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A review of the application of nanoparticles as collectors in ion flotation

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Abstract: Ion flotation is one of the most promising and unique methods for reducing or removing toxic heavy metal ions, organic pollutants, or inorganic anions and cations from mining and metallurgical wastewater. It is a cost-effective and convenient method. In ion flotation, surface-active ions are removed from aqueous solutions by adding surfactants. Therefore, the main purpose of this review article was to summarize the application of various surfactants (nanoparticle surfactants, chemical synthetic surfactants, and biosurfactants) used in ion flotation. Then, the advantages, disadvantages, and prospects of surfactants were comprehensively discussed. Recent progress regarding nanoparticle surfactants in ion flotation and the mechanism of colligends binding with nanoparticles were evaluated.

Keywords: ion flotation, nanoparticle surfactants, synthetic chemical surfactant, biosurfactants, mechanisms

1. Introduction

Heavy metal ions are widely present in wastewater from various industries. Heavy metals producing sources include mining activities, power plants, metal plating, electronic manufacturing industries, paint and paper factories, battery and metal alloy production lines, leather production units, and many others. Heavy metals threaten the human body's health and the environment, so it is necessary to find new methods to remove toxic heavy metal ions from industrial wastewater. In the last 10 years, many methods that have been used to treat valuable metal ions from aqueous wastewater are based on physicochemical, electrochemical, or advanced oxidation processes (Azimi et al., 2017; Fan et al., 2017; Hoseinian et al., 2019; Kolluru et al., 2021).

Physicochemical processes include ion-exchange, adsorption, chemical precipitation, membrane filtration, and ion flotation (Doyle 2003; Hoseinian et al., 2017). Chemical precipitation is a simple and inexpensive process for treating heavy metal-contaminated wastewater. Meanwhile, it requires large areas, long settling times, and produces large volumes of sludge that require further treatment. Unfortunately, most other methods suffer from high cost, high energy demand, generation of secondary sludge, or limited efficiency in large volumes of dilute solutions that require disposal (Azimi et al., 2017; Fan et al., 2017; Hoseinian et al., 2020; Kolluru et al., 2021; Arslan and Bulut 2022).

Among all the mentioned methods, those with cost-effective, environmentally ecofriendly, and do not produce high-volume secondary wastewater production are preferred. Flotation is a simple, low-operation cost, and selective method that has proven to be an effective process for removing ions from aqueous solutions. Ion flotation, precipitative flotation, sorptive flotation, dissolved air flotation, and foam fractionation are types of flotation methods (Rubio et al., 2002; Hoseinian et al., 2018). Precipitative flotation or sorptive flotation may be ineffective at dilute ion concentrations, producing large amounts of hazardous sludge (Schlebusch et al., 2023). However, at dilute ion concentrations, ion flotation produces small sludge volumes and requires low equipment costs (Hoseinian et al., 2019; Saleem et al., 2019; Mondal et al., 2021; Schlebusch et al., 2023).

Ion flotation was described by Sebba in 1959. He outlined the basic principles of the ion flotation process. It is an effective separation technology for removing or recovering anions and cations of heavy metals and organic pollutants from wastewaters (Doyle 2003; Peng et al., 2019).

The ion flotation process is depicted in Fig. 1. In this process, an initial homogeneous solution is rendered heterogeneous by the addition of oppositely charged surfactants. Surfactant ions with opposite charges react with targeted ions (colligends). Therefore, an insoluble complex is formed. These insoluble complexes (sublates) transfer to the foam phase by the rising bubbles of air and are removed from the solution (Wang 2006; Hoseinian et al., 2018; Chang et al., 2019; Peng et al., 2019; Xanthopoulos and Binnemans 2021). In ion flotation, the optimal surfactant concentration must be at least stoichiometric and less than critical micelle concentration (CMC) to accomplish almost full ion removal efficiently (Hoseinian et al., 2020).

Researchers have shown that ion flotation performance is affected by two categories of parameters: hydrodynamic parameters include impeller speed, bubble surface area, air flow rate, column size, gas retention, sparger geometry, and bubble size and operation parameters include concentration and type of surfactants, ion concentration, ratio of surfactant concentration to ion concentration, electrolytes, pH, flotation time, temperature, radius and ion charge, ionic strength, activity coefficient, presence of foreign ions, ratio of metal ion recovery to water recovery, and gas type. After the pH, the most important parameter in the ion flotation is the concentration of the surfactant. Choosing the suitable surfactant is an important tool for the selective separation of ions from wastewater (Rezai and Kowsaric 2019; Hoseinian et al., 2020).

According to the literature, the types of surfactants used in ion flotation include synthetic chemical surfactants, biosurfactants, and nanoparticle surfactants (Fig. 2). The most commonly used surfactants to date are chemically synthesized surfactants and biosurfactants (Chang et al., 2021).

This review article evaluates various types of surfactants utilized in ion flotation and suggests future research directions. Also, the potential of nanoparticle surfactant application for the removal or recovery of various ions from wastewater using the ion flotation process was studied. Then, the mechanism of colligends binding with nanoparticles was comprehensively evaluated.

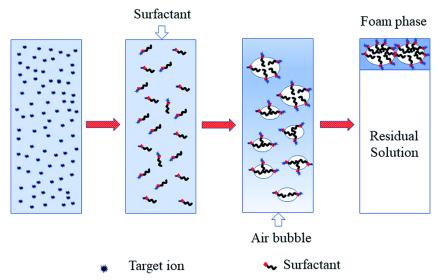


Fig. 1. Schematic of the overall mechanism of ion flotation (Chang et al., 2019)

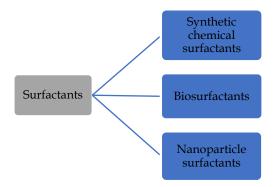


Fig. 2. General classification of surfactants

2. Synthetic chemical surfactants

Synthetic chemical surfactants such as Tetra decyl amine (TDA) (Lusher and Sebba 1965), asulphopalmitic acid (α-SPA) (Davis and Sebba 1966; Rose and Sebba 1969), Ethylhexadecyldimethylammonium bromide (EHDABr) (Grieves et al., 1973; Mcdonald and Suleiman 1979; McDonald and Ogunkeye 1981; McDonald and Jaganathan 1982), hexadecyltrimethyl ammonium chloride (HTAC) (Takayanagi et al., 1976), Dodecyl-trimethylammonium chloride (DTAC) (Kobayashi et al., 1980), Potassium ethyl-xanthate (KetX) (Stalidis et al., 1989), cetylpyridinium chloride (CPC) (Hualing and Zhide 1989), diethyl-dithiocarbamate (DTC) (Stalidis et al., 1989), lauryl amine (Zouboulis et al., 1990), Dodecyl amine (Zouboulis et al., 1990; ZHAO et al., 1996; Zouboulis et al., 1996), octylhydroxamic acid (HOHX) (Jdid et al., 1990), sodium dodecyl benzenesulfonate (SDBS) (He 1991; Ulewicz et al., 2003; Bai et al., 2018; Corpuz et al., 2018), Cetyltrimethylammonium bromide (CTAB) (Galvin et al., 1992; Choi and Choi 1996; Shakir et al., 2010; Thanh and Liu 2021), Ammonium Tetra Decyl Sulfonate (ATDS) (CHAREWICZ et al., 1999), cetylpyridinium chloride (CPCl) (CHAREWICZ et al., 1999), sodium tetradecylsulfate (STS) (Liu and Doyle 2001), sodium hexadecylsulfate (SHS) (Liu and Doyle 2001), Hexadecyl trimethyl ammonium bromide (HTAB) (Polat and Erdogan 2007; Jafari et al., 2017), Potassium amyl xanthate (PAX) (Reyes et al., 2009), sodium diethyldithiocarbamate (DEDTK) (Strel'tsov and Abryutin 2010), sodium lauryl sulfate (SLS) (Shakir et al., 2010), sodium isopropyl xanthate (SIX) (Reyes et al., 2012), N-(2-hydroxyethyl) alkyl amines (HEA) (Chekanova et al., 2012), cetylpyridinium bromide (CPB) (Stoica et al., 2015), sodium trideceth-4 carboxylate (AEC) (Lu et al., 2015), sodium alginate (Corpuz et al., 2018), D-mannitol (Bai et al., 2018), sodium stearate (Shakir et al.,) (Mafi and Khayati 2021) and Sodium dodecyl sulfate (SDS) (Zouboulis and Maris 1995; Scorzelli et al., 1999; Lazaridis et al., 2004; Medina et al., 2005; Uribe-Salas et al., 2005; Polat and Erdogan 2007; Salmani et al., 2013; Hoseinian et al., 2015; Hoseinian et al., 2017; Yenial and Bulut 2017; Zahra et al., 2017; Hoseinian et al., 2018; Lobacheva 2021; Xanthopoulos et al., 2021; Zakeri Khatir et al., 2021) were applied throughout the process of ion flotation. Table 1 summarizes the research performed in ion flotation by synthetic chemical surfactants.

Surfactants are used as collectors or frothers or both in the flotation process. Surfactants whose main role is to make the solid surface hydrophobic are called collectors. Surfactants are classified into nonionic and ionic types. Ionic surfactants are divided into cationic surfactants, whose hydrophilic groups dissociate into cations, and anionic surfactants, whose hydrophilic groups dissociate into anions, in an aqueous solution.

A typical surfactant molecule consists of a functional group (polar head) and a non-polar chain (Fig. 3a). Anionic surfactant (such as SDS) can form insoluble complexes (sublates) by absorbing colligend cations M^{2+} (such as Zn^{2+}) (Fig. 3b). These sublates are accumulated and transformed into a foam phase by rising air or gas bubbles (Uribe-Salas et al., 2005).

During ion flotation, various ions with different charges in solution compete with each other to react with collector ions, which can reduce the recovery of the desired ions. However, the presence of ions with different charges in wastewater makes a necessary to develop selective ion flotation methods for the effective recovery of desired ions. The separation of ions with different charges is generally straightforward, an ion with a higher charge has a higher selectivity for interaction with collector ions. However, some studies have shown that the selectivity of alkali and halide ions during ion flotation is based on the decreasing order of their crystal ionic radius. The selectivity of two ions with the same charge has been rarely investigated (Khatir et al., 2022; Khatir et al., 2022).

To change the selectivity coefficient in an ion flotation system, the total Gibbs free energy of adsorption of one of the cations in the system must be altered. This can be achieved by using chelating reagents or auxiliary ligands such as Ethylenediaminetetraacetic acid (EDTA) (Soliman et al., 2015), Dodecyldiethylenetriamine (Ddien) (Liu and Doyle 2009), Triethylenetetramine (Trien) (Hoseinian et al., 2021; Xanthopoulos and Binnemans 2021) and Di-(2-Ethyl Hexyl) Phosphoric Acid (D2EHPA) (Khatir et al., 2022) to increase the selectivity of ions.

According to the equilibrium of charge exchange at the interface, the selectivity coefficient (K_X^{Y}) was calculated by the Eq. (1):

$$K_X^Y = \frac{R_1}{R_2} \tag{1}$$

where R_1 and R_2 are the removal percentages of two ions that are compared with each other (Liu and Doyle 2001).

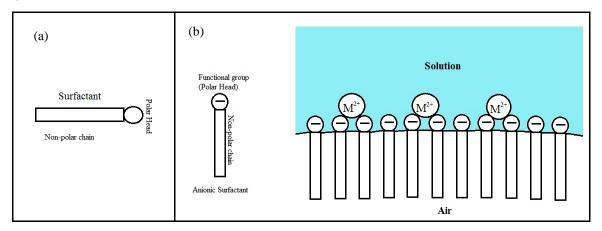


Fig. 3. (a): Typical structure of surfactant, (b): Adsorption of the anionic surfactant SDS- at the air-solution interface with colligend cation M²⁺

Collectors	Colligends	Conditions	Removal %	References
TDA	Al ³⁺	$pH = 4$, $C_{TDA} = 7.0 \text{ mmol}/\text{dm}^3$	90	(Lusher and Sebba 1965)
a CDA	Sr	pH = 3.6-3.8, $C_{collector}$ = 6.0 g/dm ³ (2.24×10 ⁻⁴ mol/dm ³), air flow rate = 1250 cm ³ /h	97.3	(Davis and Sebba 1966)
a-SPA	Y(III)	pH = 2.75, $C_{collector}$ = 1.5×10 ⁻⁴ mol/dm ³ , $C_{Metal ion}$ = 5×10 ⁻⁵ mol/dm ³ , flotation time = 30 min	99.5	(Rose and Sebba 1969)
	Cr(VI)	pH = 4.2, C _{collector} : C _{Metal ion} = 2:1, C _{collector} = 0.93 mmol/dm ³ , C _{Metal ion} = 0.46 mmol/dm ³ , Temp = 23-33°C	100	(Grieves et al., 1973)
EHDABr	Cu (II)	$pH = 6 \text{ to } 11, \text{ flotation time} = 60 \text{ min, air flow rate}$ $= 40 \text{ cm}^3/\text{min, } C_{\text{collector}} : C_{\text{Metal ion}} = 4:1, C_{\text{collector}} = 2.4 \times 10^{-4} \text{ mol}/\text{dm}^3$	99.3	(Mcdonald and Suleiman 1979)
	Zn(II)	Zn(II) = 5 ppm, flotation time = 150 min, air flow rate = 50 cm ³ /min, C _{collector} : C _{Metal ion} = 27:1, C _{collector} = 2.1×10 ⁻³ mol/dm ³	94.8	(McDonald and Ogunkeye 1981)
	Ni(II)	pH = 5, flotation time = 60 min, air flow rate = 50 cm ³ /min, C _{collector} : C _{Metal ion} = 11.8:1, C _{collector} = 8×10^{-4} mol/dm ³	87	(McDonald and Jaganathan 1982)
HTAC	Fe ³⁺ ,Mn0 ⁻ ₄	pH = 11, C _{Metal ion} = $7.45 \times 10^{-4} \text{ mol}/\text{dm}^3$	98, 97	(Takayanagi et al., 1976)
DTAC	Cd(II)	pH = 11, flotation time = 7 min, air flow rate = 10 cm ³ /min, Temp = 25°C, C _{collector} = 1.78×10 ⁻⁴ mol/dm ³	96	(Kobayashi et al., 1980)
KetX	Cu(II)	pH = 3, flotation time = 10 min, air flow rate = 400 rpm, $C_{collector}$: $C_{Metal ion}$ = 1.1:1, $C_{Metal ion}$ = 50 ppm	90	(Stalidis et al., 1989)
CPC	A wide range of ions	flotation time = 5 min, air flow rate = 100 cm ³ /min, C _{collector} = 1.8-6.0× 10^{-4} mol/dm ³ , use with Nitrogen gas	95-100	(Hualing and Zhide 1989)
DTC	Zn(II)	pH = 5, flotation time = 10 min, air flow rate = 400 rpm, C _{collector} : C _{Metal ion} = 1.1:1, C _{Metal ion} = 50 ppm	90	(Stalidis et al., 1989)

Table 1. Various synth	netic chemical sur	factants used as co	ollectors in the ion	flotation process

Lauryl amine	Cr ³⁺	pH = 7, flotation time = 10 min, C _{collector} : C _{Metal ion} = 3:1	90	(Zouboulis et al., 1990)
Dodecyl amine	Lead and ZinC	pH for lead = 7 and for Zinc = 10, flotation time = 5 min, C _{collector} = 2×10^{-4} mol/dm ³ , Ethanol = 0.5% v/v	90	(Zouboulis et al., 1990)
	Mo(VI) As(V)	pH = 3.6, flotation time = 10-15 min, air flow rate = $300 \text{ cm}^3/\text{min}$, C _{collector} = $350 \text{ mg}/\text{dm}^3$ ($1.88 \times 10^{-3} \text{ mol}/\text{dm}^3$), C _{collector} : C _{Metal ion} = $1:1$, Ethanol = $0.5\% \text{ v/v}$	99 99.4	(ZHAO et al., 1996)
	W04 ⁻	pH = 6, flotation time = 10-15 min, air flow rate = 200 cm ³ /min, C _{collector} = 350 mg/dm ³ (1.88×10^{-3} mol/dm ³), Ethanol = 0.5% v/v	100	(Zouboulis et al., 1996)
НОНХ	Zr	pH = 7.8, flotation time = 5 min, Temp = 60° C, C _{collector} : C _{Metal ion} = 3.9:1, C _{Metal ion} = 2.08×10^{-4} mol/dm ³	99.8	(Jdid et al., 1990)
	rhodmm(III) palladmm(II) platmum(IV)	pH = 5, flotation time = 25 min, air flow rate = 30 cm ³ /min, C _{collector} : C _{Metal ion} = 3:1, C _{collector} = 4×10 ⁻⁴ mol/dm ³ , SDBS most efficient collector	94 99.8 99.4	(He 1991)
SDBS	Cd(II) Zn(II)	pH = 4, flotation time = 60 min, air flow rate = 12 cm ³ /min, Temp= 20°C, C _{Metal ion} = 1×10 ⁻⁵ mol/dm ³ , C _{collector} = 1×10 ⁻⁴ mol/dm ³	90	(Ulewicz et al., 2003)
	boron	pH = 10, C _{collector} = 1×10^{-4} mol/dm ³	88.69	(Bai et al., 2018)
	Pb Cu	pH = 5.35, air flow rate = $120 \text{ cm}^3/\text{min}$	99 92	(Corpuz et al., 2018)
	$Au(CN)_2^-$	$pH = 10, C_{collector} = 7.17 \times 10^{-3} mol/dm^3$	91	(Galvin et al., 1992)
	Thoron(TH)	pH = 1.5-12.6, air flow rate = 32 cm ³ /min, C _{collector} : C _{Metal ion} = 4.38, C _{collector} = 2.2×10 ⁻⁴ mol/dm ³	99.9	(Shakir et al., 2010)
СТАВ	Pd	pH = 2, flotation time = 10 min, air flow rate = 90 cm ³ /min, C _{collector} : C _{Metal ion} = 2:1, C _{Metal ion} = 0.2 mmol/dm ³	84.1	(Thanh and Liu 2021)
	Direct Red	pH = 4, flotation time = 3 min, air flow rate = 100 cm ³ /min, C _{collector} : C _{Metal ion} = 2:1, C _{Metal ion} = 25 ppm, V = 250 cm ³	98.68	(Choi and Choi 1996)
ATDS CPC1	Zn(II) Ag(I)	pH = 6.2, flotation time = 6 min, air flow rate = 4 cm ³ /min, Temp = 22°C, Ethanol = 0.5 cm ³	for Zn = 99	(CHAREWICZ et al., 1999)
STS	Cu(II)	pH = 4-5, air flow rate = 20 cm ³ /min, Ethanol =		(Liu and Doyle 2001)
SHS HTAB	Cu(II) Zn(II) Cr(II) Ag(I)	0.4% v/v pH = 10, Ethanol = 0.5%, MIBC for Ag(I) = 0.1%, impeller speed= 750 rpm, flotation time = 16 min, air flow rate = 50 cm ³ /min	74	(Polat and Erdogan 2007)
	Cu(II)	pH = 12, flotation time = 28.5 min, MIBC=0.1% (v/v), water recovery = 24%, C _{collector} = 100 mg/dm ³ , C _{Metal ion} = 10 mg/dm ³	79	(Jafari et al., 2017)
PAX	Cu(II)	C _{collector} = 19 mg/dm ³ , propylene glycol 400 = 30 mg/dm ³ , C _{Metal ion} = 20 mg/dm ³	95	(Reyes et al., 2009)
DEDTK	Cu (II)	pH = 3, air flow rate = $1.8 \text{ dm}^3/\text{min}$, dp = 0.45 mm		(Strel'tsov and Abryutin 2010)
SLS	rhodamineB (Müller et al.,)	pH = 3.4-12.6, air flow rate = $32 \text{ cm}^3/\text{min}$, C _{collector} : C _{Metal ion} = 3:1, C _{collector} = $1.2 \times 10^{-4} \text{ mol}/\text{dm}^3$, Ethanol= 0.1% (v/v)	99.5	(Shakir et al., 2010)

SIX	Ag(II)	pH = 6, C _{collector} = 0.06 g/dm^3	97	(Reyes et al., 2012)
HEA	Cu(II) Co(II)	pH for Cu(II) = 6-10.5 pH for Co(II) = 9-10.5	99.4 96.8	(Chekanova et
	Ni(II)	pH for Ni(II) = 10.6-11.3	99	al., 2012)
СРВ	Cu(II)	pH = 9, flotation time = 10 min, $C_{collector}$: $C_{Metal ion}$ = 0.5:1, $C_{collector}$ = 0.01 mol/dm ³ , dilution ratio = 3:1, air pressure = 4.5×105 Pa	97.09	(Stoica et al., 2015)
AEC	Cd ²⁺	pH = 7.5, air flow rate = 200 cm ³ /min, C _{collector} : C _{Metal ion} = 10:1, C _{collector} = 7.5 mmol/dm ³	99.8	(Lu et al., 2015)
D- mannitol	boron	pH = 10, bubbling speed =1500 rpm, mixing time =10 min, C _{collector} : C _{Metal ion} = 25:1, C _{Metal ion} = 0.03 mol/dm ³ , C _{collector} = 1.4 mmol/dm ³	88.69	(Bai et al., 2018)
SS	Ca ²⁺	pH = 11, air flow rate = $100 \text{ cm}^3/\text{min}$, 1-butanol= $0.5\% (v/v)$, $C_{\text{collector}} = 10 \text{ ppm}$	45.67	(Mafi and Khayati 2021)
	Cu(II)	pH = 6.5, Ethanol = 0.1% v/v, C _{collector} = 50 mg/dm ³	100	(Lazaridis et al., 2004)
	Cd ²⁺	pH = 10-11, flotation time = 30 min, air flow rate = 200 cm ³ /min, C _{collector} : C _{Metal ion} = 4:1, C _{collector} = 15 to 60 mg/dm ³ , Ethanol= 0.25% v/v, C _{Metal ion} = 10 mg/dm ³	100	(Zouboulis and Maris 1995)
	Cd ²⁺	pH = 4-5, air flow rate = $2 \text{ cm}^3/\text{s}$, C _{collector} : C _{Metal} ion = $3:1$, MIBC = $0.1\% \text{ v/v}$, C _{Metal ion} = 20 mg/dm^3	99.1	(Scorzelli et al., 1999)
	Cr ³⁺	pH = 8, air flow rate = $2.1 \text{ cm}^3/\text{s}$, C _{collector} : C _{Metal} ion = $2:1$, C _{collector} = $1 \times 10^{-4} \text{ mol}/\text{dm}^3$, Ethanol = $0.1\% \text{ v/v}$	96.2	(Medina et al., 2005)
	Pb ²⁺	pH = 4, air flow rate = 6.1×10^{-5} to 12.2×10^{-5} m ³ /min, C _{collector} : C _{Metal ion} = 1 : 1 to 4 : 1, C _{collector} = 1 to 4 × 10 ⁻⁴ mol/dm ³ , Ethanol = 0.4% v/v		(Uribe-Salas et al., 2005)
	Cu(II) Zn(II) Cr(II) Ag(I)	pH = 4, Ethanol = 0.5%, MIBC for Ag(I) = 0.1%, impeller speed= 750 rpm, flotation time = 16 min, air flow rate = 50 cm ³ /min, C _{collector} = 144 mg/dm ³	74	(Polat and Erdogan 2007)
SDS	Cd ²⁺	pH = 4, flotation time = 60 min, air flow rate = 200 cm ³ /min, C _{collector} : C _{Metal ion} = 3:1, Ethanol=0.4% v/v, C _{Metal ion} = 25 mg/dm ³	92.1	(Salmani et al., 2013)
	Zn Ni	pH = 3, flotation time = 60 min, air flow rate = 1.8 cm ³ /min, C _{collector} : C _{Metal ion} = 4:1, C _{collector} = 300 ppm, Dowfroth250= 90 ppm, water recovery = 27% - 30%	92 88	(Hoseinian et al., 2015; Hoseinian et al., 2017)
	Cu		97.5	
-	Pb	pH = 9, flotation time = 3 min, air flow rate = 900		(Yenial and
	Zn Cd	rpm, C collector : C Metal ion = 1:1, C collector = 20 mg/dm ³ , water recovery = 27.5%	87 83	Bulut 2017)
	Ni Ga	pH<4, flotation time = 5 min, impeller speed = 1000 rpm, C _{collector} = 0.6×10^{-4} mol/dm ³ , C _{Metal ion} = 1.5×10^{-4} mol/dm ³	92.5 92-93	(Zahra et al., 2017)
	Ni	pH = 9.7, flotation time = 8 min, impeller speed = 800 rpm, $C_{collector}$ = 135.1 ppm, Dowfroth 250 = 20 ppm, use N ₂ gas, water recovery = 37.2%	100	(Hoseinian et al., 2018)
	Nd	pH = 3, air flow rate = $100 \text{ cm}^3/\text{min}$, C _{collector} = $21 \times 10^{-4} \text{ mol}/\text{dm}^3$, water recovery = 26.5%	85.4	(Zakeri Khatir et al., 2021)

Cu	pH = 10.2, flotation time = 300 min, C _{collector} = 5.85 mmol/dm ³ , Ethanol = 0.5% v/v, use N ₂ gas	85	(Xanthopoulos et al., 2021)
Yb	pH = 8.3, flotation time = 5 min, C _{collector} = 0.001 mol/dm ³		(Lobacheva 2021)

According to Table 1, numerous studies have demonstrated the significant value of SDS in ion flotation. SDS (Fig. 4) is one of the most widely used in ion flotation because of its foaming properties in addition to its collector properties.

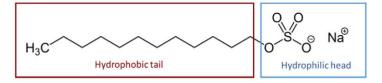


Fig. 4. Chemical structure of SDS (Wołowicz and Staszak 2020)

According to Table 1, ion flotation can effectively remove most toxic heavy metal and alkaline earth metal ions from wastewater using synthetic chemical surfactants. However, synthetic chemical surfactants have several disadvantages, including environmental toxicity, unstable chemistry, non-biodegradability and bioaccumulation, high cost, and possible interference with biological activities. For example, SDS can form bonds with macromolecules such as enzymes, peptides, and DNA. This can change their surface charge, affect the folding of peptide chains, and conflict with normal biological activity. Therefore, it is important to develop and use environmentally friendly and highly efficient surfactants for ion flotation (Mulligan 2009; Khoshdast et al., 2012; Li et al., 2013; Peng et al., 2019; Ziaee et al., 2021).

3. Biosurfactants

Biosurfactants are eco-friendly, surface-active agents produced mainly by bacteria, yeasts, and filamentous fungi. They can be found in various sources, including animals, microorganisms, and agricultural and vegetable residues. Biosurfactants are considered to be more environmentally friendly than synthetic surface-active agents because they are biodegradable and non-toxic.

Biosurfactant molecules are similar to surfactant molecules, consisting of a functional group (polar head) and a non-polar chain. The hydrophilic part can be an alcohol, carbohydrate, amino acid, carboxylic acid, or cyclic peptide, all of which have relatively strong functional groups that can form chelating structures with metal ions. The most commonly used biosurfactants are rhamnolipids, lipopeptides (e.g. surfactin), sophorolipids, glycolipids, trehalolipids, lipoproteins, lichenysin, and polymers (Mulligan 2009; Shekhar et al., 2015; Tang et al., 2018; Peng et al., 2019; Ziaee et al., 2021; Nafi and Taseidifar 2022). Biosurfactants are suitable due to their high efficacy, strong surface activity, low eco-toxicity, high environmental compatibility, biodegradability, and retention of properties over a wide pH and temperature range. Moreover, the source of these products, microorganisms, is easily accessible and reproducible. Therefore, biosurfactants are widely used as flocculants in mineral processing, collectors and frothers in froth flotation, and adsorbents in environmental treatment, among other applications (Cohen and Exerowa 2007; Liu et al., 2017; Nafi and Taseidifar 2022). Table 2 summarizes the research conducted on ion flotation using biosurfactants. Due to stringent environmental regulations, biosurfactants are a promising alternative to traditional chemically synthesized surfactants. However, they have some disadvantages that should be done in further research on the development of biosurfactants.

Rhamnolipids biosurfactants are divided into two types: monorhamnolipids and dirhamnolipids. They are more biodegradable than other synthetic surfactants and have higher frothing power. Dirhamnolipids are used more than monorhamnolipids in ion flotation. Another surfactant used in ion flotation is N-octanoyl-cys.

With the synthesis of amino acid-based surfactants, a new group of surfactants named N-octanoylcys was created (Fig. 5). N-octanoyl-cys surfactant offers several advantages for the removal of heavy

Biosurfactant	Metal ions	Conditions	Removal %	References
Tea saponin	Cd Pb Cu	pH = 6, C _{Biosurfactant} : C _{Metal ion} = 3:1	71.17 98.95 81.13	(Yuan et al., 2008)
Candida lipolytica & Candida sphaerica	Fe(III) Mn(II)	pH = 8.5, $C_{Fe(III)}$ = 61.98 mg/dm ³ , $C_{Mn(II)}$ = 4 mg/dm ³	98.7 94.5	(Menezes et al., 2011)
Rhodococcus opacus	Ni(II) Al(III)	$pH = 5, C_{Biosurfactant} = 2 g/dm^3, C_{Ni(II)} = 5 mg/dm^3,$ $C_{Al(III)} = 50 mg/dm^3, flotation time = 15 min, Gas$ $flowrate = 0.5 cm^3/s, Temp = 25^{\circ}C$	90 93	(Cayllahua and Torem 2011)
Di-rhamnolipid	Cd (II)	pH = 6.5-7, air flow rate = 300 cm ³ /min, Aeration Rate = 90 cm ³ /min	R _{Cd-fresh} = 57	(Bodagh et al., 2013)
Rhamnolipid	Cr(III)	$pH = 8$, $C_{Metal ion} = 40$ ppm, $C_{Biosurfactant} = 4.4$ ppm, flotation time = 5 min, air flow rate = 5 cm ³ /min, C FeSO ₄ for precipitation of Cr(VI) to Cr(III) = 120 ppm, Fe/Cr = 3	95	(Abyaneh and Fazaelipoor 2016)
L-cysteine (97%), octanoyl (C ₈) chloride	Hg (II) As(VI) Pb (II) Cd (II) Cr (II)	pH = 8, flotation time = 60 min, air flow rate = 2 dm ³ /min, C _{Metal ion} = 5 mg/dm ³	99.9 99.6 99.4 99.2 99.7	(Taseidifar et al., 2017)
N-octanoyl- cysteine	Cd ²⁺ Ca ²⁺	pH = 8, C _{Metal ion} = 5ppm, C _{Biosurfactant} = 0.01 mol/dm ³ , flotation time = 60 min, air flow rate = 2 dm ³ /min, Temp = 65°C	>99 97	(Taseidifar 2020)
l-decanoyl cysteine	Cd Cr Cu Ni Zn Mn	pH = 7.5, C _{Metal ion} = 5 mg/dm ³ , C _{Biosurfactant} = 0.003 mg/dm ³ , flotation time = 60 min, air flow rate = 1 dm ³ /min	98.2 98.3 99.4 97.2 99.2 97.3	(Ziaee et al., 2021)
S-octanoyl-cys	As Pb Hg	pH = 8, C _{Metal ion} = 5 ppm, C _{Biosurfactant} = 0.01 mol/dm ³ , flotation time = 60 min	99.5 99.4 99.6	(Ziaee et al., 2021)
Bio-mRL ¹ Three synthetic monorhamnolipids (Rha-C ₁₀ -C ₁₀ Rha-C ₁₂ -C ₁₂ Rha-C ₁₄ -C ₁₄)	U	pH for Bio-mRL = 5.5, pH for monorhamnolipids = 6.5, C _{Metal ion} = 442.4 μ g/dm ³ , C _{Biosurfactant} = 250 μ mol/dm ³ , Ethanol = 0.5% v/v, flotation time = 45 min, air flow rate = 50 cm ³ /min	92.6 81.9 6.6 8.7	(Hogan et al., 2022)
Sodium N- lauroylsarcosinate	Pb(II) Cu(II) Cr(III)	pH for Pb(II) = 5.69, pH for Cu(II) = 5.42, pH for Cr(III) = 4.15, C _{Biosurfactant} : C _{Pb(II)} = 2:1, C _{Biosurfactant} : C _{Cu(II)} = 3:1, C _{Biosurfactant} : C _{mixed ion} = 4:1, stirring speed = 1500 rpm, aeration rate = 0.05 Nl/min, Temp = 25°C, flotation time = 20 min	In single: >96 In mix: >70	(Jia et al., 2022)
Surfactant	Cu ²⁺ Ni ²⁺ Co ²⁺	pH = 7, C _{Surfactin} : C _{metal ion} = 1:3, C _{metal ion} = 100 µmol/dm ³ , C _{Surfactin} = 5 mmol/dm ³ , air flowrate = 0.08 dm ³ /min,	75.2% 94.7% 98.2%	(Jia et al., 2022)

Table 2. Summarizes the most important studies on ion flotation with biosurfactants

¹ biosynthetic rhamnolipid

metal ions by ion flotation. It contains several active groups, such as amine, carboxylic acid, and thiol. The biosurfactant N-octanoyl-cys is non-toxic, highly biodegradable, and environmentally friendly. It can properly orient at the water-air interface and form a monolayer on the bubble surface (Taseidifar et al., 2017).

However, biosurfactants have some disadvantages that limit their large-scale industrial application, such as high collector consumption, low ion removal efficiency, and long flotation time. Therefore, further efforts are needed to develop more efficient biosurfactants in the future.

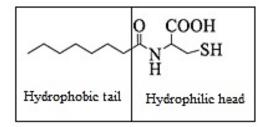


Fig. 5. Chemical structure of N-octanoyl-cys surfactant in acidic form

4. Nanoparticle surfactants

Nanoparticles are substances with at least one dimension in the nanometer range (1-100 nm). In recent years, nanoparticles and nanocolloids have acquired greater interest due to their extensive application in numerous clinical and business fields, including pharmaceuticals, biosensors, magnetic equipment, printing industries, and similar areas. Nanoparticles have attracted a lot of attention due to their high surface free energy, strong chemical activity, high reactivity due to their small particle size, very high specific surface area, and large adsorption capacity (Pokropivny et al., 2007; Müller et al., 2011).

Nanoparticles have been used as collectors, depressants, froth stabilizers (nanofrothers), and modifiers in mineral flotation. However, there are few reports on the application of nanoparticles in mineral processing processes, including flotation. Nanoparticles are divided into organic and inorganic nanoparticles such as froth flotation reagents. Inorganic nanoparticles used in mineral flotation as collectors include hydrophobic polystyrene nanoparticles (Yang and Pelton 2011; Yang et al., 2011; Mabudi et al., 2019; An et al., 2020), hydrophobic polystyrene nanoparticles (latex) (Yang et al., 2013; Razavizadeh 2019), styrene/N-butyl acrylate copolymers (Yang et al., 2013), cationic polystyrene-corepoly(n-butyl methacrylate)-shell nanoparticles (Dong et al., 2016). Organic nanoparticles include talc (Hajati et al., 2016; Choi et al., 2020) and carbon black nanoparticles (Kim et al., 2019).

One of the ion flotation technique's disadvantages is the excessive consumption of the collector. To achieve nearly complete removal of the colligend, the minimum appropriate concentration of the collector must be equal to or higher than the stoichiometric value and less than the critical micelle concentration. The benefits of using nanocollector to remove ions from wastewater include simple synthesis, low cost, high efficiency, stability in aqueous solutions, and collector consumption (Hoseinian et al., 2020; Legawiec and Polowczyk 2020).

Graphene oxide (GO) is a versatile nanoparticle collector that is used in ion flotation. Its high surface area, abundant functional groups, good hydrophilicity, good stability, low cost, and ease of production make it the ideal collector for a variety of ion flotation applications. GO's high surface area allows it to adsorb large amounts of ions, which is essential for ion flotation. GO's abundant functional groups can interact with ions through a variety of mechanisms, giving it a high degree of selectivity for different types of ions. GO's good hydrophilicity and stability make it compatible with water-based solutions and suitable for use in a variety of ion flotation conditions. GO is easy to produce, making it a viable option for large-scale ion flotation applications (Chua and Pumera 2014; Hoseinian et al., 2023).

Peng et al. used GO as a collector to remove Pb(II) from synthetic wastewater. Under optimal conditions, more than 99% of Pb(II) was removed, and the residual turbidity of the solution was 1.4 NTU. Moreover, GO could be reused after optimal desorption, and the Pb(II) removal efficiency remained at 84.9% in the sixth regeneration cycle (Peng et al., 2018).

Hoseinian et al. synthesized and used a new nanocollector of GO in the ion flotation of nickel. To increase the efficiency of the process, they functionalized GO using 2,6-diaminopyridine. The amount

of ion removal increases with collector concentration, but exceeding collector concentration enhances water recovery and decreases the selectivity of the process. They found that increasing the pH increased the removal of nickel ions. Under optimal conditions with amino-functionalized graphene oxide (AFGO) as a nanocollector (SDS concentration of 0.05 g/dm^3 , AFGO concentration of 0.1 g/dm^3 , pH = 9, impeller speed of 800 rpm, and flotation time of 10 min), they were able to completely remove nickel ions from synthetic wastewater (Hoseinian et al., 2020).

Chang et al. used ion flotation to remove Cu(II) ions using GO as a collector. Their research showed that GO's degree of oxidation affects the amount of Cu(II) ions removed during the ion flotation process, and GO's degree of oxidation may be changed to increase the amount of Cu(II) removal. They investigated 4 types of GO (GO1-GO2-GO3-GO4) with different degrees of oxidation by adding CTAB and the adsorption rate of Cu(II) on GO. They found that Cu(II) ions could be removed from synthetic wastewater at optimal conditions of pH = 6, GO concentration = 100 mg/dm³, and the various concentrations of CTAB for different degrees of oxidation of GO, which were 15, 20, 10 and 10 mg/dm³ respectively for GO4-GO3-GO2-GO1 and more than 99% of Cu(II) ions were removed from the synthetic wastewater (Chang et al., 2021; Chang et al., 2021).

Chang et al. used amidoxime-functionalized graphene oxide (AMID@GO) as a nanocollector to study the selectivity and recovery of Cu(II) ions from wastewater by ion flotation. The outcomes demonstrated that AMID@GO had higher selectivity and adsorption capability for Cu(II) than GO. Under optimal conditions (pH = 6, AMID@GO concentration = 300 mg/dm^3 , HTAB concentration = 50 mg/dm^3 , aeration rate = $100 \text{ cm}^3/\text{min}$, and contact time = 40 min) 99.26% of Cu(II) ions were removed from wastewater. Also, AMID@GO showed excellent recycling performance (Chang et al., 2021).

Chang et al. investigated ion flotation using iminodiacetic acid-functionalized graphene oxide (IDA@GO) as a nanocollector for the selective recovery of Pb(II) from a strongly acidic waste electrolyte. IDA@GO outperformed GO in terms of performance, adsorption capacity, and Pb(II) recovery rate. At pH = 2, CTAB concentration = 7.5 mg/dm^3 , reaction time = 40 min, and aeration rate = $100 \text{ cm}^3/\text{min}$, Pb(II) could adsorb up to 91.21 mg/g on IDA@GO. In addition, IDA@GO showed ideal regeneration performance and could be reused as a nanocollector in ion flotation (Chang et al., 2022).

5. Mechanism of colligends binding with nanoparticles

5.1. AFGO-SDS-Ni

According to the literature, collectors with high adsorption capacity at low concentrations are required to reduce collector consumption during the ion flotation process. AFGO, as shown in Fig. 6, has oxygenbased structures such as alkoxy, carbonyl, hydroxyl, and carboxylic acids, which contain more oxygen and nitrogen atoms and provide a large number of active sites for ion adsorption on the AFGO layer. AFGO also has multifunctional linkages suitable for complexing with colligends. FTIR analysis (Fig. 7) reveals the interactions between AFGO, colligend hydrolyzing species, SDS, and water molecules. AFGO contains primary amine groups in its structure that can form coordination compounds with metal ions through its amine functional groups (Hoseinian et al., 2020).

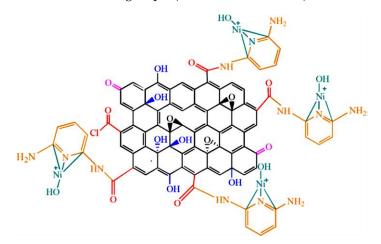


Fig. 6. The mechanism for nickel ion removal using AFGO (Hoseinian et al., 2020)

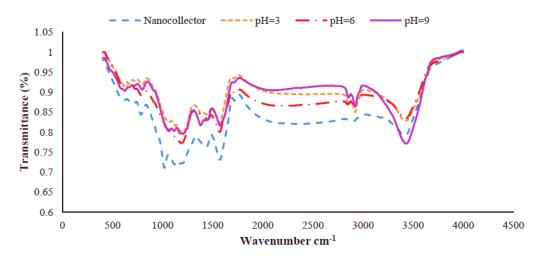


Fig. 7. FTIR analysis of nanocollector before and after nickel ion flotation (Hoseinian et al., 2020)

5.2. GO-Cu(II)-CTAB

According to FTIR and XPS analysis (Fig. 8), the adsorption mechanism of Cu(II) to CTAB on GO and GO-Cu(II) involves the formation of GO-Cu(II) flocs, hydrogen bonding, and electrostatic attraction dominating the adsorption process. All GO-Cu(II)-CTAB spectra showed two additional peaks in the FTIR spectrum following the addition of CTAB. This shift in the C=O stretching peak indicates that the carbonyl and carboxyl groups are the adsorption sites. XPS analysis was used to investigate the chemical state before and after GO as well as Cu(II) and CTAB adsorption. A new binding energy band appeared, confirming the adsorption of CTAB on GO-Cu(II).

Based on Fig. 9, the removal mechanism of Cu(II) ions by GO from wastewater can be summarized as follows: surface complexation, electrostatic attraction, and hydrogen bonding. More oxygen functional groups are created as the degree of oxidation of GO increases, acting as active adsorption sites and enhancing the removal of Cu(II). Furthermore, GO with a higher degree of oxidation was easily flocculated due to the cross-linking function of Cu(II), making bubble collision more effective. The addition of CTAB improves the hydrophobicity of GO-Cu(II) flocs and their adhesion to rising bubbles. CTAB enhanced the hydrophobicity of the GO-Cu(II) flocs and helped them adhere to the rising bubbles and accumulate in the foam phase. In conclusion, compared to GO with various oxidation degrees, GO with a higher degree of oxidation reacts more easily with Cu(II) and binds into larger flocs faster, improving its hydrophobicity with the addition of CTAB, making it easier for bubbles to collision and adhere to the rising bubbles in the foam phase. This suggests that the removal rate of Cu(II) ions by ion flotation can be accelerated by increasing the degree of oxidation of the GO nanocollector (Chang et al., 2021; Chang et al., 2021).

5.3. AMID@GO-Cu(II)-HTAB

SEM, EDX (Figure 10), FTIR analysis (Fig. 11), and zeta potential measurements (Fig. 12) indicated that the Cu(II) adsorption mechanism on AMID@GO involved ion exchange (IX), electrostatic attraction, formation of stable C=N-O-Cu(II) complexes, and interactions between HTAB and AMID@GO@Cu(II). New distinctive peaks emerged in the FTIR spectrum of AMID@GO@Cu(II) compared to that of AMID@GO, demonstrating the interaction between HTAB and AMID@GO@Cu(II). Zeta potential measurements showed that electrostatic attraction dominated the interaction between HTAB and AMID@GO@Cu(II) flocs. WCA analysis was used to elucidate the ion flotation mechanism. The WCA increased with increasing HTAB dosage, indicating a gradual improvement in the hydrophobicity of the flocs.

According to Fig. 13, Cu(II) adsorption onto AMID@GO was dominated by electrostatic attraction, IX, and surface complexation with C-N, C=N, and oxygen-containing functional groups. Then, AMID@GO flocculated into large aggregates, facilitating bubble collision. Next, the hydrophobicity of the AMID@GO@Cu(II) flocs was enhanced by the addition of HTAB. Finally, the flocs were separated from the wastewater by the flotation process (Chang et al., 2021).

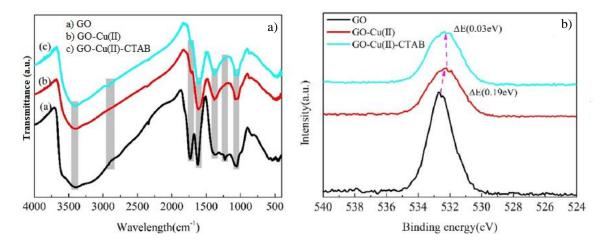


Fig. 8. a) FTIR analysis and b) XPS analysis (Chang et al., 2021; Chang et al., 2021)

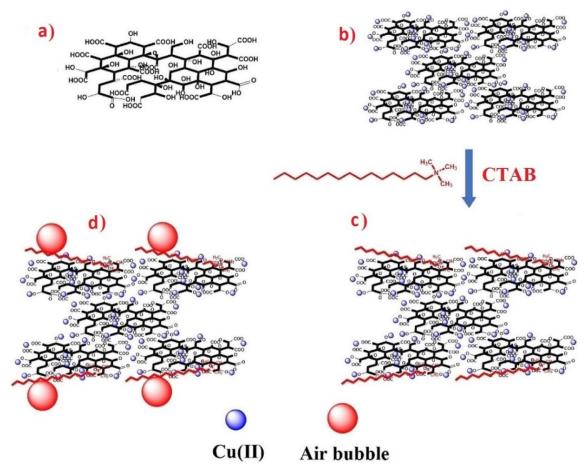


Fig. 9. Schematic for ion flotation of Cu(II) using GO as nanocollector (a): Schematic of GO, (b): Cu(II) adsorbed onto GO, (c): CTAB improved the hydrophobicity of GO-Cu(II) flocs and (d): adhesion of GO-Cu(II)-CTAB flocs to air bubbles (Chang et al., 2021; Chang et al., 2021)

5.4. IDA@GO-Pb(II)-CTAB

FTIR analysis (Figure 14) showed that Pb(II) was adsorbed onto the oxygen-containing functional groups of IDA@GO through surface complexation and IX. Zeta potential analysis (Figure 15) showed that the zeta potential of IDA@GO changed from negative to positive after conditioning with Pb(II), demonstrating that IDA@GO can recover Pb(II) by electrostatic interaction. WCA analysis showed that the WCA of IDA@GO increased as a function of CTAB dosage, indicating that the addition of CTAB increased the hydrophobicity of the Pb(II)-loaded IDA@GO surface.

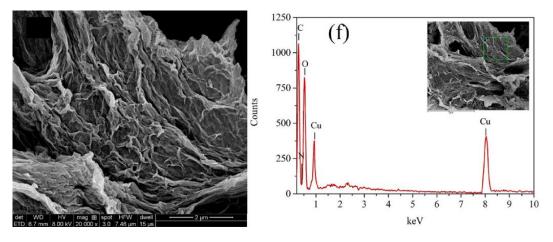


Fig. 10. SEM and EDX images of AMID@GO@Cu(II) (Chang et al., 2021)

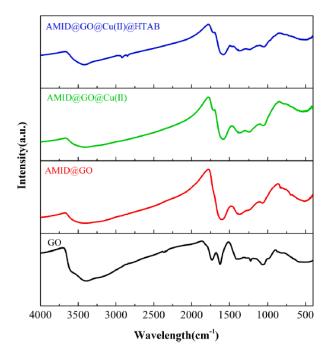


Fig. 11. FTIR spectra of GO, AMID@GO, AMID@GO@Cu(II) and AMID@GO@Cu(II)@HTAB (Chang et al., 2021)

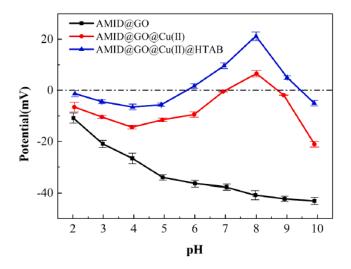


Fig. 12. Zeta potential profiles of AMID@GO, AMID@GO@Cu(II) and AMID@GO@Cu (II)@HTAB (Chang et al., 2021)

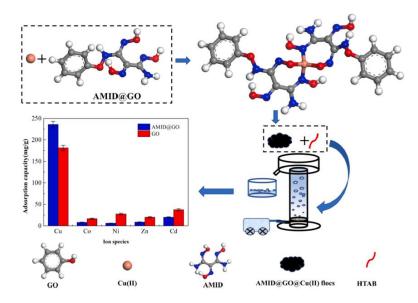


Fig. 13. Schematic Cu(II) selective recovery with AMID@GO as nanocollector (Chang et al., 2021)

According to Fig. 16, Pb(II) adsorption onto IDA@GO first occurred through IX, electrostatic attraction, and surface complexation with carboxyl acting as the adsorption sites. IDA@GO then flocculated into large flocs, increasing the likelihood of Pb(II)-loaded IDA@GO colliding with bubbles. Second, the addition of CTAB enhanced the hydrophobicity of IDA@GO-Pb(II), resulting in the removal of IDA@GO-Pb(II) flocs by rising bubbles (Chang et al., 2022). Table 3 summarizes the mechanism of colligends binding to nanoparticles.

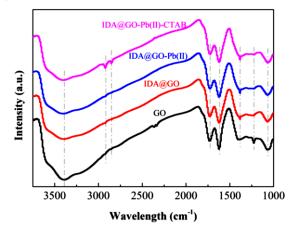


Fig. 14. FT-IR spectra of GO, IDA@GO, IDA@GO-Pb(II) and IDA@GO-Pb(II)-CTAB (Chang et al., 2022)

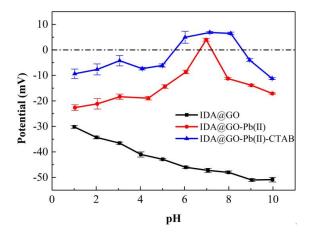


Fig. 15. Zeta potential profiles of IDA@GO, IDA@GO-Pb(II) and IDA@GO-Pb(II)-CTAB (Chang et al., 2022)

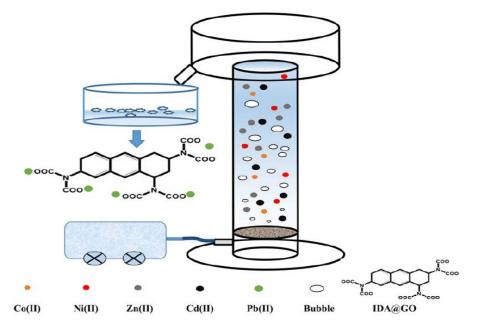


Fig. 16. Schematic for selective recovery of Pb(II) with IDA@GO as a nanocollector (Chang et al., 2022)

Nanoparticle	Colligend	Mechanisms	References
AFGO	Ni	Electrostatic attraction, coordinate bond between AFGO and ions	(Hoseinian et al., 2020)
GO with different oxidation degree	Cu	Surface complexation, electrostatic attraction, and hydrogen bonding	(Chang et al., 2021; Chang et al., 2021)
AMID@GO	Cu	Ion exchange, electrostatic attraction, and surface complexation of C=N-O-Cu(II)	(Chang et al., 2021)
IDA@GO	Pb	Ion exchange, electrostatic attraction, and surface complexation with carboxyl acts	(Chang et al., 2022)

Table 3. Summarizes the mechanism of colligends binding to nanoparticles

6. Conclusions

The review article has been presented to summarize the surfactants used in ion flotation and identify their defects to suggest future studies. The literature describes that researchers have made worthwhile efforts in ion flotation with synthetic chemical surfactants. Nevertheless, it appears that there are still many gaps in this field. In conclusion, the following issues have been identified and should be investigated in future studies:

- Chemical synthetic surfactants are easy to produce and have good selectivity and strong collection ability in ion flotation. However, their high cost, environmental toxicity, unstable chemistry, non-biodegradability, and bioaccumulation severely limit their use in environmental pollution control.
- The amount of biosurfactants or chemical surfactants required to remove any given amount of ions is determined by the stoichiometric relationship and the ion charge. This can lead to increased collector consumption, which can negatively impact the economics and downstream processes of the ion flotation process.
- Environmentally friendly biosurfactants exhibit low removal efficiency and long flotation times, which limits their large-scale industrial applications.
- Biosurfactants and chemical synthetic surfactants can be used to recover the collector, but nanocollector such as graphene oxides can be recovered and reused in the process using a simple washing method. This greatly reduces collector consumption and makes ion flotation more viable for industrial applications.

- Due to the nanoparticles having a high specific surface area and stability in aqueous solutions, their adsorption capacity increases, thereby reducing collector consumption in the ion flotation process. Nanoparticles are emerging collectors with several advantages, including high efficiency, low collector requirements, cost-effectiveness, and stability in aqueous solutions. However, additional research is needed to develop cost-reduction strategies and recyclable processes to further reduce process costs and enable industrialization.
- Various laboratory-scale studies in the field of ion flotation have been conducted on synthetic wastewater. However, it's better to investigate ion flotation's ability to recover ions from real wastewater using a nanocollector.
- One of the gaps in the current understanding of ion flotation is the lack of knowledge about the selectivity of nanocollector. Future research should focus on addressing this need, as it could have important implications for the development of new and improved separation technologies. Additionally, more systematic research should be pursued to better understand the mechanisms involved in ion flotation.
- All studies have focused on GO nanoparticles as a collector, except for GO, bentonite, montmorillonite, and molybdenite nanosheets, which, have the potential to be nanoparticle collectors due to their unique physical and chemical properties.

Overall, it is concluded that the use of nanoparticles as a collector could be more efficient and applicable if the identified shortage items in this paper are investigated.

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