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Review of tungsten resource reserves, tungsten concentrate production and tungsten beneficiation technology in China

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Abstract: Tungsten is a strategic metal that is widely used in various fields such as electron communication, aeronautics and astronautics, materials and chemical engineering, due to its special properties. China has the largest reserves of tungsten resources and the largest output of tungsten concentrate in the world, and plays a decisive role in the world tungsten market. In this work, firstly, the reserves and distribution characteristics of tungsten resources in China were summarized, and the production situation of tungsten concentrate in China was reviewed. Based on the gross domestic product per capita (GDP/capita), five different annual GDP growth rates were used to estimate China's tungsten concentrate production in the future. The estimation results suggested that if China's economy continues to grow at the current rate, the accumulative production of tungsten concentrate will exceed current reserves by approximately 2032. Then, from the aspects of process flow, beneficiation equipment and flotation reagents, the beneficiation technology development of different types of tungsten ore in China was also reviewed, including wolframite, scheelite and tungsten-bearing slime. Finally, taking several representative tungsten mines in China as examples, the classic beneficiation technology of different types of tungsten ores was elucidated in detail. Meanwhile, the development direction of tungsten beneficiation technology in the future was put forward, which was of great significance to maintain the superiority of tungsten resources in China.
Key words: tungsten; reserves; production; beneficiation; flotation; gravity separation

1 Introduction

Tungsten (W) has the highest melting point of all metals and one of the highest densities and thus has a wide application in daily life as well as in industrial and military fields. Tungsten carbide is the most important tungsten product and is used to make cemented carbides. Tungsten is also used to make alloys, corrosion-resistant coatings, catalysts, semiconductors, fire-resistant compounds, and many other items. In daily life, tungsten is used in electrical and electronic products, lighting, etc. Therefore, tungsten resources are indispensable in economic and social development [1,2].

For the sustainable mining and consumption of tungsten resources, China has established a series of legislations and regulations, such as "the Notice of the State Council on the Classification of Tungsten, Tin, Antimony, and Ion-type Rare Earth Minerals as Specific Minerals for Protective Mining [3], Notice of the Ministry of Natural Resources on Further Standardizing the Approval and Management of Rare Earth and Tungsten Ore Mining Rights [4], and Criteria for Tungsten Industry [5]".

The global tungsten reserves (W) in 2020 came to 3.4 Mt [6]. The most important tungsten minerals are scheelite ($CaWO_4$) and wolframite

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((Fe,Mn)WO_4)). In the industry, the main mineral processing methods for wolframite consist of gravity separation and flotation [7−9]. Owing to the decline of wolframite resources, scheelite, especially its recovery via flotation, is attracting increasing attention [10,11]. Although the recovery of scheelite has been successful, there are still some problems in scheelite flotation. For example, scheelite is difficult to separate from other calcium-containing minerals, such as calcite ($CaCO_3$), flourite (CaF_2), and apatite ($Ca_5(PO_4)_3(OH,Cl,F)$), and the low-temperature resistance of scheelite collectors needs to be optimized [12−15].

Many studies have focused on the supply and demand of tin [16−18], nickel [19], lead [20,21], cobalt [22], lithium [23], and rare earth elements [24] in China. China is a country with the largest tungsten reserves and is also the largest producer of tungsten concentrates. However, few studies have focused on tungsten production. Simultaneously, metal consumption is closely related to the gross domestic product per capita (GDP/capita) and thus has been widely used to predict the supply and demand of many other mineral resources [25−30]. A study by ZHENG et al [31] showed that an increase of 1% in GDP is associated with a 1.9% increase in a country's metal consumption in the same year, but the use of metals tends to reach a plateau when GDP/capita reaches 15000 US$.

In this paper, past, present, and future of tungsten resource-recoveries in China were investigated. Firstly, tungsten resources and tungsten concentrate production in China were introduced, and the future output of tungsten concentrate was evaluated by linear regression analysis based on GDP/capita. Secondly, the beneficiation technology of wolframite, scheelite, and tungsten-bearing slime was reviewed, including process flow, beneficiation equipment and flotation reagents. Thirdly, the typical beneficiation flowsheets and production practices of several tungsten plants in China were also summarized. It is of great significance to explore the development direction of tungsten resource utilization in the future.

2 Tungsten production in China

Tungsten is listed as a strategic metal in China and is also one of China's dominant mineral resources in the world because China accounts for 55% of the world's tungsten reserves; moreover, China accounted for ~82% of the total world tungsten production in 2020, followed by Vietnam (~5%), Russia (~2.5%), and United Mongolia (~2%) (Fig. 1). China's tungsten reserves, production and export volume all rank first in the world, so China plays an important role in the international tungsten market.

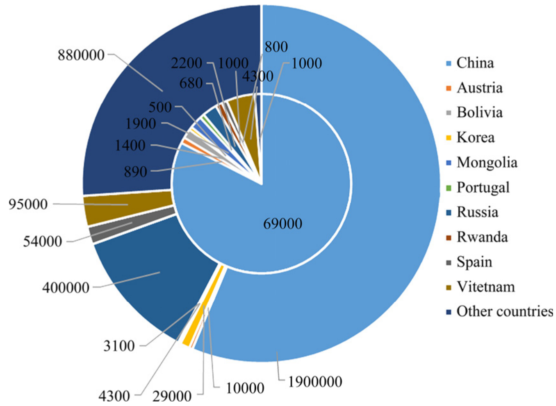

Fig. 1 Global tungsten reserves (outside) and mine production (inside) in 2020 [32] (unit: t)

2.1 Tungsten reserves and resources

According to the official data, by the end of 2018, China's potential tungsten resources were 29.70 Mt (WO_3) [33], and the detection rate of tungsten resources was 25.70%. China's remaining tungsten reserves in recent years are shown in Fig. 2. With ongoing exploration work, China's proven tungsten reserves (WO_3) reached 11.204 Mt in 2019.

Tungsten deposits in China are mainly distributed in the Nanling metallogenic belt, southeastern coastal metallogenic belt and Qinling

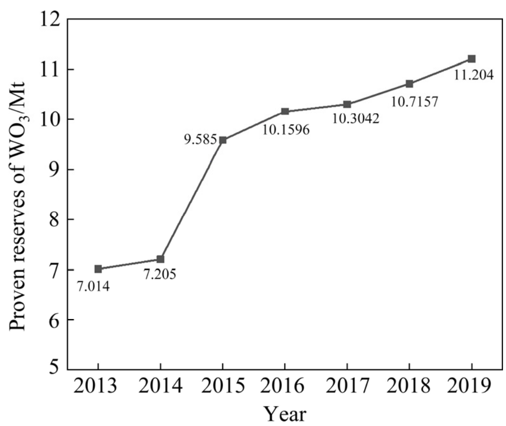

Fig. 2 China's remaining tungsten reserves [33]

metallogenic belt. Therefore, Hunan, Jiangxi and Henan are rich in tungsten resources. The remaining tungsten reserves in the three provinces account for 66.7% of China's reserves, and the potential tungsten reserves account for 49.7% of China's reserves [34−36]. Based on the generic types of tungsten deposits, coupled with orebody structure, ore composition, and surrounding rock properties, tungsten deposits in China can be roughly divided into the following four categories: quartz vein wolframite deposits, skarn scheelite deposits, porphyry tungsten deposits, and strata-bound tungsten deposits [36−38].

2.2 Tungsten concentrate production

In China, most of the tungsten ore is mined underground, and the fluctuations in tungsten grade in underground mining, beneficiation, and tailings are shown in Fig. 3(a). For the period from 1990 to 2004, the tungsten grades in both underground mining and beneficiation increased significantly; for example, the grades in underground mining and beneficiation were 0.20% and 0.26% in 1990, respectively, but 0.44% and 0.42% in 2004, respectively (Fig. 3(a)). Thereafter, tungsten grades in mining and beneficiation decreased and remained at approximately 0.3%. Before 2007, the recovery of tungsten in beneficiation was approximately 80.5%, but thereafter, it declined to approximately 73.7% (Fig. 3(b)). This is related to the large-scale recycling of scheelite after 2007. Since the beneficiation technology of scheelite is not as mature as that of wolframite and the occurrence state of scheelite in nature is more complicated, the recovery and utilization of scheelite are more difficult.

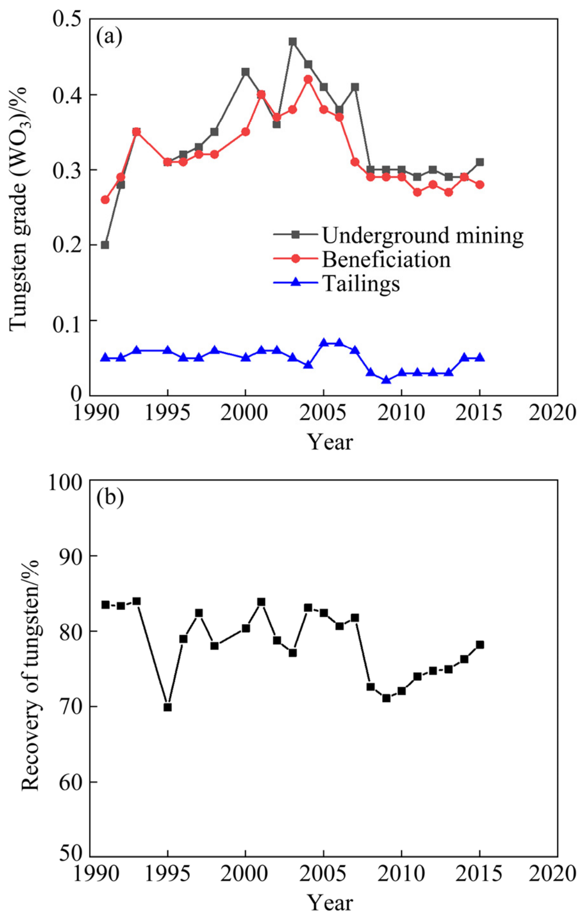

Fig. 3 Fluctuations in tungsten grade (a) and recovery of tungsten from beneficiation (b) [39]

To ensure the advantage of tungsten resources, the minimum value of mining recovery and beneficiation recovery and the comprehensive utilization ratio of tungsten mineral resources in China are required according to the current technology level of tungsten mining (Tables 1 and 2 [40]).

Table 1 Indicators for tungsten mining

Mining mode	Grade of mining (WO_3)/%	Mining recovery/%
Open-pit mining	−	92
Underground mining	≤0.2	80
	0.2 − 0.4	85
	>0.4	90

Data from Ministry of Natural Resource of China [40]

The output of tungsten in concentrate in the period from 1960 to 2018 is shown in Fig. 4(a). Production has significantly increased since 2000, reaching 92.03 kt in 2016. The end-use of tungsten was distributed as follows: cemented carbides (46.4%), specialty steels (25.6%), tungsten materials (22.4%), and chemicals (5.7%) (Fig. 4(b)) [41]. Nearly half of the tungsten was used in cemented carbide parts for cutting and wear-resistant applications, primarily in the construction, metalworking, mining, and oil and gas drilling industries. Thus, tungsten is indispensable in economic and social development.

2.3 Expected future tungsten concentrate production

The future output of tungsten in concentrate was evaluated by linear regression analysis based on GDP/capita, which in turn was calculated based

Table 2 Indicators for tungsten beneficiation

Mineral	Dissemination size of minerals/mm	Beneficiation recovery/%		
		WO_3 <0.2	0.2≤WO_3<0.4	WO_3 ≥0.4
Wolframite (≥90%)	≥0.2	75	80	82
	<0.2	70	72	81
Scheelite (≥90%)	≥0.2	70	74	76
	<0.2	68	71	72
Wolframite and scheelite (Wolframite or scheelite >10%)	≥0.2	59	62	64
	<0.2	56	60	62

Data from Ministry of Natural Resource of China [40]

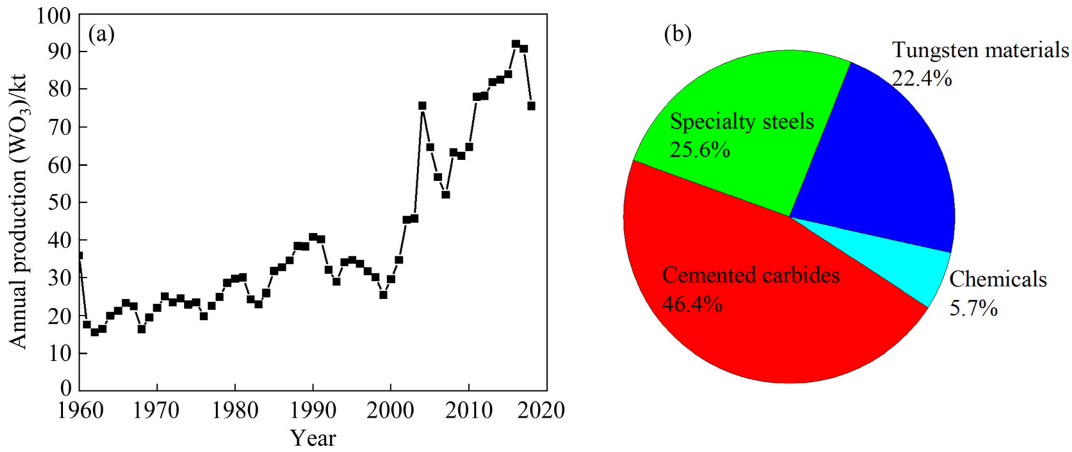

Fig. 4 Change in annual production of tungsten in concentrate (a) and end-use of tungsten (b)

on the GDP projections. The historical Chinese population and GDP data were taken from National Bureau of Statistics of the People's Republic of China [42] and World Bank [43], respectively. From 2017 to 2019, China's GDP grew at annual rates of 6.95%, 6.75% and 5.95%, respectively. Due to the impact of COVID-19, China's GDP growth rate in 2020 was only 2.30%. However, it is expected that by 2021, China will have shaken off the negative impact of the epidemic, and the annual GDP growth rate will return to the pre-epidemic level [44]; therefore, five scenarios in the period from 2021 to 2035 for China were studied, with five potential annual GDP growth rates (5.5%, 6.0%, 6.5%, 7.0%, and 7.5%). China's population in the period from 2021 to 2035 has been forecast (medium variant) by the United Nations [45].

The results of statistical analysis have shown that the Pearson's correlation coefficient (r) between GDP/capita and the output of tungsten in concentrate in China is 0.94119. Thus, the linear regression analysis was conducted using the historical production of tungsten in concentrate and GDP/capita (Fig. 5(a)), and the future output of tungsten in concentrate was estimated using the equation shown in Fig. 5(a). The historical and forecast GDP/capita values of China under the five scenarios (from 1960 to 2035) are shown in Fig. 5(b). Under these five scenarios with different annual GDP growth rates, the mine production of tungsten (WO_3) in concentrate in China by 2025 was estimated to be 128−139 kt (Fig. 5(c)). The growth rate of this production in 2025 compared with that in 2015 was calculated to be 58%−65%. By 2035, the production of tungsten (WO_3) in concentrate was estimated to be 203−262 kt, and this growth rate, compared with the production in 2015, was calculated to be 141%−212%. The cumulative tungsten production is shown in Fig. 5(d). At the end of 2015, the tungsten reserves (WO_3) and identified tungsten resources (WO_3) were estimated to be 2331 kt and 9558 kt, respectively. Based on the prediction, China's current tungsten reserves will be exhausted by approximately 2032. However, with progress in the mineral processing technology or a rise in metal

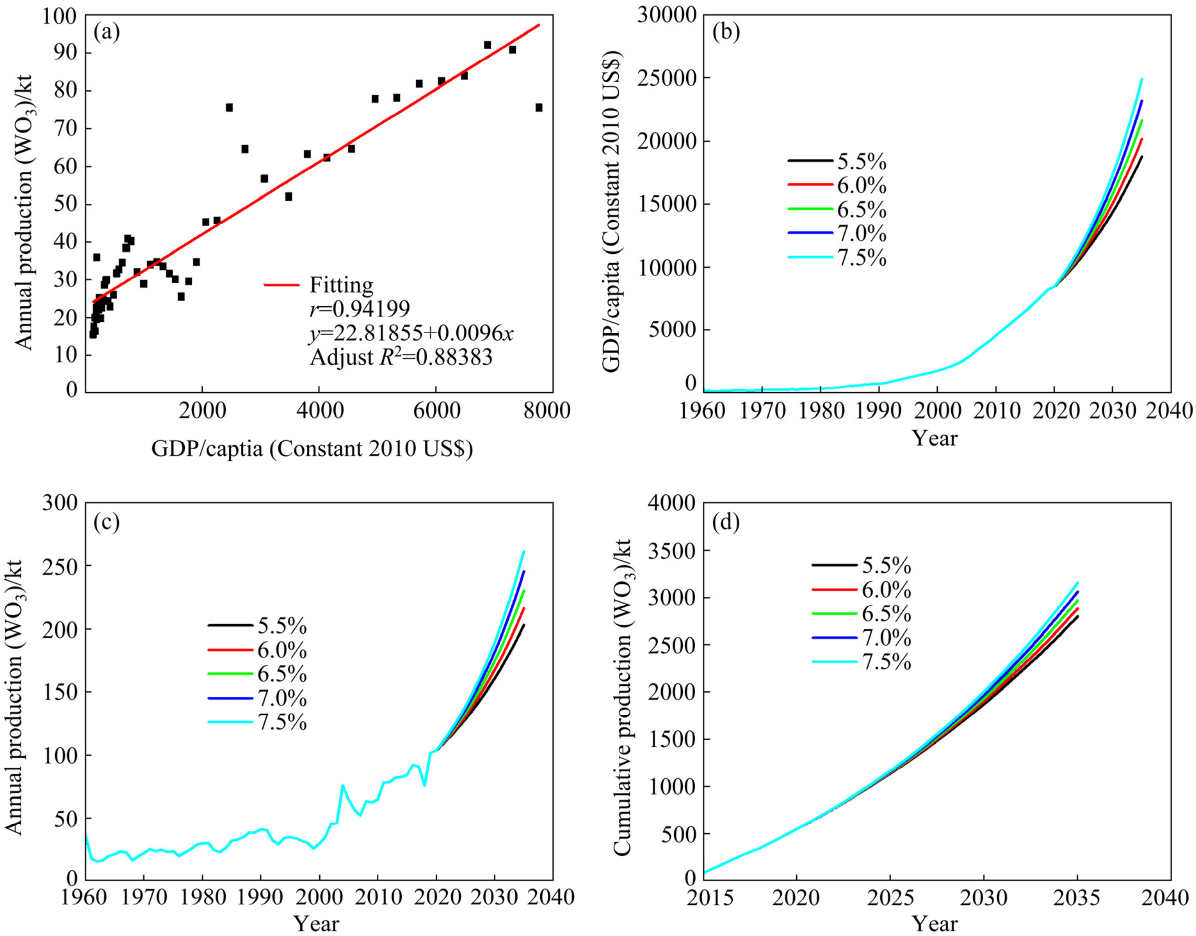

Fig. 5 Annual production of tungsten in concentrate as function of GDP/capita (constant 2010 US$) (a), GDP/capita (b), historical and forecast annual (c) and cumulative (d) production of tungsten in concentrate in China in period from 1960 to 2035 at different annual GDP growth rates

prices, today's identified tungsten resources can also become future tungsten reserves.

3 Tungsten beneficiation technology in China

The beneficiation process of tungsten ore ultimately depends on the geological morphology and mineralogy of tungsten deposits. The beneficiation methods of tungsten mainly include gravity separation, flotation, and magnetic separation [46].

3.1 Wolframite beneficiation

Gravity separation, a low-cost and environmentally friendly process, is often employed for tungsten beneficiation due to the large specific gravity of tungsten, especially for wolframite [47]. Most of China's wolframite is associated with quartz, and its grain size is relatively coarse, which makes it easy to liberate. Therefore, gravity separation is an inevitable choice for wolframite in China [48]. After years of development, a typical process, "pre-enrichment—manual selection and discarding — multisection jigging — multisection shaker—multisection grinding—shaker scavenging", has been formed [49].

The equipment is the key factor in gravity separation of wolframite. Shaker, jigger, spiral chute and cyclone are the common wolframite gravity separation equipment in China [50]. With the improvement of the manufacturing level, the enhanced gravity separators such as centrifugal concentrators and suspension vibration concentrators have been introduced into the field of wolframite gravity separation in China [51]. The centrifugal concentrator has a remarkable effect on the recovery of fine wolframite [52]. The fine-particle tungsten (<0.037 mm) from the tailings of a wolframite plant in Jiangxi Province of China was recovered by a centrifuge concentrator, and wolframite concentrate with a grade of 30.54% and recovery of 63.74% could be obtained [53]. These devices are all based on the density gap between different minerals to achieve mineral separation under different gravity fields. Different devices are often combined due to their respective advantages and disadvantages in China's wolframite plants [49,54].

Flotation has a prominent effect on fine-particle wolframite from tailings because its separation force is more precise than that of gravity separation [55,56]. As a result, the gravity−flotation combination process is usually attempted to ensure the recovery of wolframite [57−59]. Fatty acids, hydroxamic acid, arsonic acid and phosphonic acid are introduced into the flotation of wolframite. Chinese researchers found that the activation effect of metal ions is essential to the recovery of wolframite by a hydroxamic acid collector [60]. For example, under ideal pulp pH, the Pb^{2+} and $Pb(OH)^{+}$ provided by $Pb(NO_3)_2$ can be adsorbed on the wolframite surface, indicating that more adsorption sites for hydroxamic acid are available [60,61]. DENG et al [62] synthesized a novel surfactant N-(6-(hydroxyamino)-6-oxohexyl) octan amide (NHOO) for wolframite flotation. Within the appropriate range of pulp pH values, NHOO can be adsorbed on the wolframite surface by hydrogen bonds and electrostatic forces. The results of flotation tests indicated that NHOO has a better collecting capacity than benzyl hydroxamic acid and octyl hydroxamic acid due to its stronger adsorption affinity. The synthesis route of NHOO is presented in Fig. 6 [62].

The recovery of wolframite by high-gradient magnetic separators is feasible due to the weak magnetism of wolframite [63]. Magnetic separation is generally combined with gravity separation to further improve the grade of wolframite concentrate [64]. The WO_3 grade of wolframite concentrate obtained by gravity separation of wolframite slime in the Dalong Mine can be increased by 10% by magnetic separation [65]. The magnetic separation process is an effective method for the pretreatment of low-grade wolframite or to separate wolframite from scheelite [49].

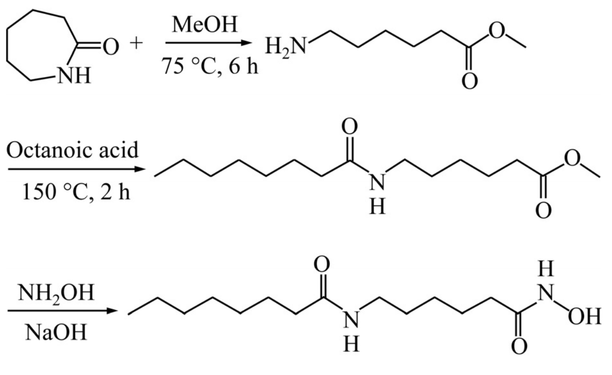

Fig. 6 Synthesis route of NHOO (Reprinted with permission from Ref. [62])

3.2 Scheelite beneficiation

Most scheelite resources in China have the characteristics of fine grain size and low grade and are refractory ores. It is necessary to separate scheelite by flotation.

3.2.1 Scheelite flotation

As the most primary technique at present, scheelite flotation is generally split into two sections in China: roughing and cleaning.

3.2.1.1 Roughing section

The roughing section recovers scheelite and removes gangue as much as possible. The flotation reagent scheme of "weak alkali medium + water glass + fatty acid" is often applied in scheelite roughing sections [66,67]. According to different alkali agents used, the process of roughing sections can be divided into the sodium carbonate method, lime method, sodium hydroxide method, and sodium carbonate + lime method [68]. Some studies have shown that sodium carbonate can not only adjust the pulp pH but also eliminate the inevitable ions in the pulp, which is suitable for scheelite with more associated calcium-bearing gangues. However, sodium hydroxide is more efficient in increasing the pulp pH [69]. SUN et al [70] summarized the experience of dozens of scheelite beneficiation plants and concluded that in general, sodium carbonate is preferred for scheelite mines with more soluble or slightly soluble minerals. Otherwise, sodium hydroxide is preferred.

3.2.1.2 Cleaning section

The quality of the final scheelite concentrate is closely related to the cleaning section. At present, the scheelite cleaning process is mainly divided into a heating flotation process and a normal temperature flotation process.

The traditional heating flotation process is also known as the Petrov process [71]. Heating can increase the floatability difference between scheelite and calcium-bearing gangues. The production index of the heating flotation process is superior and stable, but the energy consumption is high, and the operation is complicated [68]. MENG et al [72] found that HS^- generated by the hydrolysis of Na_2S can promote the desorption of fatty acid GYR on gangue mineral surfaces. Meanwhile, S^{2-} can form insoluble precipitates with free polyvalent metal ions, thus eliminating the activation of Pb^{2+} to nontarget minerals in the pulp [72].

The normal temperature flotation process is easy to operate and inexpensive, but the production index is not stable enough. The process is employed in some scheelite plants with low calcium gangue content in southern China, such as the Dangping Tungsten Mine and Xianglushan Tungsten Mine [73].

3.2.2 Flotation reagents

An effective flotation reagent scheme is undoubtedly one of the simplest ways to improve the separation effect of scheelite.

3.2.2.1 Collectors

The collectors used in scheelite flotation can be divided into four categories according to their own molecular structure characteristics: anionic collectors, cationic collectors, amphoteric collectors, and nonpolar collectors [73].

(1) Anionic collectors

Fatty acids, hydroxamic acid, arsonic acid, sulfonic acid, and phosphoric acid are all classified as anionic collectors. These collectors realize the recovery of scheelite mainly through the adsorption of anionic groups in their molecules and calcium ions on the scheelite surface [74,75].

Fatty acids and their salts are the earliest and most widely used anionic collectors of scheelite, but their low-temperature properties and selectivity are poor. The synergistic effect of polyoxyethylene ether surfactants and sodium oleate on the recovery of scheelite at low temperature was investigated by ZHU et al [76,77]. The results of the mechanism test elucidated that polyoxyethylene ether surfactants could effectively reduce the critical micellar concentration (CMC) of sodium oleate solution and form mixed micelles with sodium oleate to enhance the chemisorption of sodium oleate on the scheelite surface. Therefore, the mixture improved scheelite recovery at low temperature. The structure of the polyoxyethylene group in aqueous solution is presented in Fig. 7 [76].

Hydroxamic acid is another widely used anionic collector of tungsten ore in China. Hydroxamic acid has a better selectivity than sodium oleate due to its selective chelation, but its collection ability is poor [78]. As a result, hydroxamic acid and sodium oleate are often used in combination [12,79]. GAO et al [80] proposed a novel reagent scheme in which sodium hexametaphosphate (SHMP) was used as a depressant and octyl hydroxamic acid (HXMA-8) and sodium oleate (NaOL) were used as a mixed collector. The action mechanism schematic of the flotation reagent scheme is shown in Fig. 8 [80]. The results show that the matching of —CONHOH of HXMA-8 and O—O distance in WO_4^{2-} of scheelite is another key factor in the selective flotation separation of the two minerals, in addition to the large adsorption of SHMP on the calcite surface [80].

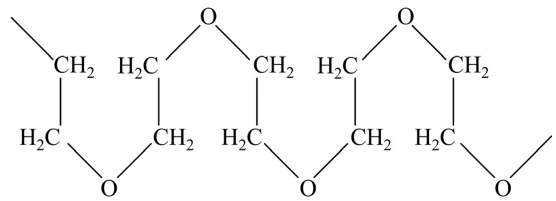

Fig. 7 Structure of polyoxyethylene group in aqueous solution (Reprinted with permission from Ref. [76])

(2) Cationic collectors

Cationic collectors mainly refer to amine collectors [81]. Relevant reports indicate that the surface potential of scheelite is more negative than that of gangue minerals over a wide range of pulp pH values. According to the difference in the point of zero charge (PZC) of different minerals, the pulp pH can be adjusted to maintain the negative charge on the scheelite surface so that the cationic collector is adsorbed on the scheelite surface by electrostatic force [82].

GAO et al [83] established a model (Fig. 9) for the adsorption of dodecyl amine (DDA) on scheelite and calcite surfaces. It is concluded that a small amount of DDA can be adsorbed on the surface of scheelite and calcite through the N—Ca bond and hydrogen bond formed between —NH_2 and mineral surfaces, while most DDA is adsorbed on the mineral surface through the electrostatic interaction between RNH_3^+ and anion groups on mineral surfaces. However, the potential of the

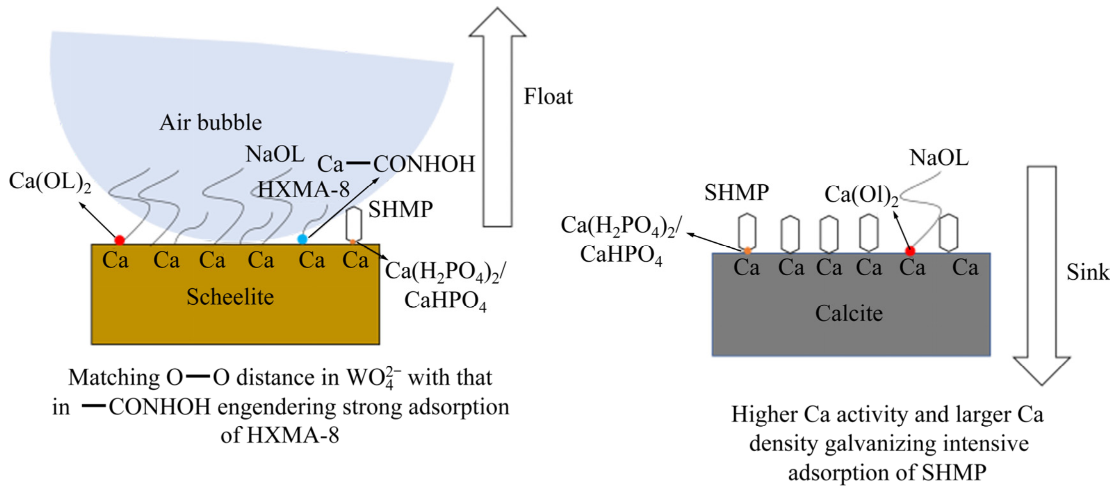

Fig. 8 Schematic diagram of separation mechanism in novel reagent scheme (Reprinted with permission from Ref. [80])

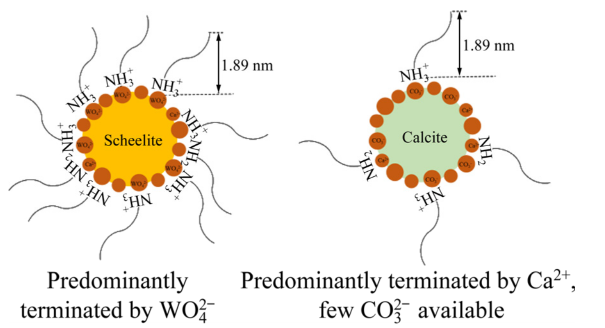

Fig. 9 Schematic diagram of adsorption behavior of DDA species on scheelite and calcite mineral surfaces (Reprinted with permission from Ref. [83])

scheelite surface was dominated by WO_4^{2-}, which can provide more adsorption sites for RNH_3^+. Therefore, a more hydrophobic surface and a better flotation recovery of scheelite were achieved [83].

YANG et al [84] demonstrated that cationic collectors have great potential in the normal temperature flotation separation of scheelite from calcium-bearing gangue. Scheelite and calcite can be separated by flotation with didecyl dimethyl ammonium chloride (DDAC) or trioctyl methyl ammonium chloride (TOAC) as collectors when the pulp pH is 8. At this point, the collecting capacity and selectivity of DDAC or TOAC are significantly better than those of oleate [84].

(3) Amphoteric collectors

Amphoteric collectors mainly refer to alkyl amine phosphonic acids with dissociative groups of two different properties and have strong adaptability to the acidity and alkalinity of pulp [13]. There is still a great deal of research to be done before the industrial application of amphoteric collectors [85].

(4) Nonpolar collectors

Nonpolar collectors mainly refer to hydrocarbon oil collectors. They are inert and often used as additives, which can enhance the hydrophobicity of collectors, promote the hydrophobic agglomeration of mineral particles, and change the foam structure, thereby improving the grade and recovery of scheelite concentrate [73,86].

(5) Anionic−cationic mixed collectors

To balance the advantages of anionic and cationic collectors, some researchers have focused on their mixtures for scheelite flotation. WANG et al [87] studied the effect of the mixed collector of dodecyl amine/sodium oleate on the flotation separation of scheelite and calcite and found that the mixed collector had good collection ability and selectivity.

(6) Metal ion−organic coordinated complexes

In contrast to the traditional method of using Pb^{2+} as an activator and benzohydroxamic acid as a collector to improve the flotation recovery of scheelite, HAN et al [88] mixed Pb^{2+} with benzohydroxamic acid in advance to obtain reactants to further improve the selective collection effect of scheelite. The mechanistic research results revealed that Pb^{2+} and benzohydroxamic acid formed complexes with “O, O” five-member ring or “N, O” four-member ring (Fig. 10 [89]). The adsorption between the collector and target mineral can be regulated directionally through the structure of metal ion−organic complexes. Meanwhile, in the metal ion coordination regulating molecular assembly model, the complexes destroyed the hydration layer structure of the hydrated lead ion component, which was conducive to the adsorption of the collector on the mineral surface. The method has been successfully applied to the Shizhuyuan Polymetallic Mine, and the tungsten recovery has increased by 8% [90].

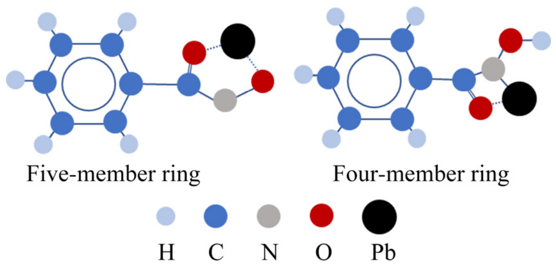

Fig. 10 Diagram of five-member and four-member ring complexes (Adapted from Ref. [89])

3.2.2.2 Depressants

Practice has proven that it is insufficient to selectively collect scheelite only with the collector. Therefore, the study of scheelite flotation reagents in China mainly focuses on depressants of calcium-bearing minerals, especially calcite and fluorite. The depressants of scheelite flotation can be categorized as inorganic and organic depressants.

(1) Inorganic depressants

The inorganic depressants studied in detail mainly include water glass (also known as sodium silicate), inorganic phosphate, sodium fluorosilicate and so on. Water glass can obviously inhibit the flotation of siliceous gangue, but its selective inhibition of calcium-bearing gangue minerals is limited [91,92]. The inhibition of water glass mainly depends on the adsorption of H_2SiO_3, $HSiO_3^-$ and SiO_2 colloids on the surface of calcium-bearing gangue minerals. Water glass is generally acidified or salted to improve the inhibition of calcite [93]. Sodium hexametaphosphate can form soluble chelates with calcium ions on the surface of calcium-bearing minerals. The dissolution of calcium ions on the gangue surface reduces the adsorption sites of the collector [94]. Similarly, sodium fluorosilicate is also used to selectively interact with calcium ions on the surface of gangue minerals to produce new substances ($CaSiO_3$, CaF_2 and $CaSiF_6$) that hinder the adsorption of collectors, thereby achieving the separation of scheelite and calcite [95].

(2) Organic depressants

According to the relative molecular mass, organic depressants can be further split into small molecular depressants and macromolecular depressants. Macromolecular depressants include sodium humate, tannin, starch, lignin sulfonate, carboxymethyl cellulose, etc., and small molecular depressants include oxalic acid, citric acid, tartaric acid, lactic acid, etc. These depressants are also selectively adsorbed on the surface of calcium-bearing gangue minerals by the binding of their characteristic functional groups to calcium ions. These functional groups include carboxyl, hydroxy, sulfonic, phosphate, and others [14,96,97]. For example, sodium phytate and calcium lignosulfonate bind to calcium ions on the calcite surface by phosphate and sulfonic groups, respectively, to produce corresponding calcium salts. Studies have shown that the depressants have little influence on the floatability of scheelite because the scheelite surface is dominated by tungstate groups, which can produce a stronger steric hindrance effect and electrostatic repulsion, preventing the adsorption of most depressants on the scheelite surface [98]. A summary of some of the latest related research on calcium-containing mineral depressants in China is shown in Table 3.

3.3 Tungsten-bearing slime beneficiation

A large amount of primary slime and secondary slime will be produced during the tungsten beneficiation process. To prevent the loss of tungsten resources, these tungsten-bearing slimes in China's tungsten plants are generally collected and stored for recycling. In addition to conventional tungsten beneficiation methods, the tungsten-bearing slime recovery process also includes selective flocculation, carrier flotation and oil agglomeration flotation [57,109,110].

Selective flocculation mainly involves adding a selective flocculant to flocculate the target mineral and then using conventional processes to recover fine-grained tungsten. The regularly employed flocculants are sodium polyacrylate, starch, gelatin, polymeric inorganic metal salts and so on. LU [111] investigated the flocculation flotation of fine-particle wolframite (<2 μm) by using polyacrylic acid (PAA) as the flocculant and sodium oleate as the collector. The results showed that the recovery could be increased by 17.83%, and

Table 3 Summary of new depressants of calcium-bearing gangues

Regent	Main functional group	Research method*	Application
Starch [99]	Hydroxyl	1, 2, 4	Laboratory
Tanning [100]	Carboxyl group, hydroxyl	1, 2, 4	Laboratory
Xanthan gum [101,102]	Carboxyl group	2, 3, 5, 6	Laboratory
Pectin [103]	Carboxyl group, hydroxyl	2, 3, 4, 5	Laboratory
Sodium alginate [98]	Carboxyl group, hydroxyl	2, 3, 4, 8	Laboratory
Sodium silicate + oxalic acid [91]	Silicate root, carboxyl group	2, 3, 4, 7	Laboratory
Dextran sulfate sodium [104]	Sulfate radical	2, 3, 4, 8	Laboratory
Calcium lignosulphonate [105]	Sulfonic acid group	2, 5, 6	Laboratory
Sodium phytate [14]	Phosphate group	2, 3, 4	Laboratory
Phytic acid [106]	Phosphate group	2, 3, 4, 5, 7	Laboratory
Etidronic acid [107]	Phosphate group	1	Industrial
Sodium hexametaphosphate [94]	Phosphate group	1, 2, 4	Laboratory
Sodium fluorosilicate [95]	Fluosilicate root	2, 3, 4, 5, 7	Laboratory
Sodium tripolyphosphate [108]	Phosphate group	1, 2, 3, 4, 6	Industrial

*1—Actual ore flotation test; 2—Microflotation test; 3—FTIR analysis; 4—Zeta potential measurement; 5—XPS analysis; 6—Adsorption test; 7—Solution chemistry analysis; 8—Contact angle measurement

the WO_3 grade could be increased by 2.94%. CHEN et al [112] studied the effect of energy input on the flocculation and flotation behavior of fine-particle scheelite (<10 μm). The results showed that under high energy input conditions, sodium oleate showed the dual function of flocculant and collector, and scheelite recovery was improved.

Carrier flotation uses coarse-particle minerals as carriers to recover fine-particle minerals. This process has strict requirements on the target minerals and carriers. PAN et al [113] used coarse-particle wolframite (>0.01 mm) as a carrier to enhance the recovery of fine-particle wolframite (<0.005 mm), and the recovery increased from 40.5% to 70.38%.

The oil agglomeration flotation process refers to the aggregation of ore particles into spherical agglomerates under the bridging action of neutral oil to recover tungsten slime [114]. Using sodium oleate as the collector and fuel oil as the binder, WEI et al [115] obtained a tungsten concentrate with WO_3 grade of 69.72%−70.65% and recovery of 90.95%−91.62% from wolframite slime with a particle size of <15 μm. The disadvantage of this process is the high production cost due to the large amount of neutral oil.

3.4 Prospect of tungsten beneficiation technology

As the grade of feeding is generally low, the tungsten beneficiation process is a relatively high energy consumption process, especially scheelite. Many attempts have been made to improve the beneficiation efficiency of tungsten. The existing problems and future development trends of tungsten beneficiation technology are summarized from the following aspects.

(1) At present, the collectors and depressants used in industry still do not have good selectivity. Although a large number of studies have been carried out on flotation reagents with good selectivity, most of these studies have been confined to laboratory-scale flotation tests, such as the depressants of sodium alginate, sodium phytate, dextran sulfate sodium, sodium alginate and vegetable gum mentioned above. Further industrial trials are needed to screen out practically and reasonably priced depressants for industrial application.

(2) The feeding grade of tungsten beneficiation needs to be further improved. It is a trend to remove gangue minerals in advance to reduce the beneficiation cost. The application of an X-ray fluorescence (XRF) intelligent preconcentrator greatly improves the efficiency of the preremoval of gangue [116]. It is worth introducing into the tungsten beneficiation process. However, attention should be paid to the accuracy of the equipment for the separation of low-grade tungsten to avoid

resource waste.

(3) The recovery of tungsten-bearing slime is not high enough. Tungsten ore is brittle and prone to excessive grinding. New equipment with a stronger separation field is needed to strengthen the separation process of tungsten-bearing slime, such as cyclonic-static flotation columns.

4 Typical beneficiation flowsheets in China

To better understand the process flow of tungsten beneficiation in China, several typical tungsten beneficiation plants are introduced, covering wolframite, scheelite, mixed ore and tungsten-bearing slime. These plants can basically represent the current level of tungsten beneficiation technology in China.

4.1 Dajishan Tungsten Mine in Jiangxi Province of China

The Dajishan Tungsten Mine is one of the earliest quartz vein-type wolframite deposits developed and utilized in China. The main metallic minerals of the Dajishan tungsten deposit are wolframite, scheelite, natural bismuth, bismuth, molybdenite, beryl, pyrite and pyrrhotite [117]. The mass ratio of wolframite to scheelite is 3:1−4:1, and gangue minerals are mainly quartz, mica, feldspar, fluorite, chlorite and calcite. The multi-element chemical analysis results of raw ore are shown in Table 4 [118].

After manual reverse selection, the typical combined process of jigger and shaking table is adopted in the tungsten roughing stage of the Dajishan Beneficiation Plant. Then, the rougher concentrate is screened and reground. The grade of concentrate is continuously improved by a jig or shaker in the tungsten cleaning stage. Finally, the wolframite and scheelite in the qualified tungsten concentrate are separated by a high gradient magnetic separator. The other valuable metals in the tailings of tungsten gravity separation are recovered by flotation. The main flowsheet of the Dajishan Plant is shown in Fig. 11 [119]. However, fine-particle tungsten in primary and secondary slimes is difficult to recover by gravity separation. To reduce the loss of tungsten resources, the flotation of tungsten slime is performed using a regent scheme of sodium carbonate + water glass +

Table 4 Multi-element chemical analysis results of Dajishan Tungsten Mine (wt.%) [118]

WO_3	Bi	Mo	S	MnO	S	Fe_2O_3	Al_2O_3	SiO_2
0.29	0.03	0.012	0.32	0.25	0.32	6.28	9.90	68.06

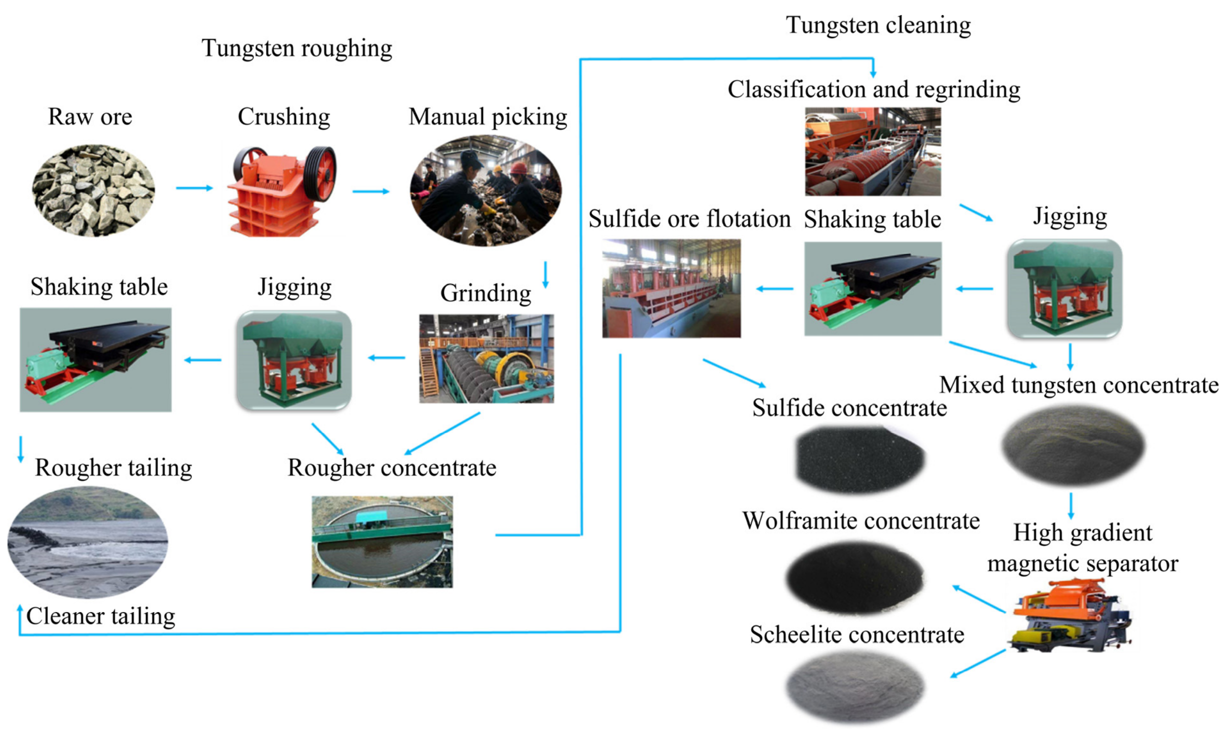

Fig. 11 Main process of Dajishan Beneficiation Plant (Drawing based on Ref. [119])

oxidized paraffin soap. The tungsten slime recovery process is shown in Fig. 12 [120]. The beneficiation index of Dajishan tungsten is as follows [118]: the WO_3 grade of gravity concentrate is higher than 70%, the WO_3 grade of flotation concentrate is higher than 50%, and the total tungsten recovery is approximately 85%. However, the recovery of tungsten-bearing slime is only approximately 50%. The amount of tungsten in slime accounts for approximately 8% of that in the raw ore. Therefore, strengthening the recovery of slime is a critical way to further improve the utilization rate of tungsten resources.

4.2 Shizhuyuan Tungsten Mine in Hunan Province of China

Shizhuyuan polymetallic ore is an extra-large tungsten−molybdenum−bismuth−fluorite skarn deposit [37]. The multi-element chemical analysis results of raw ore are shown in Table 5 [121]. The valuable metals that can be recovered in the ore include scheelite, wolframite, molybdenite, bismuth, pyrite, pyrrhotite, chalcopyrite, and cassiteritec. The mass ratio of wolframite to scheelite is approximately 1:2. The main process of the Shizhuyuan Beneficiation Plant before 2015 is presented in Fig. 13 [118].

The separation of polymetallics is a worldwide challenge for Shizhuyuan Plant. The bulk flotation process of scheelite and wolframite was employed after sulfide ore flotation in Shizhuyuan Plant. However, the flotation reagent scheme using caustic soda + fatty acids to recover wolframite and scheelite is no longer considered because it is not conducive to the subsequent recovery of fluorite. After a great deal of research, a novel flotation

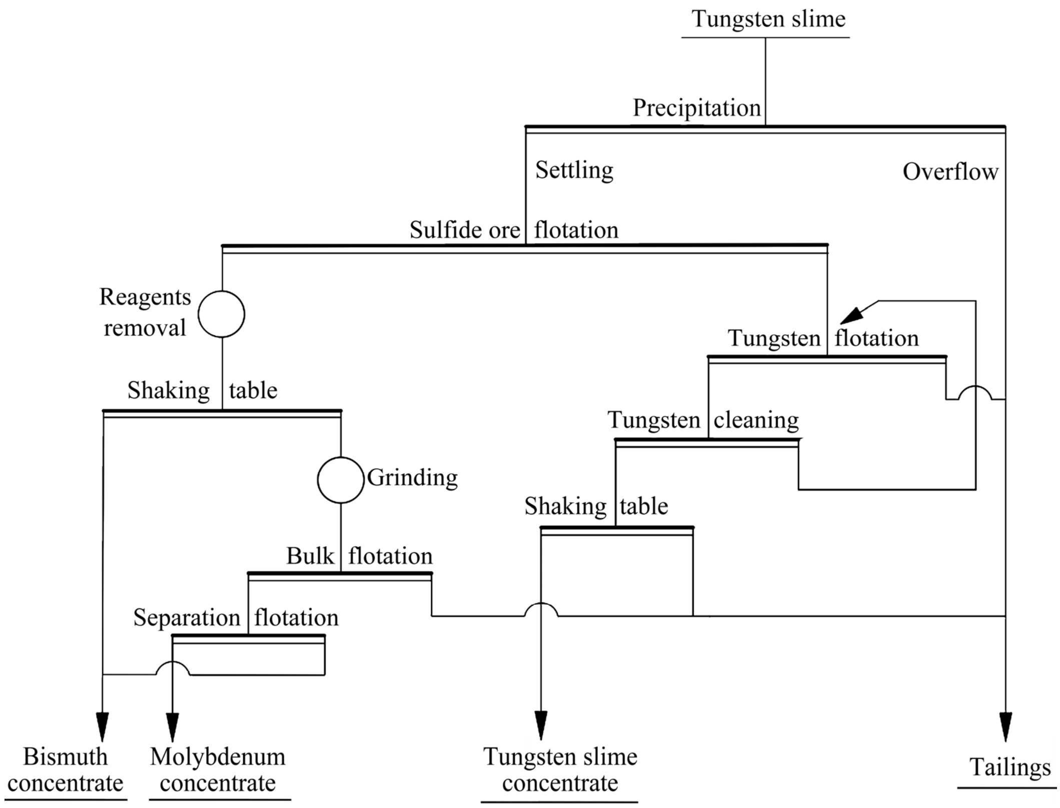

Fig. 12 Recovery process of tungsten slime (Drawing based on Ref. [120])

Table 5 Multi-element chemical analysis results of raw ore from Shizhuyuan Tungsten Mine (wt.%) [121]

WO_3	Bi	Mo	S	TFe	P	Pb	Zn	Cu
0.380	0.02	0.01	1.11	9.72	0.017	0.011	0.06	0.03
Sn	Mn	MgO	CaF_2	$CaCO_3$	Al_2O_3	Na_2O	K_2O	SiO_2
0.06	0.65	19.68	1.11	4.31	0.32	0.74	1.61	41.03

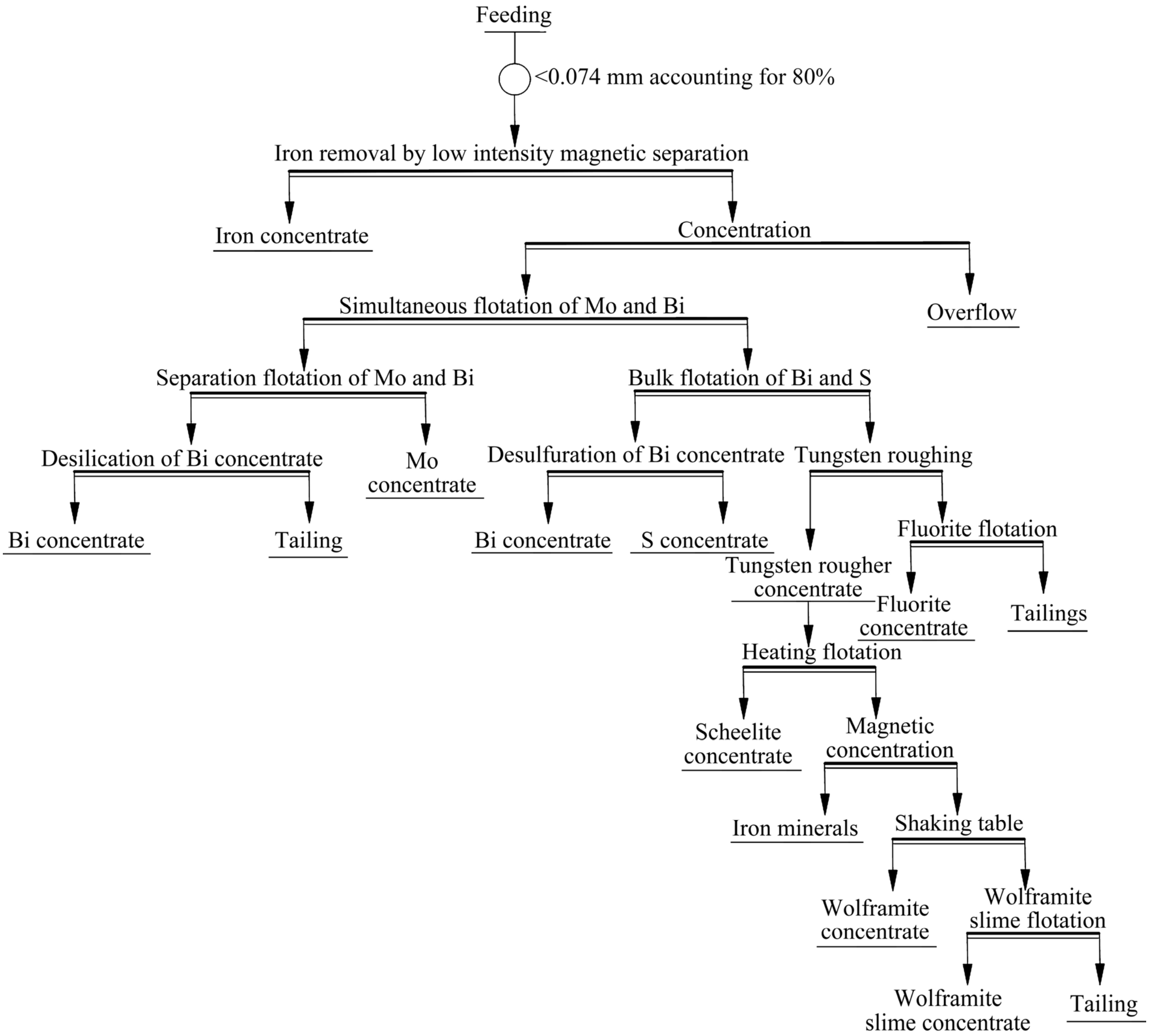

Fig. 13 Main process of Shizhuyuan Beneficiation Plant before 2015 (Drawing based on Ref. [118])

reagent scheme for bulk flotation was developed successfully with lead nitrate as an activator, sodium hexametaphosphate and aluminum sulfate as combination depressants, a chelating collector and a small amount of fatty acids as an auxiliary collector. Chelating collectors and lead nitrate activators are the core of the new process. The chelating collectors used in production are hydroxamic acid ore N-nitroso-naphthylamine ammonium. Recently, researchers from Central South University, China have innovatively developed lead(II)-benzohydroxamic acid complexes and Al−Na_2SiO_3 polymer depressants, which have been applied to Shizhuyuan Plant, greatly simplifying the flotation process and eliminating the pulp heating process (Fig. 14) [88,122].

4.3 Xianglushan Tungsten Mine in Jiangxi Province of China

The Xianglushan tungsten deposit is a skarn-type scheelite polymetallic deposit. The metallic minerals mainly include scheelite, pyrite, pyrite, chalcopyrite, molybdenite, stibnite, bismuth, wolframite, rutile and so on. The gangue minerals include quartz, potassium feldspar, plagioclase, tremolite, fluorite, apatite, calcite, garnet, and barite [37]. The multi-element chemical analysis results of raw ore are shown in Table 6 [123]. Scheelite accounts for approximately 95% of the total tungsten ore, and wolframite only accounts for approximately 4%.

Xianglushan scheelite is recovered by the whole process of normal temperature flotation

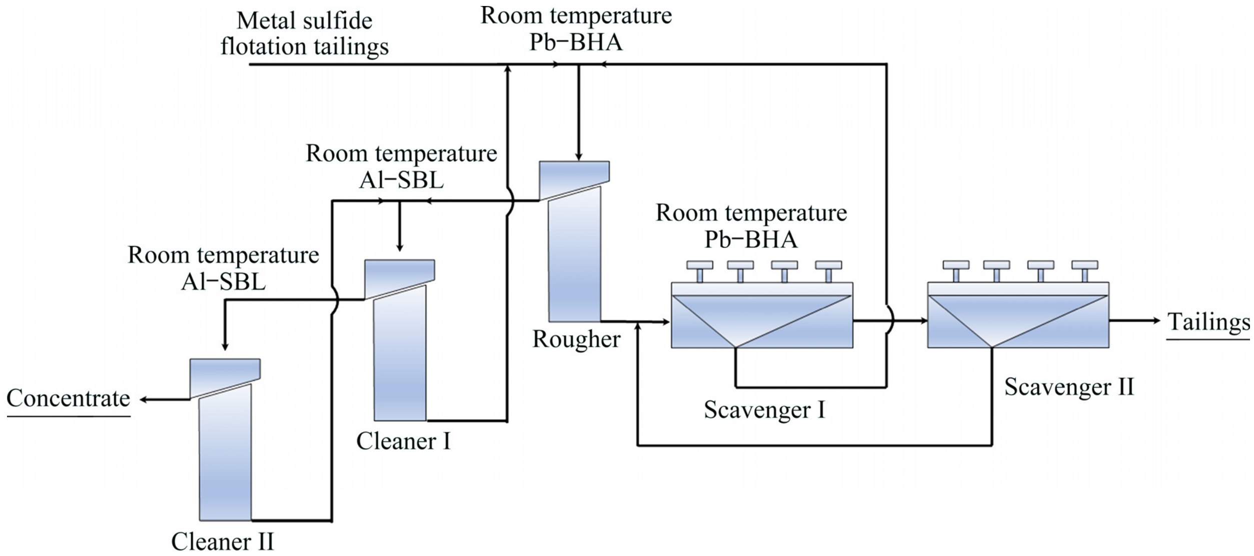

Fig. 14 Flowsheet of recovery of tungsten from molybdenum tailings [88,122]

Table 6 Multi-element chemical analysis results of Xianglushan Tungsten Mine (wt.%) [123]

WO_3	Bi	Mo	Cu	Pb	Zn	As
0.75	0.036	<0.001	0.15	0.011	0.033	<0.002
S	$CaCO_3$	CaF_2	Al_2O_3	SiO_2	Fe	
4.76	5.68	6.35	4.55	53.96	11.34	

(Fig. 15 [123]). The scheelite roughing adopts the flotation reagent scheme of sodium carbonate + sodium silicate + ZL type fatty acid sodium. A large amount of sodium silicate is added to strengthen the inhibition of gangue minerals during scheelite cleaning. The total recovery of scheelite in Xianglushan Tungsten Mine is approximately 78%, and the WO_3 grade of scheelite concentrate is more than 65% [118]. Although the energy consumption of normal temperature flotation is small, the separation efficiency between scheelite and calcium-containing gangue is lower than that of heating flotation, which will lead to a certain amount of scheelite loss. Therefore, the development of depressants with good selectivity is an alternative to improve the recovery of Xianglushan scheelite.

4.4 Sandaozhuang Tungsten Mine in Henan Province of China

Recovering tungsten from the flotation tailings of sulfide ores such as molybdenum and bismuth can improve the resource utilization rate. The Sandaozhuang mining area is a superlarge skarn-type molybdenum and tungsten deposit with proven molybdenum reserves of 672.5 kt and associated tungsten reserves of 502.5 kt. More than 95% of tungsten exists in the form of scheelite [124,125]. Luoyang Yulu Tungsten Mining Co., Ltd., is a representative enterprise in recovering scheelite from Sandaozhuang molybdenum flotation tailings. The multi-element chemical analysis results of raw ore are presented in Table 7 [126].

The relevant technical indexes and process flow of scheelite flotation are shown in Figs. 16 and 17 [127], respectively. The process consists of two sections of roughing and cleaning. In the cleaning section, a typical Petrov process is employed. Finally, tungsten concentrates with WO_3 grades of 29%−32% and recovery of 75.0%−80.2% can be obtained (Figs. 16(b−d)). However, the high energy consumption of heating flotation is the bottleneck of enterprise development. In the case of not being able to completely ban heating flotation, increasing the WO_3 grade of rough concentrate and reducing the load of the cleaning section are good choice to save energy consumption. The WO_3 grade of scheelite rough concentrate is only approximately 1.5%, which has great room for improvement compared with tungsten ore with the flotation process (e.g., the WO_3 grade of Shizhuyuan rough concentrate is 6%−20%; the WO_3 grade of Xianglushan rough concentrate is approximately 10%).

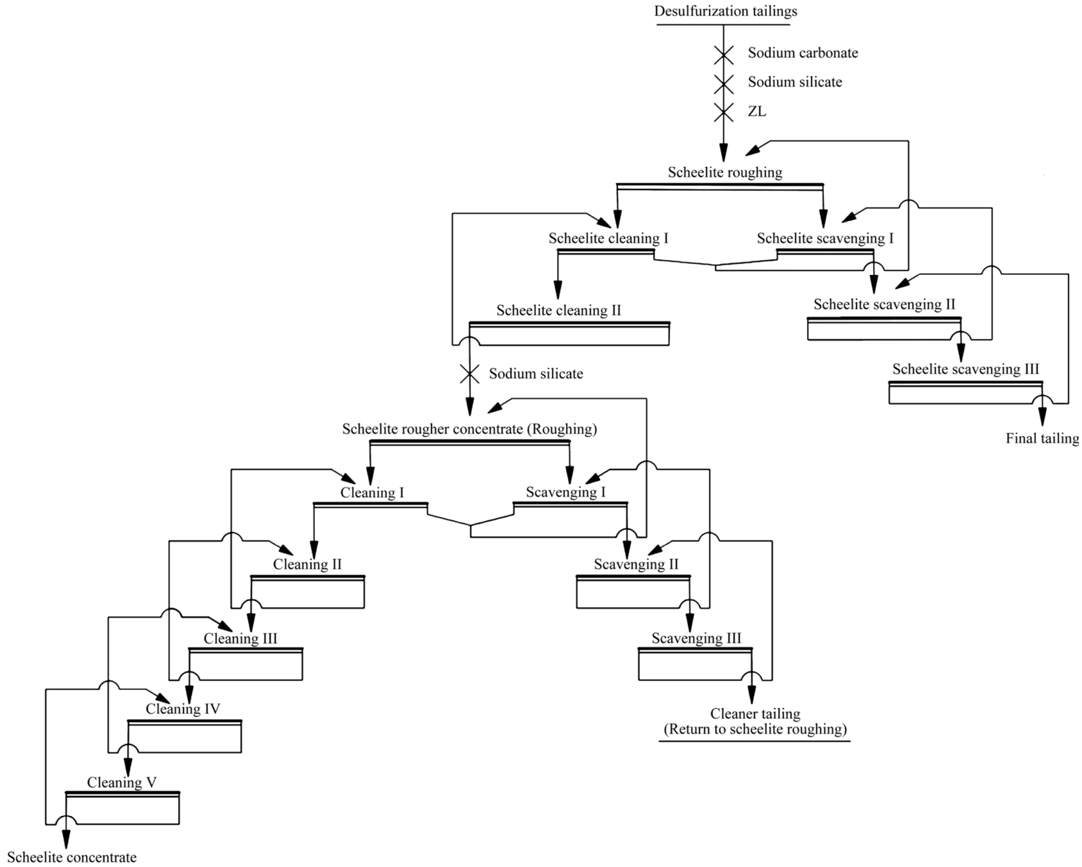

Fig. 15 Scheelite flotation process of Xianglushan Beneficiation Plant (Drawing based on Ref. [123])

Table 7 Multi-element chemical analysis results of Sandaozhuang Tungsten Mine (wt.%) [126]

WO_3	Mo	S	P	Cu	Pb	Zn
0.065	0.25	1.08	0.20	0.022	0.02	0.037
S	$CaCO_3$	CaF_2	CaO	MgO	Fe_2O_3	SiO_2
4.76	4.20	4.88	25.00	1.51	6.21	48.71

5 Conclusions

(1) China's tungsten resource reserves and tungsten concentrate output both rank first in the world. Based on GDP/capita under five different annual GDP growth rate scenarios, the production of tungsten WO_3 in concentrate by 2035 can be estimated as 203−262 kt, with a growth rate of 141%−212% compared to 2015. The cumulative tungsten production is expected to exceed China's current tungsten reserves by approximately 2032, although it is not expected to exceed the currently identified tungsten resources.

(2) Tungsten beneficiation technology has always been a research hotspot in China, regardless of the process flow, beneficiation equipment and flotation reagents. Increasing attention should be given to the industrial application and promotion of new technology. The flotation separation of scheelite and calcium-bearing gangue minerals, the recovery of tungsten-bearing slime and the beneficiation cost are still bottlenecks restricting the utilization of tungsten resources.

(3) Taking several representative tungsten mines in China as examples, the classic tungsten beneficiation processes are elaborated. The beneficiation index of wolframite is better than that of scheelite and mixed ore. Flotation reagents with high efficiency and excellent selectivity are urgently needed by scheelite plants.

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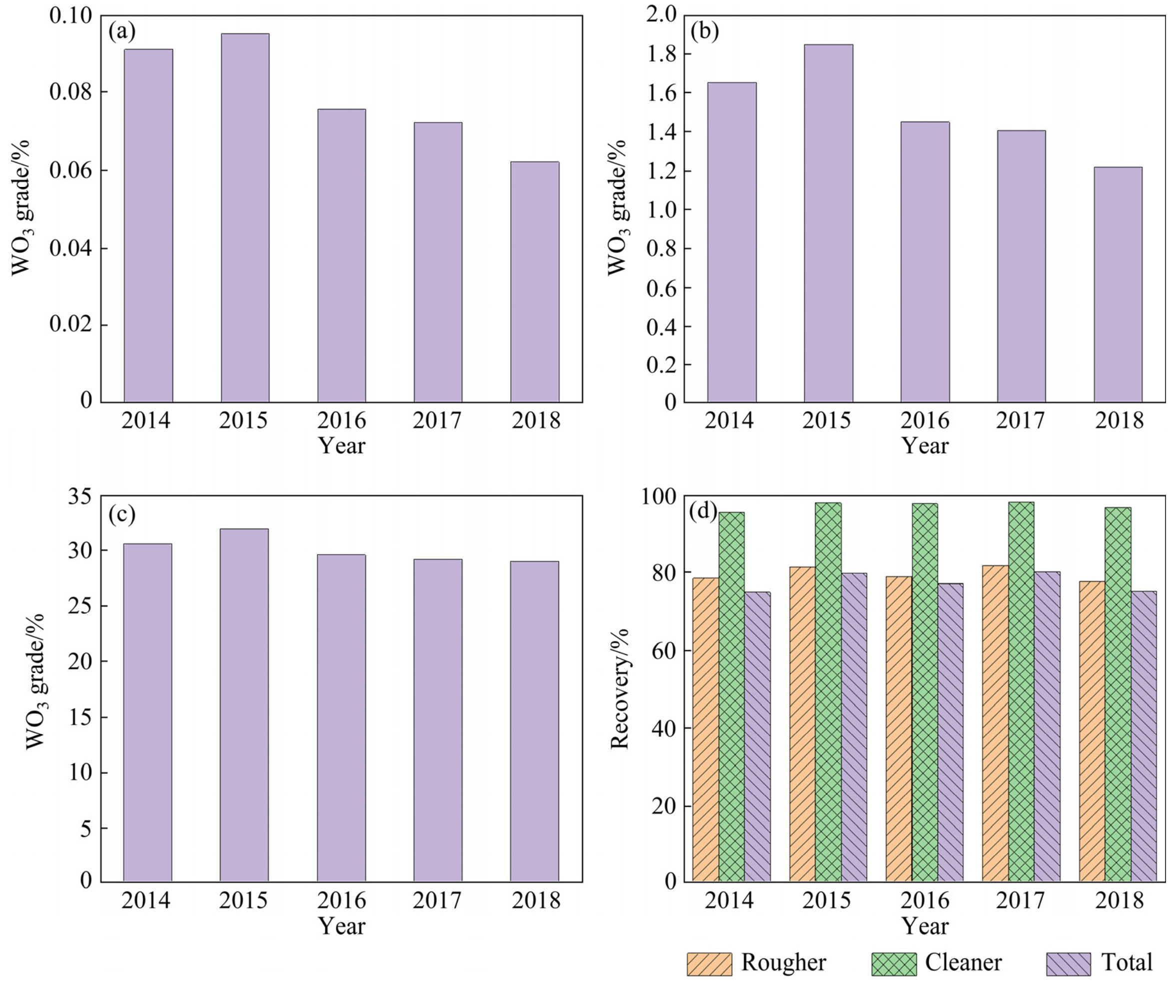

Fig. 16 Technical indicators of tungsten beneficiation: WO_3 grades of feeding (a), rougher concentrate (b) and concentrate (c), and recovery of rougher, cleaner and total (d) [127]

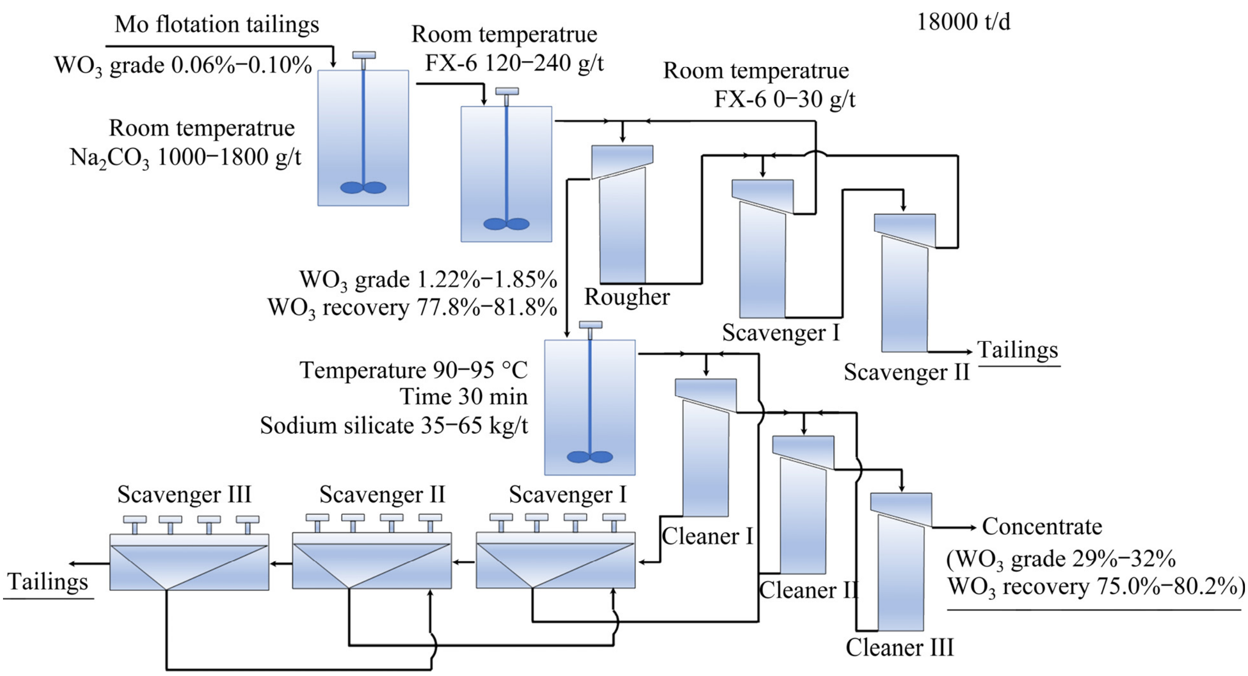

Fig. 17 Flowsheet of recovery of tungsten from molybdenum tailings (Drawing based on Ref. [127])

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中国钨资源储量、钨精矿生产和钨选矿技术综述

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摘 要：钨是一种战略金属，由于其特殊的性能，因此被广泛应用于电子通信、航空航天、材料和化工等领域。中国是世界上钨储量最大、钨精矿产量最大的国家，在世界钨市场中起着举足轻重的作用。本文首先总结中国钨资源的储量和分布特点，并对中国钨精矿生产现状进行综述。以人均国内生产总值(GDP/capita)为基础，采用 5 个不同的 GDP 年增长率来估算未来中国的钨精矿产量。估算结果表明，如果中国经济继续以目前的速度增长，到 2032 年左右，钨精矿的累计产量将超过当前储量。然后，从工艺流程、选矿设备和浮选药剂等方面综述中国黑钨矿、白钨矿和含钨矿泥等不同类型钨矿的选矿技术发展情况。最后，以中国几个具有代表性的钨矿为例，详细阐述不同类型钨矿的经典选矿工艺。同时，提出未来钨矿选矿技术的发展方向，对保持中国钨矿资源优势具有重要意义。

关键词：钨；储量；生产；选矿；浮选；重选

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