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# Extraction and recycling technologies of cobalt from primary and secondary resources: A comprehensive review

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**Abstract:** Cobalt has excellent electrochemical, magnetic, and heat properties. As a strategic resource, it has been applied in many high-tech products. However, the recent rapid growth of the battery industry has substantially depleted cobalt resources, leading to a crisis of cobalt resource supply. The paper examines cobalt ore reserves and distribution, and the recent development and consumption of cobalt resources are summarized as well. In addition, the principles, advantages and disadvantages, and research status of various methods are discussed comprehensively. It can be concluded that the use of diverse sources (Cu–Co ores, Ni–Co ores, zinc plant residues, and waste cobalt products) for cobalt production should be enhanced to meet developmental requirements. Furthermore, in recovery technology, the pyro-hydrometallurgical process employs pyrometallurgy as the pretreatment to modify the phase structure of cobalt minerals, enhancing its recovery in the hydrometallurgical stage and facilitating high-purity cobalt production. Consequently, it represents a promising technology for future cobalt recovery. Lastly, based on the above conclusions, the prospects for cobalt are assessed regarding cobalt ore processing and sustainable cobalt recycling, for which further study should be conducted.

**Keywords:** cobalt recovery; copper–cobalt ore; nickel–cobalt ore; zinc plant residue; waste cobalt products

## 1. Introduction

As science and technology have advanced and developed, the focus of metal supply has gradually shifted from bulk metals such as steel, aluminum, and copper to metals of “special interest” such as lithium, cobalt, indium, and rhenium. Undoubtedly, “special interest” metals, also known as critical metals, are indispensable for high-technology products and emerging innovations, which have recently been the focus of worldwide attention.

Cobalt, a so-called “devil’s metal”, is similar to iron and nickel in terms of its physical and chemical properties [1]. Traditional industries mainly use cobalt to produce heat-resistant, hard, and magnetic alloys [2–4]. Cobalt heat-resistant alloys are particularly popular in gas turbine blades, impellers, and chemical equipment because of their excellent thermal resistance. The recent development of the new energy industry stimulated cobalt consumption in electrode materials [5], and the applications and production of cobalt and its compounds rapidly increased [6]. The irreplaceability of cobalt in various industries is a vital reason why cobalt is defined in national strategies for the raw materials of key metal minerals. Because of the rapidly increasing demand and resource security of cobalt, the secure supply of cobalt

minerals and raw materials has received worldwide attention to ensure the safe development of new energy fields, and cobalt resources have been classified as critical minerals by various countries [7]. In 2009, Japan initiated the Strategy for Securing Rare Metals, followed by the European Commission’s issuance of Key Mineral Raw Materials of the European Union in 2010 and China’s introduction of the National Mineral Resources Plan (2016–2020) in 2016. The US Department of the Interior also published a list of critical minerals in 2022.

Although China is the world’s largest cobalt refiner, it has only 1.88% of global cobalt reserves. Thus, the production of domestic cobalt enterprises is dominated by imported cobalt ore, with an external dependence of more than 90% [8], which poses a considerable risk to the cobalt supply. In addition, with the changing cost of ore imports and the international situation, the cobalt smelting industry faces enormous challenges. Therefore, the efficient recovery of cobalt-containing resources is crucial before cobalt can be replaced, and research on the extraction technology of cobalt-containing resources has become a hot spot in the development of the cobalt industry.

In recent decades, much research has been conducted on cobalt recovery technologies, particularly with regard to

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waste batteries. However, few systematic reviews of the recovery of cobalt-containing resources are available. Herein, the resource characteristics, supply, and demand of cobalt are briefly introduced, and the technology and methods for extracting cobalt from different ores and waste batteries are systematically reviewed. The principles, advantages, and disadvantages of processes are compared in detail. Finally, according to the characteristics of resources and technology, cobalt extraction technology and measures to address the risks of the cobalt supply are discussed.

## 2. Resources and consumption of cobalt

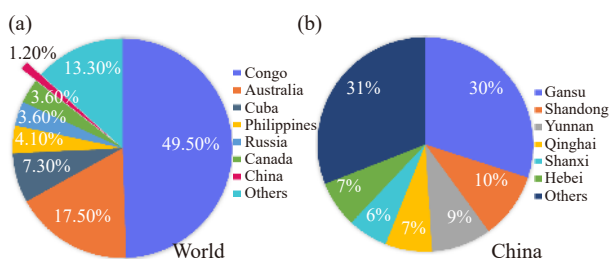
### 2.1. Mineral resources

Nearly 100 cobalt-containing minerals [9] occur in nature, with confirmed terrestrial cobalt mainly comprising sedimentary copper–cobalt deposits, laterite–nickel ore deposits, and magmatic Ni–Cu sulfide deposits. Meanwhile, seafloor cobalt resources mainly occur in manganese nodules. The deposits, distribution, reserves, and content of cobalt are

shown in Table 1. It is seen that earth has no cobalt-only deposits, and cobalt resources are associated with copper, nickel, and zinc ores. Fig. 1(a) shows the distribution and proportion of the world's cobalt-containing deposits, which are concentrated in the Democratic Republic of the Congo (DRC), Australia, Cuba, the Philippines, Russia, Canada, and other countries [18–19]. More than 70% of the world's cobalt supply is controlled by countries such as the DRC, Australia, and Cuba. Having limited cobalt reserves, China only possesses 137400 tons of dispersed deposits. As shown in Fig. 1(b), these deposits are located in Gansu, Xinjiang, Yunnan, Shanxi, Qinghai, and Hubei [20–21]. The reserves of Gansu, Shandong, and Yunnan provinces account for half of the domestic cobalt reserves [22]. The largest cobalt deposit is situated in Jinchuan, Gansu Province, China. However, the average cobalt grade within this mine is merely 0.019wt%. The recovery process poses considerable challenges and expenses compared to cobalt-rich nations, imposing severe resource constraints on the cobalt industry.

**Table 1.** Main types, distribution, and content of the world's cobalt deposits [10–17]

Types	Region (Country)	Reserves / Mt	Average cobalt content / wt%
Sedimentary copper–cobalt ore	Mutanda (DRC); Otkamo (Finland); Wubaoshan Cobalt Deposit (China)	11060	0.29
Laterite–nickel ore	Nkamouna (Cameroon); Jakarta (Brasil)	11202	0.08
Magmatic Ni–Co sulfide	Sudbury Basin (Canada); Norilsk (Russia); Yongchang Nickel Mine (China)	12194	0.07
Hydrothermal and volcanogenic Cu (–Zn–Co)VMS	Outokumpu (Finland); Windy Craggy (Canada)	919	0.13
IOCG–Cu–Au (–Ag–U–REE–Co–Ni)	Olympic Dam (Australia); Idaho (America); Shilu Iron Deposit (China)	1067	0.04
Metastded.-roch-hosted Co–Cu–Au	Blackbird distract (America); NICO (Canada); Dahenglu Cobalt Ore (China)	115	0.23
Fe–Cu–Co sharn and replacement	Magnitogorsk (Russia); Yamato (Japan); Tonglushan Mining (China)	760	0.02
Polymetallic (Ag–Ni–As–Bi)–Co-rich veins	Bou-Azzer (Morocco); Cobalt-Gowganda (Canada)	23	0.92
MVT Zn–Pb (–Co–Ni) sulfide	Madison (America); Mt. Isa (Australia)	2	0.27
Seafloor Fe–Mn(–Ni–Co) nodules	Pacific prime crust zone	35936	0.38



**Fig. 1.** Distribution and proportion of cobalt-containing deposits of cobalt reserves in the world [18–19] and China [20–21].

### 2.2. Development and consumption of cobalt

The development of cobalt mines is important to the world cobalt industry since cobalt is a critical metal. Fig. 2 shows the statistics of cobalt mine production [23]. Despite minor fluctuations, the global production of raw ore still exceeds 100000 t·a<sup>-1</sup>. In terms of supply structure, the DRC is the

largest producer, followed by Russia, Australia, the Philippines, and Cuba, while the cobalt output of other nations is negligible. For example, the DRC produced 120000 tons of cobalt ore in 2021, accounting for 72.6% of the world's output, while only 2300 tons of cobalt ore were produced by China, merely 1.3% of global production. Consequently, as a major cobalt processing country, China relies heavily on imports of mineral resources to develop its cobalt industry. Therefore, although global cobalt production will continue to rise steadily, the dependence on the DRC will not change temporarily for cobalt-poor countries.

Moreover, because of the high strength and electrochemical performance of cobalt and its compounds, cobalt and cobaltate products are used extensively to produce power batteries [24], and cobalt cannot be replaced by other metals in the modern economic and social evolution. Fig. 3(a) illustrates the consumption structure of refined cobalt in China over the last ten years. In 2011, the highest consumption of

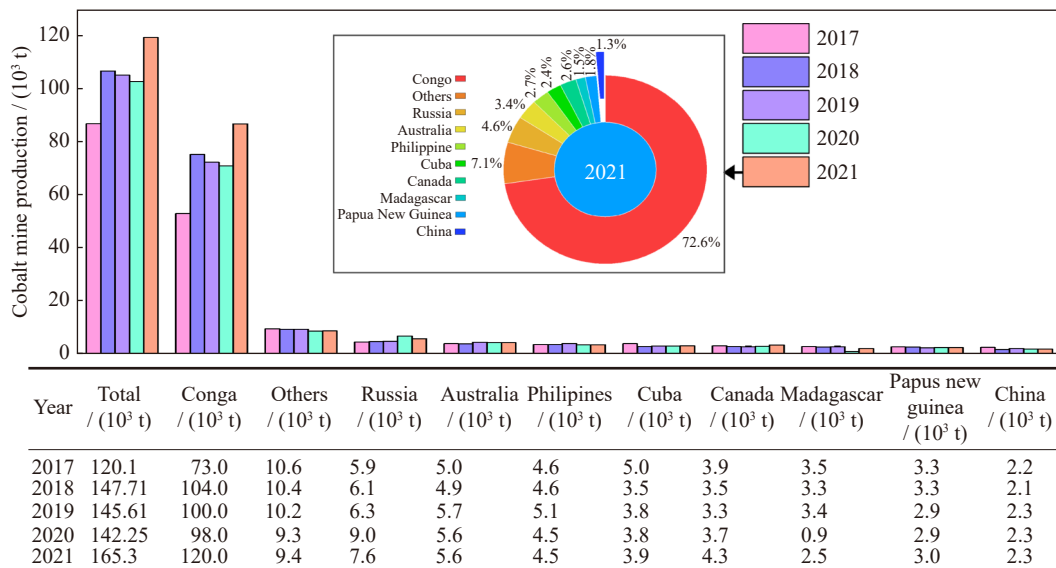


Fig. 2. Statistics of global cobalt mine production from 2017 to 2021 [23] (The inset shows the production proportion of different countries in 2021).

refined cobalt was in the battery industry, accounting for 63% of the industrial total [25]. Cobalt also has other industrial applications, for instance, as catalysts or drying agents in glass ceramics and high-temperature alloy production. The consumption of refined cobalt in battery materials will increase to 84.4% by 2021, indicating that cobalt is predominantly used for battery production in China, which further under-

scores the need for China to implement spent lithium-ion recovery. Fig. 3(b) illustrates cobalt consumption in the world and China from 2017 to 2021 [26]. As society advances, worldwide demand for cobalt steadily rises. Despite the impact of COVID-19 on China's cobalt industry, its consumption has remained relatively stable, solidifying its position as the world's largest producer of refined cobalt.

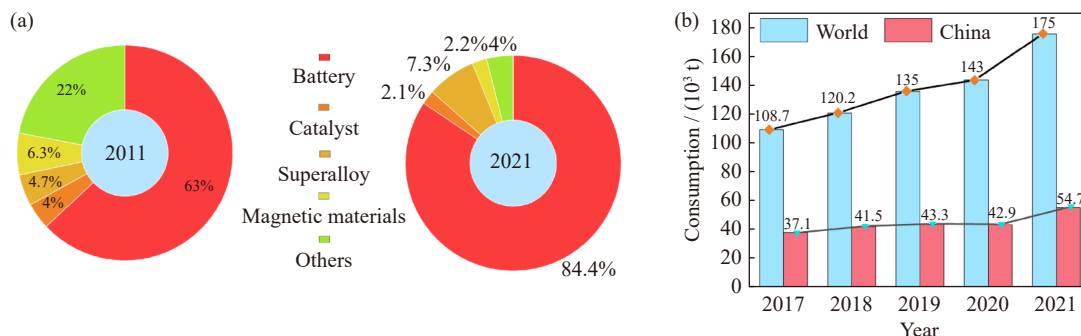


Fig. 3. (a) Consumption structure of refined cobalt in China: 2011 and 2021; (b) consumption tendency of cobalt from 2017 to 2021 in worldwide and China.

### 2.3. Cobalt-containing secondary resources

With the changing consumption structure in China, new challenges have arisen in the recycling of secondary cobalt resources, and cobalt recovery from battery materials has become increasingly prominent. Compared to alloy and catalyst materials, spent lithium-ion batteries possess a more complex structure and composition, necessitating an improvement in waste battery recovery processes to guide other waste cobalt products toward alleviating resource shortages and environmental pressures. Because of the higher energy density and improved stability of cobalt salts or cobalt-containing materials, batteries incorporating cobalt exhibit enhanced charging and discharging capabilities and heightened safety measures. Consequently, adding cobalt for electrode material modification is a fundamental driver behind the es-

calating demand for this element within the battery industry. However, electrode materials, such as lithium cobalt oxide ( $\text{LiCoO}_2$ ), nickel-cobalt multiple oxides ( $\text{Li}(\text{Ni}_x\text{Co}_y\text{Mn}_{1-x-y})\text{O}_2$ ) [27], and lithium iron phosphorus oxide ( $\text{LiFePO}_4$ ), inevitably experience structural collapse after a certain number of cycles [28]. After benefiting from the convenience of batteries, humans have recently faced the challenge of disposing of spent lithium-ion batteries. The total amount of power battery waste increased to 200000 tons in 2020 and is expected to reach 750000 tons by 2025 [29]. The metal resources contained within such a vast quantity of waste severely damage soil and water [30]. Nevertheless, it also represents a valuable asset that can be effectively utilized toward achieving sustainable resource development goals. This fact is particularly relevant for China as a major consumer of cobalt that heavily relies on imports of cobalt minerals. The recovery of

cobalt-containing secondary resources could provide a vital source of raw materials, ensuring security guarantees for Chinese cobalt industry development.

The idea of recycling secondary resources was introduced earlier, with countries such as the United States, Belgium, and Russia establishing production lines to produce metal cobalt, oxides, and chlorides from cobalt-bearing slag and secondary materials in the last century. However, the recovery technology developed much later, and renewable cobalt technology has slowly developed in China. In 2011, only 2500 tons of China's cobalt supply, merely 10% of the total supply, were recycled through regeneration technology [31]. Recently, domestic cobalt resources have been nearly depleted, substantially increasing import costs from \$1400 per ton (in 2016) to \$2100 per ton (in 2020). Simultaneously, the issue of environmental pollution caused by waste batteries has escalated. Research on related recycling processes and technologies has intensified to sustain the development of the cobalt industry and safeguard ecological balance. In 2021, more than 20% of the total output (25000 tons) was derived from secondary resource recovery by domestic companies.

### 3. Smelting of cobalt-containing minerals

Cobalt extraction from ores primarily relies on the smelting process of the main metal, with cobalt recycled through

by-products. Table 2 lists several secondary resources that can be utilized for cobalt recovery, revealing a relatively substantial content of cobalt in various ores and resources [32–47]. For instance, Jinchuan Company [35] and Huayou Cobalt Company [48] have established production departments to recover cobalt from ores to improve their economic benefits while producing nickel and copper products.

The cobalt extraction methods from mining resources such as copper–cobalt deposits and nickel–cobalt deposits encompass pyrometallurgy, hydrometallurgy, and pyro-hydrometallurgical processes. The pyrometallurgical process involves incorporating a portion of the cobalt into the slag phase. Consequently, the slag contains a certain amount of cobalt recoverable through enrichment for subsequent use. The data in rows 11–15 of Table 2 demonstrate that during the smelting process of nickel sulfide concentrate, the cobalt content exceeds 2wt% in the soot, smelting slag, and converter-blowing slag produced by the flash smelting process. Particularly noteworthy is that the amount of cobalt surpasses 7wt% in converter slag. Thus, nickel metallurgy enterprises, such as Norilsk Nickel, Otokunpu, and Jinchuan Company, have established dedicated workshops for cobalt recovery from slag. According to the principle of pyrometallurgy, the key step in cobalt recovery from waste involves introducing coke powder and calcine into cobalt slag, followed by reduction and vulcanization processes conducted in a dilu-

**Table 2.** Summary of the metallic element contents in the processing of copper–cobalt deposits (Nos. 1–8), nickel–cobalt sulfide deposits (Nos. 9–17), and some secondary resources (Nos. 18–21)

No.	Raw material	Element content / wt% (excluding those marked with g·L <sup>-1</sup> )								Refs.
		Co	Ni	Cu	Fe	S	Mg	Zn	Cd	
1	Cu–Co concentrate	2.20	0.30	2.50	4.00	0.05	4.82	—	—	[32]
2	Copper–cobalt ore	0.65	0.007	1.60	4.36	—	4.53	0.01	0.008	
3	Reduction leachate	2.05 g·L <sup>-1</sup>	0.018 g·L <sup>-1</sup>	4.94 g·L <sup>-1</sup>	1.80 g·L <sup>-1</sup>	—	0.39 g·L <sup>-1</sup>	0.04 g·L <sup>-1</sup>	0.002 g·L <sup>-1</sup>	
4	Chemical cleaning liquid	24.56 g·L <sup>-1</sup>	0.21 g·L <sup>-1</sup>	0.04 g·L <sup>-1</sup>	0.005 g·L <sup>-1</sup>	—	0.005 g·L <sup>-1</sup>	0.49 g·L <sup>-1</sup>	0.01 g·L <sup>-1</sup>	[33]
5	Extract liquid	24.53 g·L <sup>-1</sup>	0.21 g·L <sup>-1</sup>	0.002 g·L <sup>-1</sup>	0.002 g·L <sup>-1</sup>	—	0.002 g·L <sup>-1</sup>	0.002 g·L <sup>-1</sup>	0.001 g·L <sup>-1</sup>	
6	Cobalt chloride solution	83.2 g·L <sup>-1</sup>	0.004 g·L <sup>-1</sup>	<0.002 g·L <sup>-1</sup>	0.002 g·L <sup>-1</sup>	—	0.002 g·L <sup>-1</sup>	<0.002 g·L <sup>-1</sup>	—	
7	Cu–Co smelting slag	0.70	—	1.40	38.73	0.51	0.41	3.25	—	[34]
8	Converter blows slag	7.50	30.0	0.50	6.00	—	—	—	—	[35]
9	Electrolytic slag	8–10	28–32	0.4–0.5	5–6	—	—	—	—	[36]
10	Nickel sulfide ore	0.11–0.13	5–6	2.5–3.0	31–33	24–26	7.0–9.0	—	—	[37]
11	Cu–Ni concentrate	0.25	12.0	42.0	30.0	—	—	—	—	[38]
12	Middle nickel matte	0.62	22.0	19.7	26.0	27.0	—	—	—	[39]
13	Flash smelting slag	0.2–0.3	0.7–1.2	0.50	43.0	1.40	8–10	—	—	[40]
14	Flash furnace soot	6.74	2.76	0.21	40.93	1.54	5.30	—	—	[41]
15	High nickel matte	30.67	0.293	43.33	1.50	16.50	—	—	—	[39]
16	Electrolytic purification slag	3.33	25.52	0.48	7.68	23.78 (SO <sub>4</sub> <sup>2-</sup> )	—	0.051	—	[42]
17	Zinc smelting slag	6.79	—	—	1.88	—	—	—	1.30	[43]
18	Magnetic materials	36.78	—	3.92	9.45	—	—	—	—	[44]
19	Spent catalyst	50.30	0.064	0.002	0.105	—	0.005	—	—	[45]
20	Anode materials	40.19	0.06	0.012	—	—	0.012	0.025	—	[46]
21	Spent cobalt aluminum coating	41.89	0.060	0.012	0.063	—	0.012	0.025	—	[43,47]

Note: “—” represents a lack of data. The real amount of MgO presents the Mg contents.

tion electric furnace. Subsequently, the resulting cobalt matte further undergoes blowing in a converter to obtain a cobalt-rich matte [49]. Despite their simplicity and ease of operation, pyrometallurgical recovery processes suffer from limitations such as insufficient processing capacity, considerable cobalt losses, and inadequate cobalt matte metallization. In contrast, hydrometallurgical processes focus on the leaching, multistage extraction separation, and purification of cobalt oxide minerals. Although these processes have accumulated considerable experience in achieving a high metal recovery rate through leaching, they are characterized by substantial acid consumption and variable difficulties in removing impurities. The pressurized ammonia leaching method has been developed for cobalt ore recovery. This method is based on the principle that  $[\text{Co}(\text{NH}_3)_n]^{3+}$  exhibits excellent stability and high solubility under high oxygen pressure conditions, effectively avoiding excessive impurities in solution and reducing consumption of acidic and alkaline reagents. The combined process of pyrometallurgy and hydrometallurgy is widely employed in cobalt metallurgy because of its strong adaptability toward various raw materials. This process can effectively treat cobalt–sulfur concentrate, arsenic–cobalt ore, cobalt-bearing alloy waste, and other materials. The subsequent sections will focus on the advancements in cobalt smelting technology using copper–cobalt deposits, nickel–cobalt deposits, and zinc plant residue as case studies.

### 3.1. Recovery of cobalt from copper–cobalt ore

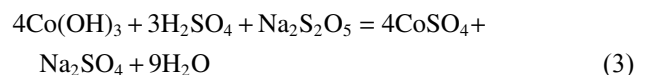
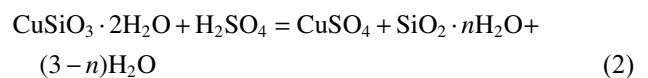
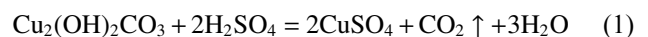
The Katanga region in the DRC is renowned for its abundant copper and cobalt resources, establishing itself as the world's largest producer of cobalt minerals [50]. Copper–cobalt deposits in the DRC are present in oxide and sulfide forms, with oxides occurring near the surface at a depth of 120 m [51]. However, sulfides occur deeper underground, posing considerable extraction challenges. This section presents a comprehensive overview of recovery methods for copper–cobalt deposits based on mineral types and their respective recovery statuses.

#### 3.1.1. High-grade copper–cobalt oxide ore

The pyrometallurgical process for copper–cobalt oxide using a blast furnace or electrolytic furnace is inefficient, primarily because of the raw export policy [52]. Hydrometallurgical processes, which involve mineral leaching, impurity separation, and cobalt precipitation, yielding cobalt oxide and pure cobalt solution as main products, are mainly employed for copper–cobalt oxide with variations depending on ore types and grades.

The high-grade Cu–Co oxide ore finds extensive applications in various industries and can be extracted through leaching, followed by iron removal purification, cobalt ion precipitation, and extraction. Preleaching crushing substantially enhances the leaching efficiency of valuable metallic elements. Zhang [53] determined that the optimal crushing method for oxidized ore, slime-bearing ore, and viscous ore was semi-self-grinding. High-grade oxidized ores are leachable using reduction leaching or selective leaching methods.

However, the reduction leaching method is more widely employed because of its shorter process and simpler operation. Generally, reductants include sodium metabisulfite [54], sodium sulfite [33], sulfur dioxide [55], and ferrous sulfate [56]. The toxicity of sulfur dioxide and the introduction of  $\text{Fe}^{2+}$  through ferrous sulfate pose challenges to cobalt purification, making sodium metabisulfite and sodium sulfite the preferred reductants. Xie [57] investigated the reduction leaching of cobalt-rich hard manganese ore with sodium metabisulfite ( $\text{Na}_2\text{S}_2\text{O}_5$ ), and the primary reactions in the leaching process are presented in Eqs. (1)–(3). The solution reduces high-valence cobalt while dissolving copper and iron. Under the conditions of pulp concentration 33%, sulfuric acid solution pH range from 1.5 to 1.8 and sodium metabisulfite dosage 8 kg/t, the leaching rate of cobalt reached 90.38% after 5 h of stirring leaching. The leaching solution contains various impurity metal ions such as zinc, manganese, aluminum, magnesium, and calcium, necessitating subsequent extraction and chemical impurity removal processes to obtain a qualified cobalt solution.



#### 3.1.2. Low-grade copper–cobalt oxide ore

Copper-rich cobalt deposits in Katanga have been exploited to provide high-quality raw materials for copper and cobalt smelting worldwide. However, the shallow surface deposits of copper-rich cobalt are nearly depleted, making the recovery of copper and cobalt from low-grade oxidized or sulfurized ores a crucial direction for future development in this industry. The low-grade copper–cobalt ore exhibits many elements and complex phases, as demonstrated in Table 3. In this context, the content of valuable metallic elements is relatively low, with cobalt typically not exceeding 1wt% and copper being less than 10wt%. Therefore, the leaching system of low-grade ores presents several challenges, such as a low leaching rate, high acid consumption, and extensive equipment requirements. Addressing these issues means enhancing the copper and cobalt grade before leaching through mineral separation. Fig. 4(a) illustrates the recovery process for low-grade copper–cobalt ore. On the basis of the characteristics of copper–cobalt ore, Ou [58] developed a process that involves the first flotation of copper–cobalt sulfide followed by the flotation of Cu–Co oxide without desliming. This process adopts the combined processing technology for beneficiation, smelting, and closed-circuit test processes for micro-fine middles and raw ores, respectively. The result was sulfide ore concentrate with a copper grade of 31.52wt% and a recovery rate of 33.25%, as well as oxidized ore concentrates with a copper grade of 23.76wt% and a recovery rate of 47.14%. Additionally, the study explored the process involving sodium sulfite reduction, acid leaching, copper ex-

**Table 3. Main phases in low-grade copper–cobalt ore**

Types	Mineral phase	Molecular formula
Oxide	Malachite	$\text{Cu}_2(\text{OH})_2\text{CO}_3$
	Chrysocolla	$(\text{Cu}, \text{Al})_2\text{H}_2\text{Si}_2\text{O}_5(\text{OH})_4 \cdot n\text{H}_2\text{O}$
	Cuprite	$\text{Cu}_2\text{O}$
	Cobaltite	$\text{CoO}(\text{OH})$
	Cobalt copper ore	—
	Copper–cobalt-rich psilomelane	$(\text{Li}, \text{Al}, \text{Cu}, \text{Co})\text{MnO}_2(\text{OH})$
	Cobalt-bearing dolomite	—
	Cobalt malachite	—
	Rutile	$\text{TiO}_2$
	Hematite	$\text{Fe}_2\text{O}_3$
	Quartz	$\text{SiO}_2$
Sulfide	Chalcocite	$\text{Cu}_2\text{S}$
	Chalcopyrite	$\text{CuFeS}_2$
	Bornite	$\text{Cu}_5\text{FeS}_4$
	Covellite	$\text{CuS}$
Carbonate	Dolomite	$\text{CaMg}(\text{CO}_3)_2$
	Calcite	$\text{CaCO}_3$

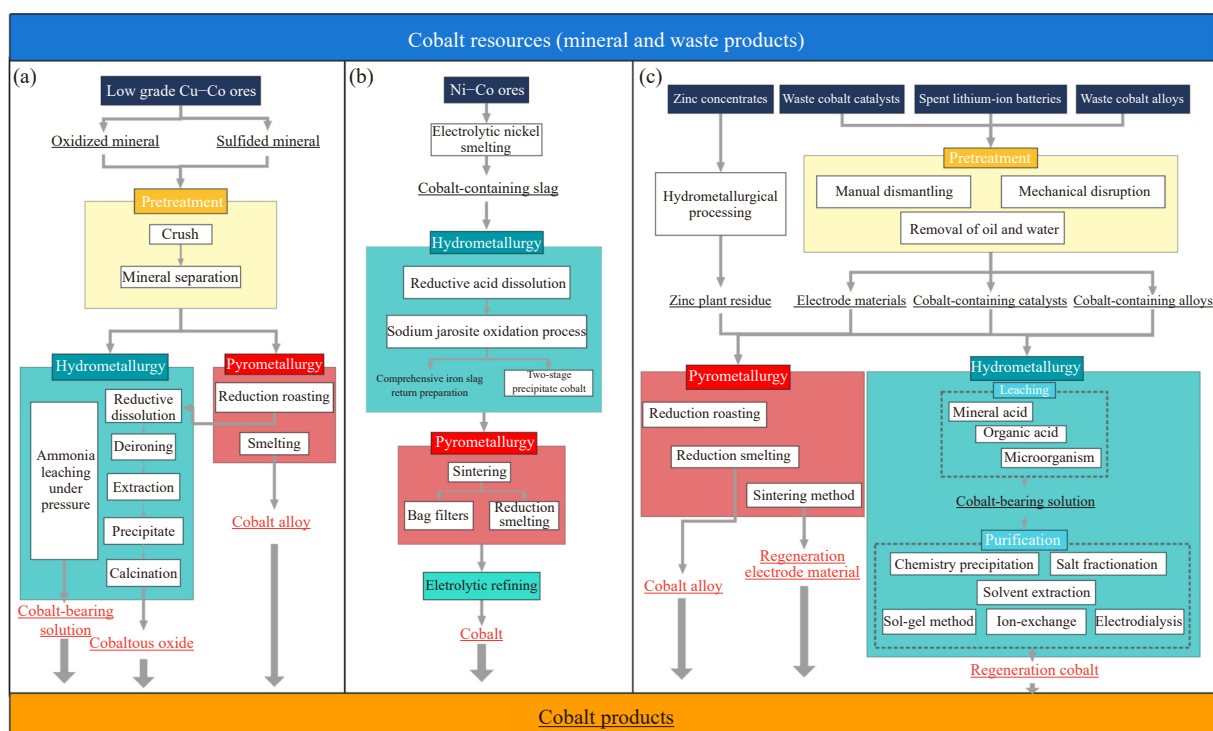
traction, and electrodeposition, neutralization removal of iron, cobalt precipitation and purification, and deep solution purification [33], as shown in Table 2 (Nos. 4–8). Finally, pure cobalt oxide was obtained with recovery rate for cobalt and copper at 85.10% and 89.78%, respectively. Liu *et al.* [59] modified the acid leaching process by incorporating sodium sulfite and investigated the impact of the activity, leaching time, and cobalt precipitation agent dosage on the cobalt precipitation ratio. Under optimal conditions, copper and cobalt achieved leaching ratios of 99.06% and 98.87%,

respectively. Additionally, they proposed a purification process involving “M5640 copper extraction, iron removal through sodium jarosite treatment, aluminum removal using sodium carbonate, and calcium and magnesium removal via sodium fluoride treatment, followed by evaporation crystallization for cobalt product preparation”. Furthermore, the use of flotation and grinding equipment for low-grade copper–cobalt ore [53], as well as the application of microbial leaching technology for mixed ores containing Cu–Co oxide and Cu–Co sulfide [60], has gained considerable traction in the research on Cu–Co ore.

3.1.3. Copper–cobalt sulfide ore

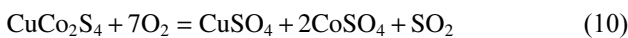
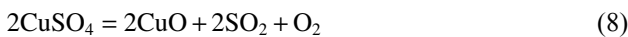
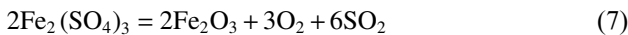
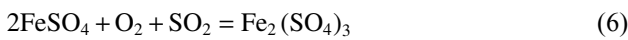
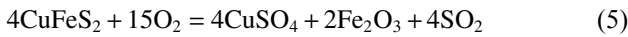
(1) Sulfide roasting–leaching process.

The current treatments for sulfide copper–cobalt ore are diverse, with the predominant method being the sulfide roasting of Cu–Co sulfide concentrate. The reaction equations for this process are Eqs. (4)–(10). The iron sulfide in the Cu–Co sulfide minerals undergoes selective oxidation to form hematite as copper and cobalt convert into sulfate. According to their differing solubilities, the copper and cobalt sulfates can be separated from hematite through water leaching. Yu *et al.* [61] investigated the impact of roasting temperature and duration, as well as the two-stage leaching process, on the leaching efficiencies of copper and cobalt. The results revealed that after roasting at 620°C for 5 h, the leaching efficiencies of copper and cobalt in the first stage of low acid leaching were 93.81% and 85.5%, respectively. The content of copper and cobalt in the first stage of leaching slag were 1.75wt% and 1.52wt%, respectively. Subsequently, the leaching slag underwent a second stage with an initial acid concentration of 100 g·L<sup>-1</sup>. At this stage, copper and cobalt had total leaching efficiencies reaching 98.03% and 91.93%, respectively,



**Fig. 4. Schematic of recovery steps of major cobalt resources: (a) copper–cobalt ores; (b) nickel–cobalt sulfide ores; (c) zinc plant residue and waste cobalt products.**

while their concentrations remained below 1%. The relationship between the standard enthalpy of reaction and temperature in the sulfated roasting process was analyzed by Sun *et al.* [62] from a thermodynamic perspective. Notably, precise roasting temperature control is crucial for obtaining  $\text{CuSO}_4$  and  $\text{Fe}_2\text{O}_3$ .



#### (2) Pressure leaching process.

The pressure leaching process is used to extract nonferrous metals such as uranium, tungsten, molybdenum, vanadium, and titanium [63]. In the 1950s, Americans achieved industrial production of pressure leaching for cobalt, nickel, and copper concentrate treatment. Subsequently, this technology matured and became widely used in smelting Ni–Co concentrate, Cu–Ni matte, and zinc concentrate. Compared to other methods, pressure leaching can conveniently produce elemental sulfur from sulfide ore, facilitating transportation and storage. Additionally, it operates at higher temperature with better kinetic conditions, resulting in faster reaction rates while reducing environmental pollution during metallurgical processes without roasting. Lan [64] and Li *et al.* [65] utilized a pressure leaching process to treat Cu–Co sulfide concentrate, wherein an oxygen pressure leaching reaction occurred in a pressurized kettle under oxidizing conditions [66]. At a reaction temperature of 180°C, a reaction time of 2 h, a liquid/solid (L/S) ratio of 5, and an oxygen partial pressure of 600 kPa, the leaching ratios for cobalt and copper were above 98.5% and 99%, respectively, higher than those achieved through sulfated roasting processes (98.03% for copper and 91.93% for cobalt), indicating that pressure leaching is more effective in extracting copper and cobalt from concentrate. However, the substantial fluctuation in sulfur content observed in Cu–Co sulfide concentrate during practical applications can profoundly impact the acidity of the terminus of the oxygen pressure leaching process, resulting in unstable hydrolysis of  $\text{Fe}^{3+}$  to  $\text{Fe}_2\text{O}_3$ . Therefore, Li *et al.* [67] studied the behavior of iron and manganese during the pressurized leaching of Cu–Co sulfide ore, finding that the precipitation ratio of iron decreases with increasing acidity at the terminus, meanwhile, the slag contains less than 0.1wt% manganese, and no precipitation occurs. Changes in initial ferric acid concentration lead to precipitation of hematite, basic ferric sulfate, and hydrate.

#### (3) Pyrometallurgical process.

The pyrometallurgical process, a traditional step in copper smelting, can also be utilized for Cu–Co sulfide ore. Smelt-

ing primarily aims to recover copper, resulting in cobalt enrichment as an impurity element in converter slag, approximately 70% of the cobalt from the ore will enter into converter slag. Blowing effectively reduces crude copper's impurity content, while cobalt is abundant in the blowing slag and can serve as a raw material for cobalt recovery [68–69]. However, because the high iron-to-cobalt mass ratio in the blown slag is greater than 10, their separation and effective recovery pose considerable challenges, limiting the availability of reports on cobalt product preparation from Cu–Co sulfide ore through roasting. Ye *et al.* [70] and Tang *et al.* [34] started from the traditional Cu–Co sulfide ore recovery process and proposed the oxidation–reduction blowing process of matte according to the principle of the affinity of oxygen and sulfur to different metals. They controlled the distribution of cobalt elements in matte and slag phases by changing the conditions of oxidation and reduction smelting to improve cobalt recovery and obtain a cobalt-rich alloy. And the distribution ratio of cobalt in matte and slag can be enhanced by reducing temperature and promoting oxidation, facilitating higher enrichment in low-iron cobalt matte. Moreover, the lower temperature and higher iron in the matte contribute to increased enrichment and recovery of cobalt during the reducing–matting smelting process.

### 3.2. Recovery of cobalt from nickel–cobalt ore

Global nickel reserves amount to approximately 470 million tons and are primarily categorized into sulfide and laterite ores [71–72], whose products have found extensive applications in the chemical industry, metallurgy, battery production, and construction due to their exceptional stability, mechanical strength, and ductility. As discussed in Section 2 regarding cobalt ore reserves, nickel sulfide concentrate and limonite laterite–nickel ore contain approximately 3wt% and approximately 1wt% cobalt, respectively [73]. High-grade cobalt by-products generated during nickel metallurgy processes can serve as a recycled resource for efficiently addressing the issue of depleted cobalt resources.

#### 3.2.1. Nickel sulfide ore

Nickel sulfide ore accounts for 30% of the total reserves, yet it yields approximately 60%–70% of the extracted nickel. To recover cobalt from nickel sulfide ore, Jinchuan Nickel Production Company has established a cobalt production line with an annual output capacity of 15000 tons, designed to acquire cobalt-containing slag generated during nickel smelting [74]. The electro-nickel products were produced through a combination of pyrometallurgical and electrolytic refining processes, as depicted in Fig. 5. Moreover, the valuable cobalt-containing slag generated during anolyte purification can be directly recovered as metallic cobalt or processed into various cobalt products within the dedicated workshop.

The currently marketed cobalt products primarily include electric cobalt, cobalt oxide, cobalt powder, and various cobalt salts. Fig. 4 shows the preparation of electrolytic cobalt, an efficient process to produce pure cobalt metal. The cobalt content in nickel electrolyte-derived cobalt slag typically

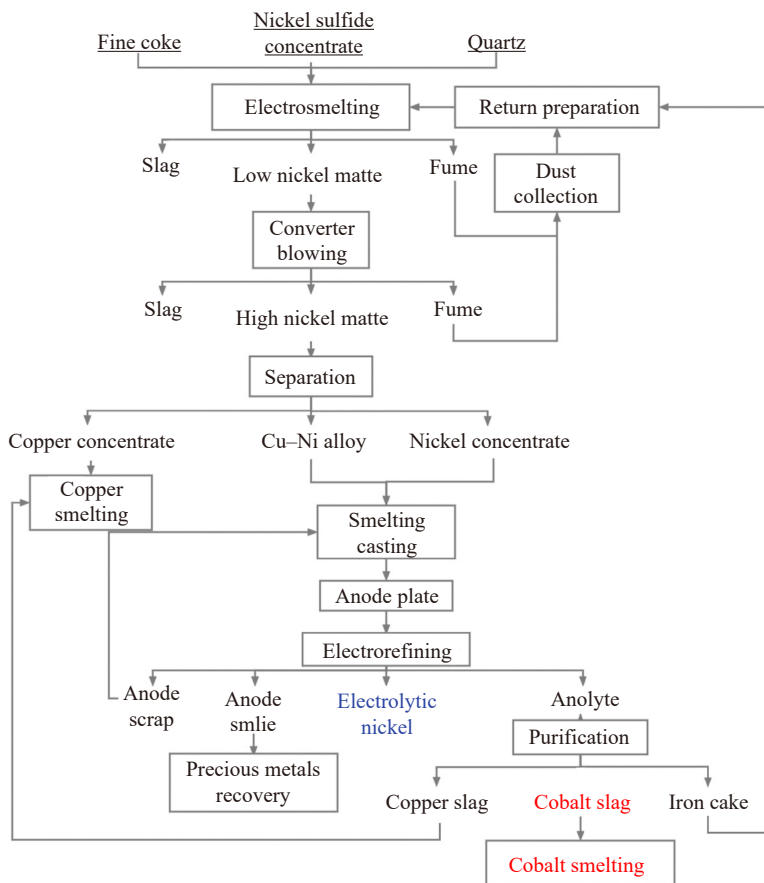


Fig. 5. Production of electro-nickel products and cobalt-containing slag formation.

ranges from 8wt% to 10wt%. Slurry reduction acid dissolution is achieved using sodium sulfite and nickel electrolytic anodic solution, while potassium ferricyanide is employed to indicate the completion of reduction dissolution. Because  $Fe^{3+}$  ( $E^\ominus = -0.440$  V) has a smaller potential than  $Co^{2+}$  ( $E^\ominus = -0.277$  V) and  $Ni^{2+}$  ( $E^\ominus = -0.250$  V), a substantial amount of  $Na^+$ ,  $SO_4^{2-}$ , and  $Fe^{3+}$  ( $16\text{ g}\cdot\text{L}^{-1}$ ) dissolved in solution under strict control of the precipitation temperature at  $65\text{--}75^\circ\text{C}$  and the pH at  $1.5\text{--}2$  [75]. This result caused sodium jarosite precipitation with excellent filtration performance. Table 4 displays the elemental composition of the solution before and after iron removal, indicating a reduction in iron ion content to below  $0.005\text{ g}\cdot\text{L}^{-1}$ . Subsequent multistage cobalt sedimentation, sintering, and reduction melting processes result in crude cobalt with a high-purity range of  $96\%\text{--}98\%$  [76]. Electrolytic refining is commonly employed to achieve high-purity products. The migration behaviors of cations and anions on the electrodes in the electrolytic cell are as follows: cobalt exhibits an anodic polarization potential of  $0.1$  V, while metals with negative potentials such as Fe ( $E^\ominus = -0.440$  V), Mn ( $E^\ominus = -1.029$  V), Ni ( $E^\ominus = -0.250$  V), Pb ( $E^\ominus = -0.126$  V), and Zn ( $E^\ominus = -0.763$  V) also dissolve sim-

ultaneously on the anode plate. Notably, the standard potential of Cu is  $+0.337$  V, and the formation of Cu–Co solid solutions can lead to its dissolution and eventual entry into the anode slime. Although Co has a standard potential of  $-0.277$  V at the cathode, its polarization potential falls below  $-0.3$  V under electrochemical polarization. Therefore, impurities such as Cu, Ni, Pb, Zn, and Fe should deposit on the cathode after the depolarization. Diaphragm electrolysis was performed using an electrolyte of extremely high purity to achieve super-pure cobalt at the cathode. Table 5 shows the electrolyte element content and technical indexes for electrolytic cobalt production.

### 3.2.2. Laterite ore

With industrial growth, nickel sulfide ore consumption has continuously increased while resource reserves have dwindled. As of 2020, nickel laterite accounts for  $60.14\%$  of the world’s output of nickel resources [77]. Nickel laterite, an oxide ore, is formed through long-term weathering and leaching metamorphism of olivine-containing nickel. It can be classified into two types based on chemical composition: silicon magnesium nickel ore and limonite with higher iron and cobalt contents. The latter type accounts for  $70\%$  of the

Table 4. Element content in solutions before and after removal by sodium jarosite

Process	Co	Ni	Fe	Cu	Mn	Zn	Pb	Na	Si	$Cl^-$	$SO_4^{2-}$	pH
Before	19	80	16	0.4	0.09	0.03	0.004	17.5	0.33	8.34	250	1.5–2
After	19	80	0.005	0.35	0.06	0.02	0.002	25	0.25	15	220	3–3.3

**Table 5. Element content of cobalt electrolyte and technical indexes**

Element content / (g·L <sup>-1</sup> )						Technical index				
Co	Fe	Cu	Pb	Zn	Ni	Current density / (A·m <sup>-2</sup> )	Voltage / V	Temperature / °C	d / mm	Δh / mm
100–120	0.0008	0.0003	0.0001	0.005	0.03	350–500	3–5	55–66	180	30–50

Notes: *d* is the electrode spacing, and  $\Delta h$  is the liquid level.

laterite mineral resources' nickel reserves.

The enrichment and recovery of cobalt in limonitic ore occurs within the hydrometallurgy process, including high-pressure acid leaching (HPAL), atmospheric pressure acid leaching (APAL), and reduction roasting-ammonia leaching (RRAL). These methods yield cobalt-containing leachate, and extensive research has enhanced the leaching and separation of cobalt [78]. HPAL is a competitive method for cobalt recovery from limonitic ore because of the acceleration of chemical reaction rates at high temperature and pressure, resulting in over 95% cobalt recovery [79], which requires an elevated temperature (240–270°C) and pressure (3000–5000 kPa) [80–81]. Guo *et al.* [82] investigated the leaching behavior of cobalt from nickeliferous limonitic laterite ore by the HPAL process. The effects of various parameters, including sulfuric acid added, leaching time and temperature, and L/S ratio, on cobalt leaching behavior showed more than 96wt% Co extracted under optimum conditions. The commercial HPAL operations include Murrin, Ravensthorpe, Ambatovy, Ramu River, and Coral Bay [83]. The APAL process is typically conducted under atmospheric pressure (101,325 Pa) at 50–110°C [84], resulting in lower capital and operational costs compared to HPAL [85–86]. Li *et al.* [87] used phosphoric acid for the selective leaching of cobalt from limonitic laterite. The results indicate that 89.8wt% of cobalt was successfully extracted from calcined limonitic laterite, and the resulting FePO<sub>4</sub> precipitate exhibited a purity level of 94%, which should provide additional economic benefits. The RRAL (Caron) process is a well-proven technology, wherein limonitic ore undergoes initial roasting at 850°C to selectively reduce cobalt [88]. Subsequently, metal cobalt leaches by ammonia/ammonia carbonate solution [89], finally recovering cobalt sulfide. The RRAL method is unique because it can recycle ammonia, lowering reagent costs. Additionally, alternative methods such as heap leaching, nitric acid leaching, hydrochloric acid leaching, and bio-leaching have also been explored for cobalt recovery [78,90–94].

The efficient separation of metal elements in cobalt-containing leachate represents the ultimate objective of cobalt resource recovery, and various corresponding technologies have undergone development. For instance, solvents such as hydroxy-oxime, phosphinic acid, D2EPHA (phosphoric acid, Weakacid IX resin), and trialkyl amine have been utilized for cobalt ion extraction, leading to considerable advancements in this field [83,95]. Simultaneously, precipitation [96–97] and ion exchange [98–99] processes are continuously undergoing optimization to achieve higher efficiency in the separation and recovery of cobalt.

### 3.3. Recovery of cobalt from zinc plant residue

The hydrometallurgical process of sphalerite serves as the

primary zinc production technology, constituting over 80% of China's total output [100–102]. Typically, a cobalt enrichment level ranging from 0.9% to 1.2% can be achieved in Zn–Co leaching residue. After purification, the cobalt content can be increased to 6wt%–12wt%. Therefore, separating and recovering cobalt from zinc–cobalt slag avoids pollution and improves the utilization rate of cobalt resources to alleviate its scarcity. Comprehensive studies have been conducted on the selective leaching of zinc and cobalt to achieve effective separation and recovery of metal resources from zinc–cobalt slag. Wang *et al.* [103] investigated the citric acid in valuable metals from purification residue. The results demonstrated a high leaching rate of 97.64%. Huang *et al.* [104] proposed an alkaline glycine solution to separate zinc and cobalt, which selectively dissolves 93.81% of zinc from the slag, leaving cobalt in the residue. Moreover, various leachates, such as sulfuric acid, hydrochloric acid, nitric acid, glycine, antiphrasis, oxalic acid, aspartic acid, Na<sub>2</sub>CO<sub>3</sub>, NaOH, and ammonium also obtained important results [105–107].

The separation and recovery of cobalt-containing leaching solutions primarily involve oxidation precipitation and solvent extraction. In the former process, Co<sup>2+</sup> in the leaching solution is oxidized to Co<sup>3+</sup> by oxidizing agents [108–110], followed by hydrolysis of Co<sup>3+</sup> to generate precipitation by adjusting the pH (Fig. 6(a)). Washing, selective leaching, purifying, and precipitating oxidized cobalt produces a cobalt-rich hydroxide precipitate with an assay of (45 ± 4)% Co at a (61 ± 14)% Co recovery. The critical step is controlling the oxidation conditions during the selective precipitation process [101]. The application of solvent extraction is extensively researched to separate zinc and cobalt. Fig. 6(b) illustrates the extraction sequence of P204 for various metal ions [111]. Controlling the pH of the water phase is necessary during the extraction process to ensure efficient separation of the target metal ions [112]. For example, the diluent used by Kongolo *et al.* [113] was kerosene, and a 20% P204 extractant was employed for the simultaneous extraction and recovery of cobalt and zinc. A two-stage extraction process separated zinc and cobalt at pH = 2.5 and 1.5, respectively.

## 4. Recovery of cobalt from secondary resources

With the advancement of traditional industries and the emergence of new industries [114], the recycling of cobalt from waste materials such as alloy catalysts and batteries cannot be overlooked, alongside the decline in cobalt ores. To evaluate the industrial flow state of cobalt, the substance flow analysis (SFA) method [115–116] is employed to investigate the cobalt flow in China, as illustrated in Fig. 7 [24].

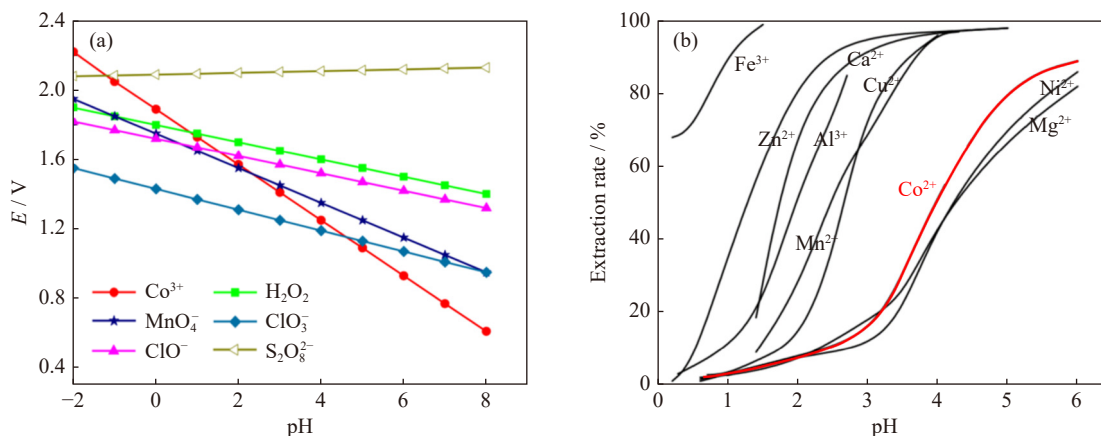


Fig. 6. (a) Relationship between the oxidation potential of an oxidizer and pH. Reprinted from *Hydrometallurgy*, 193, Y.K. Huang, Z.F. Zhang, Y.J. Cao, et al., Overview of cobalt resources and comprehensive analysis of cobalt recovery from zinc plant purification residue-A review, 105327, Copyright 2020, with permission from Elsevier; (b) sequence of metal extraction by P204 at different pH values [111].

Cobalt from natural sources is observed to enter the industry through mining. The cobalt content of cobalt-containing products is concluded in Table 2 (Nos. 18–21).

Generally, waste cobalt products exhibit a simple elemental composition and possess considerably higher cobalt content compared to minerals, enabling the use of mineral-based processes for cobalt recovery. Because the structure of spent lithium-ion batteries is the most complex, and the amount of scrap is the largest, prioritizing recycling is imperative. At the same time, the recovery of cobalt from waste alloys and catalysts used in traditional industries is also summarized.

#### 4.1. Recovery of cobalt from spent lithium-ion batteries

The cobalt recovery process in lithium-ion batteries comprises battery pretreatment and metallurgical purification of cobalt resources. Battery pretreatment releases residual en-

ergy from the battery and classifies it preliminarily for safety and optimal cobalt recovery efficiency. Because of the intricate nature of battery materials, various techniques, such as crushing and flotation [117–118], magnetic separation [119], and ultrasound, are employed together to enhance the separation process. Following battery pretreatment, the initially enriched cathode materials can undergo further purification through metallurgical methods for cobalt recovery. The metallurgical approaches utilized for cobalt recovery from electrode materials include pyrometallurgy, hydrometallurgy, and pyro-hydrometallurgy. Fig. 4(c) presents a schematic of the key steps.

##### 4.1.1. Pyrometallurgical processing for cobalt recovery in electrodes

The pyrometallurgical process for cobalt recovery from spent lithium-ion batteries involves separating cobalt from impurities at high temperature through reduction smelting

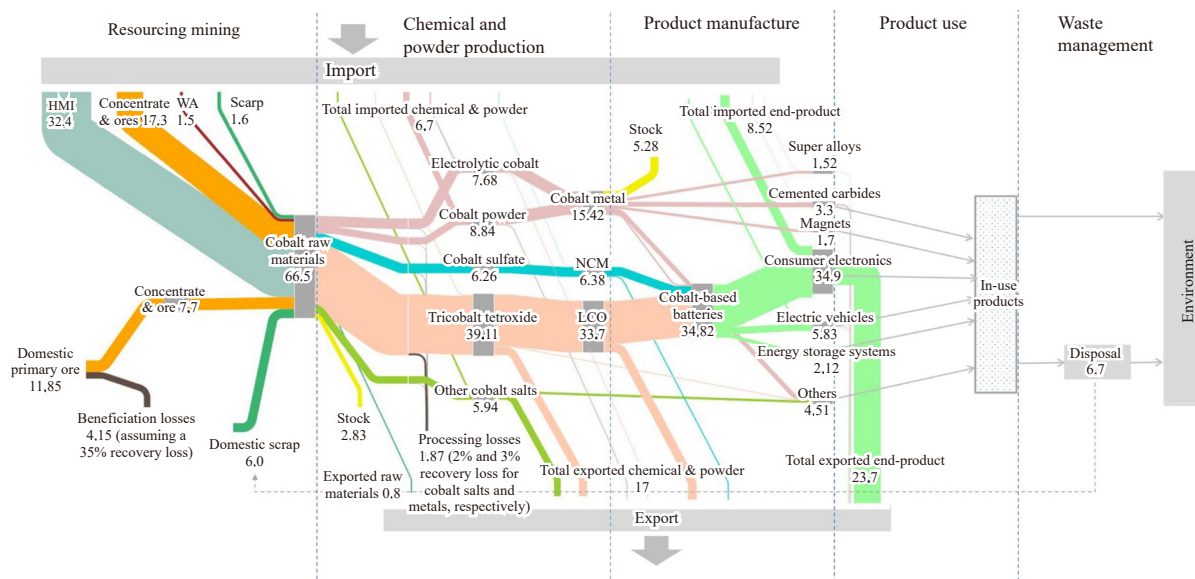


Fig. 7. Schematic of cobalt flows in China for 2015. Reprinted from *J. Clean. Prod.*, 275, Z.Y. Chen, L.G. Zhang, and Z.M. Xu, Analysis of cobalt flows in mainland China: Exploring the potential opportunities for improving resource efficiency and supply security, 122841, Copyright 2020, with permission from Elsevier.

and high-temperature sintering reduction roasting, producing cobalt-containing products.

#### (1) Reduction smelting process.

The reduction smelting process utilizes carbon, aluminum, and other reducing agents to reduce and melt electrode materials at high temperature to recover cobalt with a cobalt alloy. In industry, the reduction smelting process has a strong adaptability of raw materials, a simple process, and high efficiency, making it a promising candidate for continuous production in the future. The current production by reduction smelting has been established by Belgium Meimeike Company, Switzerland Batec Company, and Japan Mitsubishi Company [120]. However, there are limitations in the purity of the obtained product and environmental concerns regarding the release of harmful gases such as CO and CO<sub>2</sub> during recovery. Additionally, further improvement still requires substantial investments.

#### (2) High-temperature sintering process.

Because of the degradation of electrode materials and subsequent loss of metal ion activity, lithium-ion batteries are scrapped after a certain number of cycles [121–122]. Zhang

*et al.* [123] proposed an experimental approach for directly preparing electrode materials from discarded lithium-ion batteries, as depicted in Fig. 8(a). Initially, trifluoroacetic acid (TFA) was used to extract the positive electrode material from the aluminum foil. Subsequently, Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and other metal salts were added to the positive electrode material to adjust its stoichiometric ratio, followed by two-stage roasting to prepare Li(Ni<sub>1/3</sub>Co<sub>1/3</sub>Mn<sub>1/3</sub>)O<sub>2</sub>. The electrochemical analysis presented in Fig. 8(b)–(c) demonstrates that the material's initial charge–discharge capacities are 201 and 155.4 mAh·g<sup>-1</sup>, respectively, with a capacity retention rate of 83.01% after 30 cycles. Although sintering is a cost-effective and eco-friendly process, it poses challenges regarding process control and results in unstable electrochemical performance.

#### 4.1.2. Hydrometallurgical processing for cobalt recovery in electrodes

##### (1) Selective leaching process.

The hydrometallurgical recovery of cobalt from a spent battery typically involves dissolving the battery material and purifying the resulting solution to obtain a high-quality co-

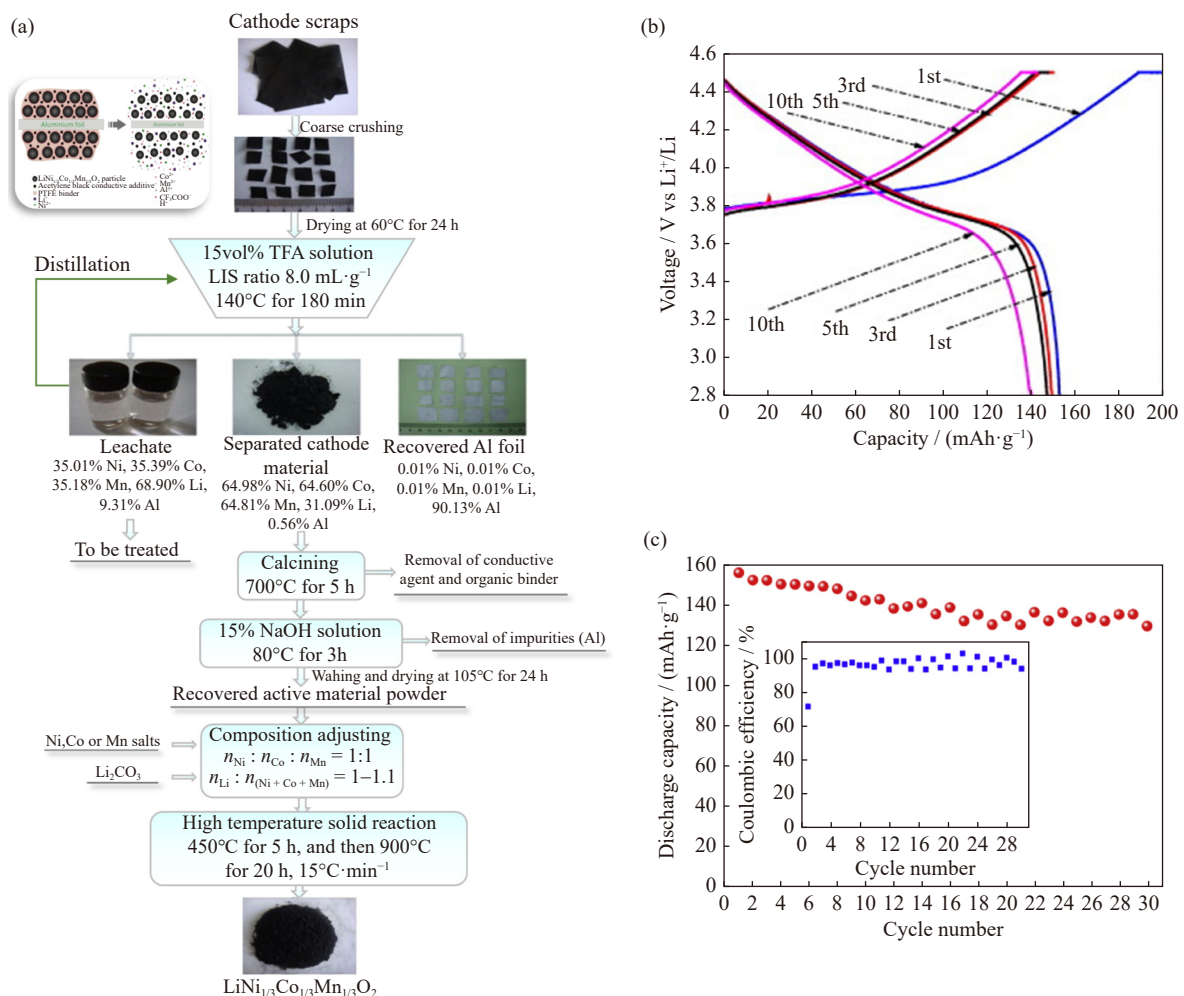


Fig. 8. (a) Experimental scheme for directly preparing electrode materials from spent lithium-ion batteries and (b–c) electrochemical performance test of regenerated electrode materials. Reprinted from *Waste Manage.*, 34, X.H. Zhang, Y.B. Xie, H.B. Cao, F. Nawaz, and Y. Zhang, A novel process for recycling and resynthesizing LiNi<sub>1/3</sub>Co<sub>1/3</sub>Mn<sub>1/3</sub>O<sub>2</sub> from the cathode scraps intended for lithium-ion batteries, 1715, Copyright 2014, with permission from Elsevier.

balt solution [124]. Various leaching systems and lixiviants have been developed, and the leaching rates of cobalt ions under different leaching agents and conditions are summarized in Table 6. The leaching system can be categorized as follows: inorganic acid systems, such as sulfuric acid, hydrochloric acid, and nitric acid; organic acid systems, such as malic acid, citric acid, and oxalic acid; and ammonia leaching systems. Table 6 illustrates the effective leaching of cobalt in cathode materials under mild experimental conditions with the addition of a reductant [56,62,125–149]. This effectiveness is due to the lower chemical bond energy between divalent cobalt and oxygen compared to trivalent cobalt and oxygen when a reductant is present in the leaching solution, which breaks the chemical bond between high-priced cobalt and oxygen [128–150]. Consequently,  $\text{Co}^{3+}$  can be reduced to

$\text{Co}^{2+}$  [126,133,151], making it readily soluble in acid and further improving the leaching rate of cobalt.

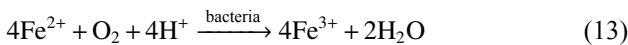
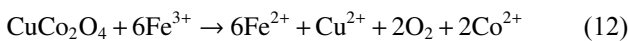
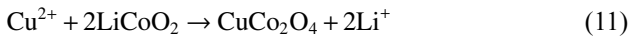
Moreover, the use of microorganisms in the leaching process of spent lithium-ion batteries has been studied. The microbial leaching approach enables selective cobalt extraction through direct action by specific microorganisms on cobalt within the batteries or indirect action via microorganisms and their metabolites. Zeng *et al.* [152] investigated the factors influencing the cobalt leaching ratio from waste  $\text{LiCoO}_2$  batteries by *Thiobacillus ferrooxidans* (A.f) and found that after 10 d, the cobalt leaching ratio was only 43.1%. However, when a small amount ( $0.75 \text{ g} \cdot \text{L}^{-1}$ ) of  $\text{Cu}^{2+}$  was present, cobalt in spent  $\text{LiCoO}_2$  cells was completely leached (99.9%) after only 6 d. On the basis of related studies [153–154], Zeng *et al.* proposed a  $\text{CuCo}_2\text{O}_4$  catalytic mechanism, and the reac-

**Table 6. Summary of the cobalt leaching ratio under different conditions**

No.	Leachates	Molecular formula	Raw materials	Reductant	Molecular formula	Optical leaching conditions		Co / wt%	Refs.
						Temperature / °C	Time / min		
1	Sulfuric	$\text{H}_2\text{SO}_4$	LNCM	—	—	90	180	68	[125]
2	Sulfuric	$\text{H}_2\text{SO}_4$	LCO	—	—	70	240	66.2	[126]
3	Sulfuric	$\text{H}_2\text{SO}_4$	LCO	Sodium Sulfite	$\text{NaHSO}_3$	70	240	91.6	[127]
4	Sulfuric	$\text{H}_2\text{SO}_4$	LCO	Hydrogen peroxide	$\text{H}_2\text{O}_2$	80	120	92.3	[127]
5	Sulfuric	$\text{H}_2\text{SO}_4$	LCO	Hydrogen peroxide and ultrasound	$\text{H}_2\text{O}_2$	30	30	94.63	[128]
6	Sulfuric	$\text{H}_2\text{SO}_4$	LNCM	Sodium pyrosulfite	$\text{Na}_2\text{S}_2\text{O}_5$	60	60	>90	[129]
7	Sulfuric	$\text{H}_2\text{SO}_4$	LNCM	Starch	$(\text{C}_6\text{H}_{10}\text{O}_5)_n$	80	120	96.73	[130]
8	Hydrochloric	$\text{HCl}$	Spent-LIBs	—	—	50	90	>99	[131]
9	Hydrochloric	$\text{HCl}$	LCO	Hydrogen peroxide	$\text{H}_2\text{O}_2$	80	90	99.4	[132]
10	Nitric	$\text{HNO}_3$	LCO	Hydrogen peroxide	$\text{H}_2\text{O}_2$	75	30	>95	[133]
11	Nitric	$\text{HNO}_3$	Spent-LIBs	Hydrogen peroxide	$\text{H}_2\text{O}_2$	90	30	98.01	[134]
12	Phosphoric	$\text{H}_3\text{PO}_4$	LCO	Hydrogen peroxide	$\text{H}_2\text{O}_2$	50	60	99	[135]
13	Malic	$\text{C}_4\text{H}_6\text{O}_5$	LCO	Hydrogen peroxide	$\text{H}_2\text{O}_2$	70	40	93	[136]
14	Malic	$\text{C}_4\text{H}_6\text{O}_5$	LNCM	Hydrogen peroxide	$\text{H}_2\text{O}_2$	70	20	98.06	[137]
15	Citric	$\text{C}_6\text{H}_8\text{O}_7$	LNCM	Hydrogen peroxide	$\text{H}_2\text{O}_2$	90	30	91	[136]
16	Citric	$\text{C}_6\text{H}_8\text{O}_7$	LCO	Hydrogen peroxide	$\text{H}_2\text{O}_2$	90	300	99.07	[138]
17	Citric	$\text{C}_6\text{H}_8\text{O}_7$	Spent-LIBs	Hydrogen peroxide	$\text{H}_2\text{O}_2$	80	90	95	[139]
18	Oxalic	$\text{H}_2\text{C}_2\text{O}_4$	LCO	—	—	95	150	97	[140]
19	Oxalic	$\text{H}_2\text{C}_2\text{O}_4$	LCO	Hydrogen peroxide	$\text{H}_2\text{O}_2$	80	120	>98	[62]
20	Ascorbic	$\text{C}_6\text{H}_8\text{O}_6$	LNCM	—	—	65	20	~100	[141]
21	Ascorbic	$\text{C}_6\text{H}_8\text{O}_6$	LCO	Ascorbic	$\text{C}_6\text{H}_8\text{O}_6$	70	20	94.8	[56]
22	Acetic	$\text{CH}_3\text{COOH}$	LNCM	Hydrogen peroxide	$\text{H}_2\text{O}_2$	60	60	93.62	[142]
23	Acetic	$\text{CH}_3\text{COOH}$	LNCM	Hydrogen peroxide	$\text{H}_2\text{O}_2$	50	30	~37	[143]
24	Succinic	$\text{C}_4\text{H}_6\text{O}_4$	LCO	Hydrogen peroxide	$\text{H}_2\text{O}_2$	70	40	~100	[144]
25	Tartaric	$\text{C}_4\text{H}_6\text{O}_6$	LNCM	Hydrogen peroxide	$\text{H}_2\text{O}_2$	70	30	98.64	[145]
26	Citric and ascorbic	$\text{C}_6\text{H}_8\text{O}_7$ and $\text{C}_6\text{H}_8\text{O}_6$	LCO	Ascorbic	$\text{C}_6\text{H}_8\text{O}_6$	80	300	80	[146]
27	Phosphoric and citric	$\text{H}_3\text{PO}_4$ and $\text{C}_6\text{H}_8\text{O}_7$	LNCM-523	Citric	$\text{C}_6\text{H}_8\text{O}_7$	90	30	91.63	[147]
28	Maleic and ascorbic	$\text{C}_4\text{H}_4\text{O}_4$ and $\text{C}_6\text{H}_8\text{O}_6$	LCO	Ascorbic	$\text{C}_6\text{H}_8\text{O}_6$	80	60	>90	[148]
29	Ammonia–ammonium sulfate	$\text{NH}_3$ – $(\text{NH}_4)_2\text{SO}_4$	LNCM	—	—	80	40	94	[149]

Notes: “—” represents a lack of data, LCO means  $\text{LiCoO}_2$ , and LNCM means  $\text{LiNiCoMnO}_2$ . Other elements in these materials may have been unlisted (e.g., Mn, Li, and Ni).

tions of the leaching process are directed as follows in Eqs. (11)–(13). First, cationic exchange occurs between added  $\text{Cu}^{2+}$  and the cationic  $\text{Li}^+$  in  $\text{LiCoO}_2$  to form an intermediate product  $\text{CuCo}_2\text{O}_4$  on the solid surface that does not precipitate. In the presence of iron ions, high-state cobalt is reduced, and  $\text{CuCo}_2\text{O}_4$  dissolves into the solution. The remaining  $\text{Cu}^{2+}$  undergoes another cationic exchange reaction with  $\text{LiCoO}_2$  to promote continuous dissolution.



## (2) Separation and recovery process.

After the leaching process, cobalt and other elements are enriched in the solution, which is subsequently recoverable. Various methods exist for separating cobalt from leachates, including chemical precipitation, solvent extraction [155], electrolytic deposition [156], ion exchange, electrodialysis, and salting-out. Nayl *et al.* [157] and Sun *et al.* [62] investigated the factors influencing cobalt precipitation when utilizing ammonium oxalate as a precipitating agent. Kang *et al.* [120] employed a sulfuric acid-hydrogen peroxide system for leaching the cathode material, and cobalt ions in the solution were precipitated by oxalic acid, resulting in a cobalt recovery rate exceeding 98%. Chen *et al.* [158] studied cobalt recovery from waste lithium cobaltate materials using phosphoric acid as a leaching agent and precipitator. The cobalt recovery rate reached 99%, while the purity of the cobalt phosphate ( $\text{Co}_3(\text{PO}_4)_2$ ) product reached 97.1% under optimal precipitation conditions: a phosphate concentration of  $0.7 \text{ mol}\cdot\text{L}^{-1}$ , liquid-to-solid ratio of  $20 \text{ g}\cdot\text{L}^{-1}$  hydrogen peroxide of 4vol%, temperature at  $40^\circ\text{C}$ , and a reaction time of 60 min.

As a highly efficient method for separating metal ions, solvent extraction is used to recover valuable metals by leveraging the selectivity of organic solvents. According to differences in solution environments, cobalt extraction is divisible into chelating, basic, acidic, and neutral systems, as well as other types of systems. Torkaman *et al.* [159] investi-

gated the extraction efficiency of various elements in a battery using three acidic extractants (Cyanex301, D2EHPA, and Cyanex272) and two alkaline extractants (Alamine336 and TOA), as illustrated in Fig. 9. In comparison to other extractants (D2EHPA and Cyanex272), Cyanex301 exhibited superior performance in cobalt extraction, but Cyanex272 effectively facilitated the separation of Co and Ni. Traditional liquid–liquid extraction employs an organic solvent and aqueous solution as two immiscible phases, but the former substance is typically a toxic, combustible, and volatile compound. Ionic liquids have emerged as a sustainable alternative solvent for extraction and hold great potential in separation applications [160–161]. Numerous studies have investigated the use of ionic liquids, such as phosphonium-based [162–164] and fatty-acid-based [165] ionic liquids, for cobalt extraction from batteries [166–171]. Ilyas *et al.* [172] developed the trihexyl(tetradecyl)phosphonium bis-2,4,4-(trimethylpentyl) phosphinate to selectively separate cobalt from an HCl-leached solution of spent lithium-ion batteries. Similarly, cobalt was recovered from the cathode degradation process of lithium-ion battery waste through functional ionic liquid extraction by Morina *et al.* [173], enabling effective battery recycling. The results demonstrate that over 70wt% of lithium can be separated from cobalt without adding organic acid or EDTA, highlighting the efficient separation of lithium and cobalt via the 3-methyl-1-octylimidazolium thenoyl-trifluoroacetone, Omim-TTA, ionic liquid. Jing *et al.* [174] synthesized the new task-specific ionic liquids of  $[\text{C}_8\text{H}_{17}\text{NH}_2][\text{Cyanex272}]$  with good performance for selective extraction and separation of  $\text{Co}^{2+}$  by a one-step reaction of n-octylamine and Cyanex 272. The separation factor of  $\text{Co}^{2+}$  and  $\text{Ni}^{2+}$  ( $\beta_{\text{Co/Ni}}$ ) was 976.9, and high-purity  $\text{CoSO}_4\cdot 7\text{H}_2\text{O}$  (99.7wt%) was obtained. Moreover, important advancements have occurred in cobalt recovery from spent lithium batteries using deep eutectic solvents [175–184], which are considered analogs to ionic liquids [185–186].

## 4.1.3. Pyro-hydrometallurgical processing for cobalt recovery in electrodes

A single method has limitations regarding cobalt recovery

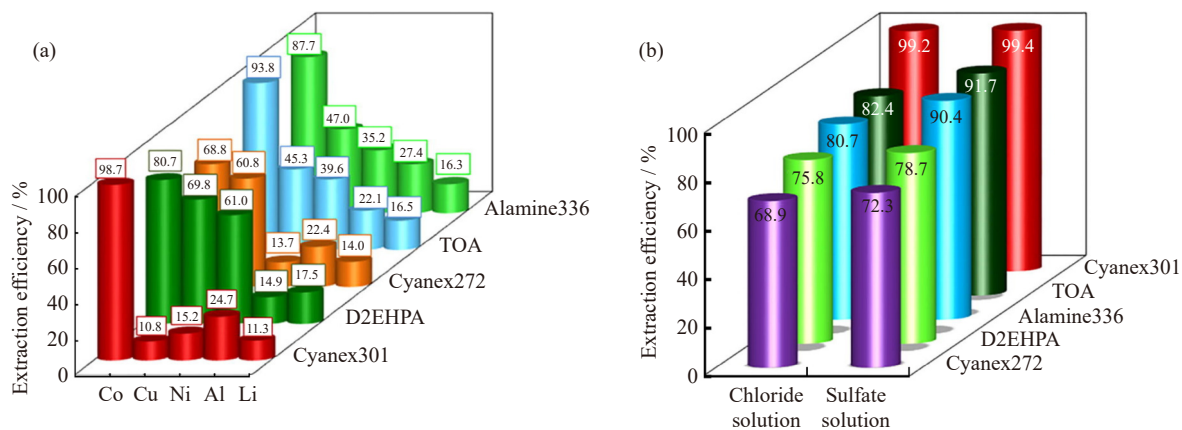


Fig. 9. (a) Extraction efficiency of three acidic extractants and two alkaline extractants for cobalt and others in lithium-ion batteries and (b) influence of chloride ions and sulfate ions on cobalt extraction. Reprinted from *Sep. Purif. Technol.*, 186, R. Torkaman, M. Asadollahzadeh, M. Torab-Mostaedi, and M.G. Maragheh, Recovery of cobalt from spent lithium ion batteries by using acidic and basic extractants in solvent extraction process, 318, Copyright 2017, with permission from Elsevier.

and product quality, but cobalt in raw material can be transformed into a form suitable for hydrometallurgical recovery after pyrometallurgical pretreatment. Therefore, cobalt resources from spent lithium batteries can be efficiently recovered through the combined process of pyrometallurgy and hydrometallurgy.

Unlike reduction smelting, the combined process involves high-temperature roasting with the addition of a reducing agent to convert an elemental metal or low-cost metal oxides, enabling the subsequent recovery of valuable components such as cobalt and nickel through hydrometallurgical processes. The reduction roasting process is commonly used for cobalt from lithium-ion batteries, offering advantages such as preventing toxic slag formation, minimizing heating waste, and mitigating fluorine pollution. Moreover, carbon materials on the negative electrode can effectively reduce cobalt oxides during roasting, enhancing leaching kinetics conditions and facilitating increased recovery of valuable metals. Liu *et al.* [187] reduced the high-valence cobalt in cathode material using hydrogen; then, lithium leached into the solution and left cobalt in the slag in the water-leaching process. Wang *et al.* [188] investigated the reduction roasting of  $\text{LiCo}_2\text{O}_4$  in argon using cathode aluminum foil as the reducing agent. The resulting slag was enriched with  $\text{CoO}$ , which was leached by sulfuric acid to obtain a cobalt sulfate solution with a concentration of more than  $160 \text{ g}\cdot\text{L}^{-1}$ . Evaporation and crystallization were used to produce  $\text{CoSO}_4$  products. In addition, numerous scholars have studied the carbon reduction process as a reductant. Huang *et al.* [189] proposed the technical route of using vacuum carbothermal

reduction–magnetic separation to separate nickel–cobalt and established a calculation model of  $\text{Co}^{2+}$  and  $\text{Ni}^{2+}$  reduction temperature by analyzing the heat transfer process in the vacuum reduction process. Hu *et al.* [190] studied the carbon reduction thermodynamics of metal oxides (Co, Ni, and Mn) under different pressure conditions, and they noted that the carbothermal reduction reaction has a lower initial temperature and is more complete under vacuum conditions. Fu *et al.* [191] proposed a microwave-assisted carbothermal reduction process, along with fumaric acid leaching, for recovering valuable metals from cathode materials. This process demonstrates that the cathode material is reduced to Co, Ni, MnO, and  $\text{Li}_2\text{CO}_3$ .  $\text{Li}_2\text{CO}_3$  can be directly leached by water, with a remarkable recovery rate of 99%. Under optimized leaching conditions in fumaric acid, the water-leaching slag rich in Ni, Co, and Mn achieves a leaching ratio exceeding 96%, enabling the production of various metal salt products through stepwise precipitation methods. Fig. 10 shows the microwave carbothermal reduction device [191].

#### 4.2. Recovery of cobalt from waste cobalt-containing alloys

China generates a substantial amount of cobalt alloy waste, including waste from magnetic, high-temperature, and hard alloys. There are obvious differences in the composition of cobalt alloys [192] for different purposes, resulting in the diverse treatment methodologies. The principle, however, is to recover and recycle cobalt resources from waste products through either pyrometallurgical or hydrometallurgical processes.

Sm–Co permanent magnets can be recovered through

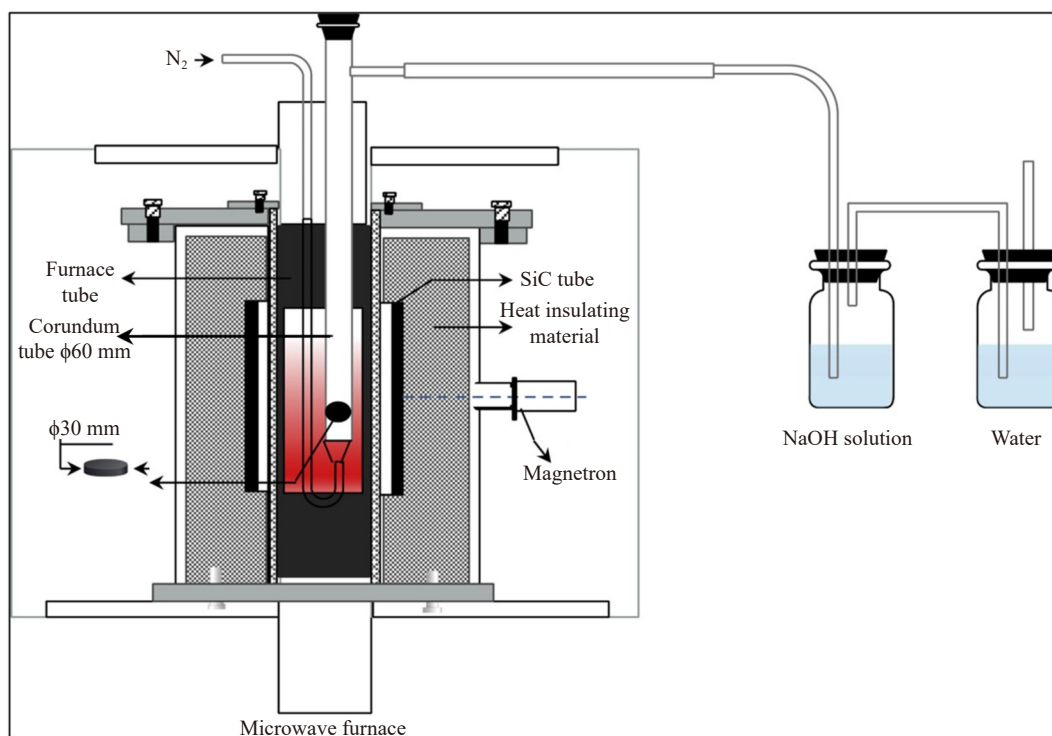


Fig. 10. Schematic of the microwave carbothermal reduction device. Reprinted from *J. Alloys Compd.*, 847, Y.P. Fu, Y.Q. He, J.L. Li, *et al.*, Improved hydrometallurgical extraction of valuable metals from spent lithium-ion batteries via a closed-loop process, 156489, Copyright 2020, with permission from Elsevier.

various methods, including direct recycling, the pyrometallurgical process, and hydrometallurgical recovery. Direct recycling is the most efficient and cost-effective method for treating Sm–Co processing waste, with minimal environmental impact [193–194]. The pyrometallurgical process has high efficiency but requires high energy consumption due to its operation under high-temperature conditions, resulting in solid waste that needs further treatment [195]. The hydrometallurgical process uses acid leaching, solvent extraction, or selective precipitation to recover cobalt [196–197]. Acid leaching is crucial for removing impurities and efficiently extracting high-value metals. Commonly used acids include  $H_2SO_4$ , HCl, and  $HNO_3$ , in which HCl is typically chosen to facilitate solvent extraction of target metal elements.

The cobalt content in high-temperature alloys is typically approximately 5wt%. Common methods for recovering cobalt include electrochemical dissolution and pressure acid leaching [198–199]. Zhang *et al.* [200], Song *et al.* [201], and Maharana *et al.* [202] used the electrochemical dissolution method to extract metal elements from cobalt-based high-temperature alloys (Ni 70wt% and Co 5wt%), resulting in a cobalt–nickel solution subsequently recovered through extraction separation techniques. Wang *et al.* [203] employed acid leaching and  $CoSO_4$  crystallization to treat cutting waste, achieving initial cobalt separation before using hydrometallurgical processes to produce  $Co_2O_3$  products.

The recovery of hard alloys is particularly remarkable because of its main component, WC, and the presence of 5%–10% Co [204–208]. WC does not react with  $H_2SO_4$ , HCl, and  $HNO_3$ . The recovery of hard alloys can be achieved through various methods, including oxidation, electrolysis, zinc melting, and the  $H_3PO_4$  method [209–210]. Because of the favorable results achieved through hydrometallurgical processes in recovering other alloy materials, numerous scholars are dedicating their efforts toward employing these processes for cemented carbide recovery.

### 4.3. Recovery of cobalt from waste cobalt-containing catalysts

Molybdenum–cobalt catalysts receive extensive use in the fertilizer, petroleum, and synthetic ammonia industries because of their exceptional catalytic activity. Over a certain period of reaction, catalysts will undergo deactivation, aging, or poisoning, leading to a decline in their effectiveness and generating waste materials [211–212].

As molybdenum exhibits amphoteric properties, cobalt separation and recovery from waste molybdenum–cobalt catalysts can be achieved through acid leaching or alkali leaching [213–215]. In China, acid leaching is the most commonly used method for treating waste catalysts because of its simplicity and low cost, and commonly employed leaching systems include  $H_2SO_4$  and mixed-acid solutions [216]. However, it requires substantial acid consumption and high equipment demand. The alkaline leaching method is currently the most extensively studied technique, with three main types [217]: oxidation alkaline leaching, high-pressure

alkaline leaching, and alkali roasting–water leaching [218]. In the oxidation alkaline leaching method,  $H_2O_2$  or  $KClO$  is primarily used as an oxidant to extract cobalt from waste catalysts by adding basic solutions such as those of NaOH,  $Na_2CO_3$ , and  $NH_3$  [219]. High-pressure alkaline leaching is one of the earliest and most well-established methods for recovering waste catalysts [220]. The alkali roasting and water leaching method is widely employed to recover waste molybdenum–cobalt catalysts. This approach involves converting molybdenum oxide and sodium salt into the corresponding sodium molybdate under high-temperature conditions, while cobalt oxide remains unreactive with sodium salt. Subsequently, hot water leaching effectively enables separation of cobalt and molybdenum [221–222].

## 5. Conclusions and prospects

The rapid consumption of cobalt has resulted in the near depletion of exploitable cobalt ore. The minerals used for cobalt production primarily consist of nickel–cobalt ore, copper–cobalt ore, zinc plant residue, and waste cobalt products. Among these minerals, nickel–cobalt ore, copper–cobalt ore, and zinc plant residue receive direct use as metallurgical raw materials for various cobalt products' manufacturing. As cobalt predominantly occurs in associated metal ores, its recovery will be strengthened by advances in primary metal metallurgy. Waste cobalt products possess a higher composition and grade of cobalt and include lithium-ion batteries, which hold great potential as recycled raw materials for obtaining cobalt. Therefore, the use of diverse sources to produce cobalt will be enhanced to meet developmental requirements.

Pyrometallurgical processes are characterized by ease of operation, simplicity, strong adaptability to raw materials, high efficiency, and direct obtainment of cobalt products. However, these processes suffer from substantial cobalt losses and inadequate cobalt matte metallization, which demonstrate promising applications in treating high-grade cobalt ore or low-purity cobalt products. Meanwhile, hydrometallurgical processes offer advantages such as efficient cobalt recovery, particularly for oxide ores, and the production of high-purity cobalt products. Nevertheless, atmospheric pressure leaching presents challenges, including large acid consumption and incomplete impurity separation. Although selective leaching methods such as high-pressure leaching, ammonia leaching, and microbial leaching can reduce leaching agent consumption while being environmentally friendly through selective extraction of cobalt, these methods entail disadvantages such as complex operational procedures and higher costs. In the future, an unrestricted supply of raw materials and high recovery efficiency will be necessary because of the decreasing cobalt grade in minerals and increasing waste cobalt products. Therefore, pyro-hydrometallurgical processes that integrate various methods have greater potential for advancement.

This review indicates several future research topics for cobalt mineral and secondary resources: (1) advancements in

marine cobalt mining technologies to enhance resource inputs into the cobalt industry; (2) development of short-process recovery methods to minimize cobalt losses during processing; (3) investigation of organic acid leaching and ammonia leaching technologies; (4) and exploration of separation applications involving ionic liquids and deep eutectic solvents.

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## Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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