mm fraction, and improve the recovery of the graphite mineral.

SEM imaging analysis was conducted to investigate the differences in the surface morphology of large-flake graphite between the two closed-circuit process concentrates obtained from the enhanced gravity concentration-precise flotation process and conventional multi-stage grinding and flotation process. The SEM images with the same magnification of the two concentrates are shown in Fig. 9.

As can be clearly seen from Fig. 9, compared with the concentrate obtained from the conventional process, the concentrate from the novel process has many large flakes with the largest size of 310.4 mm×204.3 mm. SEM images visually confirm the results of the study shown in Fig. 8. In addition, the flakes in the concentrate of the novel process have smooth edges and surfaces. In contrast, the flakes in the concentrate of the conventional process have uneven fractured edges. The results indicate that the novel enhanced gravity concentration-precise flotation process significantly improved the concentrate's size and quality.

To characterize nanoscale morphology differences of the large-flake graphite in the two concentrates from the innovative and conventional process technology, the surfaces were imaged in the Peak Force Tapping mode of AFM, and the results are shown in Fig. 10.

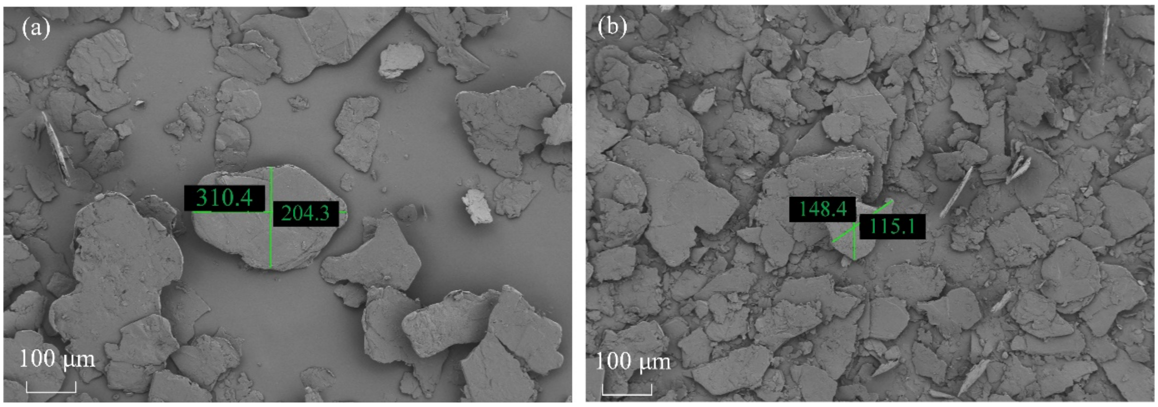

Fig. 9. SEM images of the closed-circuit process concentrates of the novel process (a) and the conventional process (b)

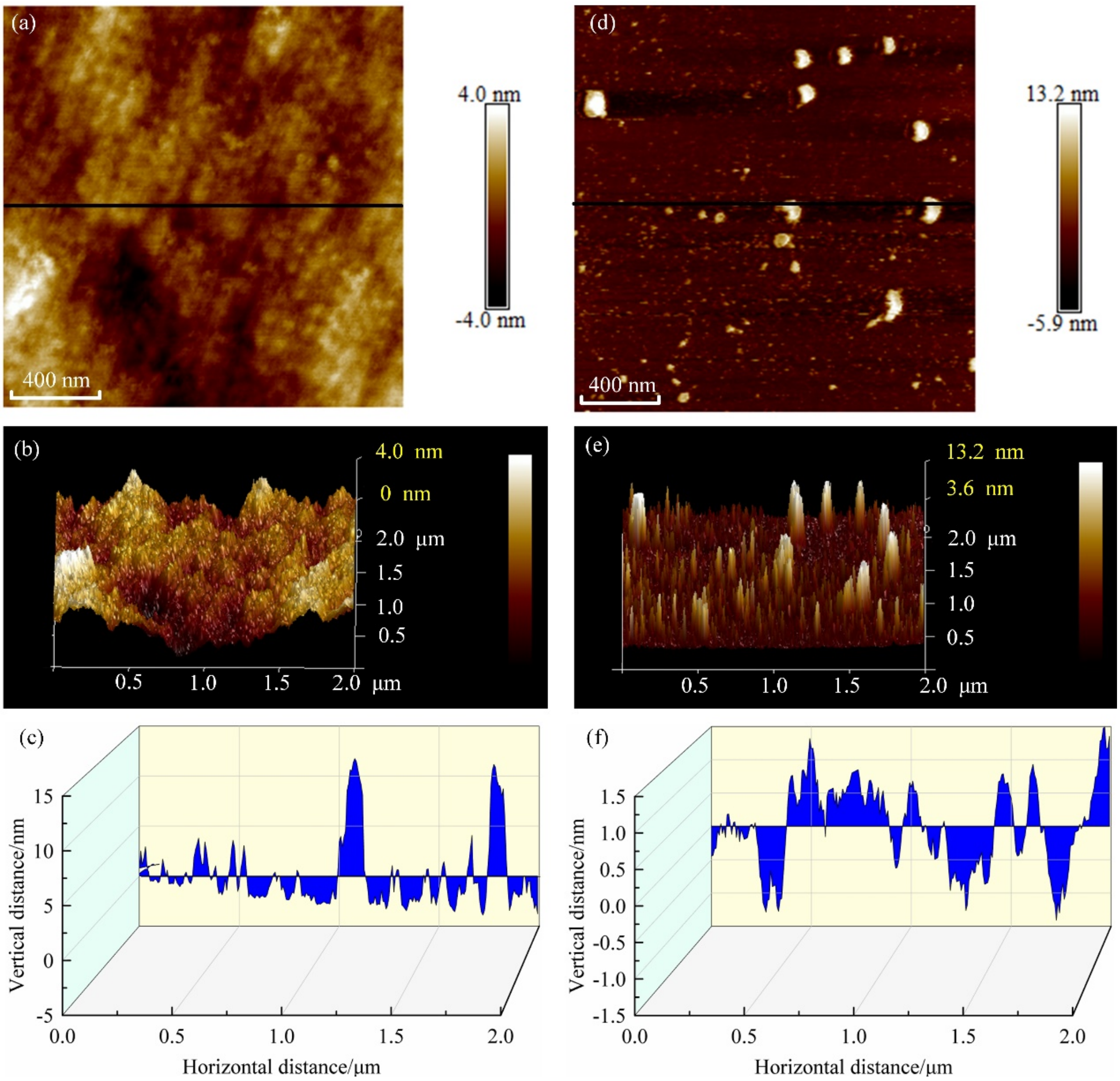

Fig. 10. Height images in 2D, 3D, and cross-section drawing of the large-flake graphite in the two concentrates ((a)-(c): the novel process concentrate; (d)-(f): the conventional process concentrate)

Fig. 10 (a) and (b) are height images of the novel process concentrate in 2D and 3D, respectively, which show that the surfaces of the large-flake graphite particles were smooth. In contrast, the height images of the conventional process concentrate in 2D and 3D (Fig. 10 (d) and (e)) confirm that the surfaces of the particles were rough with some protrusions. The cross-section drawing in Fig. 10 (c) clearly shows the vertical fluctuations along the solid line in Fig. 10 (a). These fluctuations span the range between −1.5 and 1.5 nm, which implies that the height difference on the novel process concentrate surface is between 0 and 3 nm. As for the conventional process concentrate, Fig. 10 (f) shows pronounced vertical fluctuations of the solid line in Fig. 10 (d) in comparison with those of the former. These fluctuations span the range between −5 to 15 nm, which implies that the height difference on the conventional process concentrate surface is between 0 and 20 nm. In short, the large-flake surface of the novel process graphite concentrate is smoother than that of the conventional process graphite concentrate, it is because, since the coarse grained gangue such as quartz and feldspar can be separated as early as possible by the adoption of the novel EGC technology, thus reducing the scratching damage of these gangue on flake graphite, especially the large flake graphite during subsequent grinding which adopt the same grinding stages of three in the innovative and conventional process technology.

In a word, the novel enhanced gravity concentration-precise flotation process not only significantly improved the yield of the large-flake graphite, which could effectively protect the crystal structure of the large-flake graphite, but also improved the recovery of flake graphite. Consequently, the innovative technology shows a promising application prospect for protecting the large flake graphite from being destroyed and is worthy of further study.

4. Conclusions

The hard gangue minerals (quartz and feldspar) would seriously damage the large-flake graphite during grinding, so it is necessary to separate gangues from graphite at a coarser feed size by enhanced gravity concentration. Based on this, the graphite with enhanced hydrophobicity by kerosene was more conducive to enrich into HQ dominated by the large-flake graphite and effectively separated from LQ dominated by gangues.

The novel enhanced gravity concentration-precise flotation process significantly improved the large-flake graphite yield of closed-circuit concentrate from 2.72% to 4.74% compared to the concentrate of the conventional method, especially for the −1.0+0.3 mm fraction. The novel method can effectively protect the crystal structure of the large-flake graphite, resulting in its surface much smoother and significantly improve its yield.

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References

BAIJU, K.R., SATISH-KUMAR, M., KAGI, H. et al., 2005. *Mineralogical characterization of graphite deposits from Thodupuzha–Kanjirappally Belt, Madurai Granulite Block, Southern India.* Gondwana Research; 8, 223–230.

BARMA, S.D., BASKEY, P.K., RAO, D.S. et al., 2019. *Ultrasonic-assisted flotation for enhancing the recovery of flaky graphite from low-grade graphite ore.* Ultrasonics Sonochemistry; 56, 386–396.

CEN, D., ZHANG, T., YU, Y. et al., 2018. *Experimental research on extraction and purification of large flake graphite from graphite ore in Heilongjiang.* Metal Mine; 504, 89–93.

CHENG, F., ZHANG, T., YU, Y. et al., 2017. *Experimental research on beneficiation of large flake graphite ore in Madagascar.* Non-Metallic Mines; 40, 76–78.

DING, Y., 2010. *Investigation on the preparation of high purity quartz from low-grade silica.* Shenyang: Northeastern University.

FENG, Y., 2015. *Study on regrinding technology and flotation rate of flake graphite.* Wuhan: Wuhan University of Technology.

HE, F., ZHANG, L., QIU, Y. et al., 2017. *Experimental research on beneficiation of large flake graphite in eastern area of Mozambique.* Bulletin of the Chinese Ceramic Society; 36, 4205–4210, 4216.

LAO, D., SHEN, S., LI, C. et al., 2014. *Study on technological process of stage grinding–flotation and mesh separation beforehand for flake graphite ore.* China Non-metallic Minerals Industry; 113, 32–35, 47.

LIU, G., 2021. *Preparation and properties of inorganic silicate super-hydrophilic coating.* Guangzhou: Guangzhou University.

LIU, L., GUO, L., SUN, H. et al., 2022. *Comparative study on flotation technology of crystalline graphite from Akesai in Gansu Province.* Non-Metallic Mines; 43, 56–59, 63.

LIU, H., LAO, D., LI, C. et al., 2015. *Flotation technical study of Luobei flake graphite in Heilongjiang Province.* China Mining Magazine; 24, 182–185.

LIU, X., ZHANG, L., LI, X., 2014. *Beneficiation experiment of a graphite in Luobei of Heilongjiang Province.* Metal Mine; 5, 105–109.

MA, Y., HUANG, Y., LIU, L., 2021. *Genesis of the Tianping flake graphite deposit at the western margin of Yangtze Block, SW China.* Ore Geology Reviews; 139, 104434.

MA, F., TAO, D., 2022. *A study of mechanisms of nanobubble-enhanced flotation of graphite.* Nanomaterials; 12, 3361.

MA, F., TAO, D., TAO, Y. et al., 2021. *An innovative flake graphite upgrading process based on HPGR, stirred grinding mill, and nanobubble column flotation.* International Journal of Mining Science and Technology; DOI: 10.1016/j.ijmst.2021.06.005.

NI, C., ZHANG, Q., JIN, M. et al., 2022. *Effect of high-speed shear flocculation on the flotation kinetics of ultrafine microcrystalline graphite.* Powder Technology; 396, 345–353.

NING, G., LI, P., ZHANG, T. et al., 2012. *Hydrophobic nano-silica for the surface modification of graphite flake.* Propellants, Explosives, Pyrotechnics; 38, 520–524.

NIU, M., LIU, L., CHEN, L. et al., 2018. *Study on the protection of large-scale graphite by particle bed breakage–grading separation technology.* Conservation and Utilization of Mineral Resources; 8.

QIU, Y., 2016. *Research of flotation behaviors and new technology on flaky graphite.* Wuhan: Wuhan University of Technology.

QIU, Y., YU, Y., ZHANG, L. et al., 2016. *Dispersion and agglomeration mechanism of flaky graphite particles in aqueous solution.* Journal of Dispersion Science and Technology; 38, 796–800.

REN, P., 2018. *Mineral processing technology of graphite and mica in graphite ore from Sichuan Nanjiang.* Mianyang: Southwest University of Science and Technology.

REN, W., 2021. *Study on flotation behavior of graphite with different characteristics.* Baotou: Inner Mongolia University of Science & Technology.

SHALAMOV, V., VARLAM, A., TIKHONOVICH, G., 1981. *Graphite.* New York.

SHI, G., WANG, Z., ZHAO, J. et al., 2011. *Adsorption of sodium ions and hydrated sodium ions on a hydrophobic graphite surface via cation–π interactions.* Chinese Physics B; 20, 068101.

VILAVERDE, C., SANTOS, R.M., PAIVA, M.C. et al., 2015. *Dispersion and re-agglomeration of graphite nanoplates in polypropylene melts under controlled flow conditions.* Composites Part A: Applied Science and Manufacturing; 78, 143–151.

WAKAMATSU, T., NUMATA, Y., 1991. *Flotation of graphite.* Minerals Engineering; 4, 975–982.

WANG, D., WANG, G., MIAO, X. et al., 2022. *Activated carbon fibers with different hydrophilicity/hydrophobicity modified by pDA–SiO_2 coating for gravity oil–water separation.* Separation and Purification Technology; 303, 122179.

XIA, Y., XING, Y., GUI, X. et al., 2022. *Probing the interactions between collector molecules and hydrophobic graphite surfaces using chemical force microscopy.* Applied Surface Science; 597, 153760.

YANG, D.-S., ZEWEIL, A.H., 2009. *Ordered water structure at hydrophobic graphite interfaces observed by 4D ultrafast electron crystallography.* Proceedings of the National Academy of Sciences; 106, 4122–4126.

YOU, D., HE, A., LI, G., 2020. *Research progress of graphite flotation technology.* Modern Mining; 616, 130–134, 142.

ZHANG, F., LI, H., ZHANG, Y. et al., 2024. *Experimental study on crystalline graphite flotation column.* Nonferrous Metals (Mineral Processing Section); 11, 80–86, 104.

ZHOU, N., CHENG, X., 2016. *Research on the flotation process for large flake graphite ore in Inner Mongolia Autonomous Region.* Contemporary Chemical Industry; 45, 182–184, 188.

ZHOU, S., TONG, Z., DONG, L. et al., 2023. *A comparative study on the influence of single and combined ultrasounds assisted flake graphite flotation.* Ultrasonics Sonochemistry; 99, 106551.

ZHU, B., GUAN, H., LIU, W., 1999. *Study on improving the wettability of water to flake graphite by surfactant.* Journal of Wuhan Yejin University of Science and Technology (Natural Science Edition); 22, 242–244.