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Review

A review of removal technology for antimony in aqueous solution

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ABSTRACT

Antimony (Sb) and its compounds, toxic metalloid, have been classified as high-priority pollutants. Increasing Sb released into the water environment by natural processes and anthropogenic activities, which exposure threatens to human health and ecosystems. Therefore, it is of unquestionable importance to remove Sb from polluted water. Keeping in view the extreme importance of this issue, we summarize the source, chemistry, speciation, distribution, toxicity, and polluted situation of Sb about aqueous solution. Then, we provide the recent and common technology to remove Sb, which are based on adsorption, coagulation/flocculation, electrochemical technology, membrane technology, ion exchange, etc. In this review, we focus in detail on the adsorption method, researchers at present have been investigating to discover more advanced, cost-effective, eco-friendly, reusable adsorbents. However, to date the Sb-containing wastewater treatment technologies are not sufficiently developed and most of research have been tested only in controlled lab conditions. Few reports are available that include field studies and applications. We critically analyzed the salient features and removal mechanisms, evaluating benefits and limitations of these technologies, hoping to provide more references for further research. Finally, we considered the Fe- or Mn-based technologies was the most promising technique to remove Sb for field application.

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Introduction

In the past few decades, antimony (Sb) is increasingly discharged and distributed into environment. The contamination of the water environment by Sb had already revealed adverse effects also caused crises to emerge on a global scale. An ever-growing number of research articles have described this critical issue (Wang et al., 2011). Antimony is a nonessential toxic metalloid, with chemistry similar to metalloid arsenic. It exhibits properties of both metals and nonmetals (Li et al., 2014), belonging to Group 15 of the periodic chart (Filella et al., 2002). Antimony is naturally present in aquatic systems, introduced by natural processes and anthropogenic activities, making human life miserable in Sb-geogenic areas (Liu et al., 2010; Sánchez-Rodas et al., 2016). Natural processes including volcanic activities, rock weathering, biological activities, water-rock interactions, soil runoff, etc. All these factors that promote the dissolution, migration and transformation of Sb compounds. Anthropogenic activities release lots of Sb-pollutants through excessive exploitation and overuse of the Sb containing products (e.g. catalysts, batteries, raw material for flame retardants, the semi-conductor industry, fossil fuel combustion, timber preservatives, etc.). In particular anthropogenic activities result in increasing concentrations of Sb being emitted into atmosphere, soil and aquatic systems (Filella et al., 2002; He et al., 2012; Ungureanu et al., 2015; Hu et al., 2017).

Long-term exposure to a high level Sb-contaminated water poses adverse effects, which lead to complex health issues. Many countries and regions have taken steps to control the concentration of Sb in aqueous solution. The Council of the European Union (CEC) and the United States Environmental Protection Agency (USEPA) had classified Sb and its compounds as priority pollutants, established the maximum admissible concentration of Sb in drinking water as 6 and 10 µg/L, respectively. China had set threshold concentration of Sb in drinking water to 5 µg/L, which is even more stringent than the international emission standards (He et al., 2012).

Sb exists in the environment mainly in four oxidation forms [Sb(-III)/Sb(0)/Sb(III)/Sb(V)]. In aquatic environments,

inorganic species Sb(III) and Sb(V) comprise the two predominant valence states, Sb(III) mainly exists in reducing environments, but Sb(V) generally exists in oxidizing environments, which highly depend on the prevailing pH-Eh and ionic conditions (He et al., 2012). Inorganic Sb compounds are found to have stronger toxic effects than organic forms. Long-term exposure and incessant consumption of Sb-rich aqueous solution cause various types of chronic and acute diseases.

Based on the extensive literature investigation, we found that Sb removal technologies are mainly divided into adsorption (Xi et al., 2013; Qi and Pichler, 2016; Qi et al., 2017), coagulation/flocculation (Du et al., 2014; Guo et al., 2009; Wu et al., 2010), electrochemical technology (Song et al., 2015; Di et al., 2017), membrane technology (Kang et al., 2000; Du et al., 2014), ion exchange (Riveros, 2010), etc. In general, the membrane technology and ion exchange achieves a high removal efficiency, however, initial investment and operating costs prevented the development of the technologies. Coagulation and electrochemical technologies are the most practical methods, nevertheless, they carry the risk of secondary pollution. In this review, we focus on the adsorption method and adsorbent's development, which roughly classified into inorganic adsorbents, organic adsorbents, clay and soil constituents, compound adsorbents, etc. Many adsorbent materials are sensitive to environmental factors, therefore, environmental conditions of the aqueous solution affects adsorption capacity and regeneration performance. Recently researchers have focused on several aspects of adsorbents, such as low-cost, rapid reaction, high selectivity and efficiency, reusability, ease of operation and maintenance, availability and flexibility.

In this review, we attempt to summarize Sb removal technologies. The main topics of this review are presented in the following sections: (1) Overview of the chemistry, speciation and distribution of Sb in aqueous solution; (2) Comprehensive summary of the current main achievements, providing references for further investigation; (3) Critically analyzed the salient features and removal mechanisms, evaluating benefits and limitations of these technologies;

(4) We considered the Fe- or Mn-based technologies was the most promising technique to remove Sb for field application.

1. The chemistry, speciation and distribution in aqueous solution

1.1. Antimony chemistry and speciation

Antimony is a metalloid with atomic number 51 and an atomic mass 121.76 that possesses $5s^25p^3$ outer orbital electron configurations (Miao et al., 2014). Sb, an amphoteric element, occurs in natural waters and exists mainly in particulate and soluble forms. Sb is most problematic to the ecosystems due to its relative mobility over a wide range of pH-Eh conditions (Herath et al., 2017). Soluble Sb commonly occurs in four different oxidation states [Sb(-III)/Sb(0)/Sb(III)/Sb(V)] (Li et al., 2018). Its solubility, toxicity and other properties strongly depend on the pH, redox potential environment (Kong et al., 2015). Inorganic species Sb(III) and Sb(V) comprise the two predominant valence states, which are frequently found in aerobic surface water and anaerobic groundwater, respectively (Filella et al., 2002). Sb(V) generally present stable in oxygenated systems, whereas Sb(III) regularly present in anoxic media (groundwater or pore water, etc.). The oxidation state has a critical influence on the transformation and immobilization of Sb, it found evidence that Sb(V) may be more soluble and mobile than Sb(III) (Johnson et al., 2005; Wu et al., 2011).

Apart from redox conditions, the pH value is another important factor influencing the form of Sb. Sb(III) exists in the form of $Sb(OH)_3$, $SbO(OH)$ or $HSbO_2$ in environmental conditions of pH 1.0–11.0, while under extreme acidic conditions (pH < 1.0) Sb(III) exists in the positively ionic form SbO^+ or $Sb(OH)_2^+$ (Bergmann and Koparal, 2007). Under strong alkaline conditions (pH > 11.0) the prevalent forms of Sb(III) are negatively charged ions SbO_2^- or $Sb(OH)_4^-$. The species of Sb(V) exist dominantly as $Sb(OH)_6^-$ in environmental conditions of pH 1.0–14.0, whereas under strong acidic conditions (pH < 0) the form of Sb(V) is SbO_2^+ (Pitman et al., 1957). The Sb species found in actual wastewater in Xikuangshan (Sb mine area in China) showed that the Sb(V) is the dominant species in aerobic water, mainly present in the form of $Sb(OH)_6^-$, while Sb(III) could be detected at trace concentrations in a small number of samples (Liu et al., 2010). The interference factors control Sb's mobility and transformation, such as Fe- and Mn-based minerals, humic acids (HAs), pH-Eh conditions, DOM and interfering ions. The solubility of Sb(III) is low, but it has stronger affinity than Sb(V) to hydroxides of iron or Mn, and HAs in the soil or sediments (Liu et al., 2015; Wu et al., 2010).

Antimony, a cumulative and genotoxic toxic element, which converted into organic species such as dimethylantimony (DMSb) and trimethylantimony (TMSb) (Bentley and Chasteen, 2002). Moreover, Sb, weak ecotoxicological effect, which presented serious threat to human health (Hammel et al., 2000). Soluble inorganic Sb species are easily combined with internal sulfhydryl groups in the human body (He et al., 2019). Moreover, Sb(III) easily reaches to critical biological targets, and retained for longer periods of time in organisms (Wang et al., 2017). Sb and its compounds can destroy the

ion balance in cells and cause cellular hypoxia (Yang et al., 2015). Inhalation exposure to antimonial has been reported to cause pneumonitis, fibrosis, bone marrow damage and carcinomas (Kang et al., 2000). Moreover, excessive long-term intake of Sb cause severe health implications including direct lung damage even induced cancer (Qi et al., 2017).

1.2. Distribution and pollution by Sb in aqueous solution

The distribution of Sb in aqueous solution has not been widely researched. In unpolluted natural water bodies, Sb occurs naturally at low concentration. Total dissolved Sb concentration in freshwaters and groundwater have been reported to typically range from around dozens of ng/L to a few $\mu\text{g/L}$ depending on the physical and chemical conditions of geographical location (Ulrich, 2000). The average concentration of Sb in untapped natural water usually less than $1.0 \mu\text{g/L}$ (Filella et al., 2002). The concentrations of Sb(III) and Sb(V) in the Yangtze River were in the range of $0.029\text{--}0.736 \mu\text{g/L}$ and $0.121\text{--}2.567 \mu\text{g/L}$, respectively (Wu et al., 2011). However, the vicinity of Sb ore fields such as active mines and smelter areas for ceramics and flame retardants, volcanic zones and geothermal springs, the concentration of Sb could reach up to several hundred $\mu\text{g/L}$, even mg/L , which is several dozen times higher than the maximum safe standards (Hu et al., 2016).

About some surveys, the level of Sb in geothermal groundwater was reported in the range $0.06\text{--}26 \mu\text{g/L}$ (Aksoy et al., 2009). Groundwater in the vicinity of abandoned Sb mines in Slovakia presented Sb concentration up to $9300 \mu\text{g/L}$ (Hiller et al., 2012). Rapid industrialization could be responsible for much higher Sb concentrations in water, especially in proximity to smelting, power plants and mining areas. China possesses the most abundant Sb reserves and contributes significantly to global anthropogenic emissions. The concentration of dissolved Sb in surface water in the vicinity of mining areas in China ranged from 4.58 to 29.4 mg/L (He et al., 2012). In the Qingfeng River near the world's largest Sb mine in the Xikuangshan area, the concentration of Sb reached $6384 \mu\text{g/L}$ (Wang et al., 2011). A study revealed that the Sb concentration in the river receiving seepage flow from ore residues near the Xikuangshan area was between 0.33 and 11.4 mg/L (Liu et al., 2010). Most of the nonferrous metal minerals are associated with Sb (Rakshit et al., 2011). The manufacturing and consuming of products containing large quantities of Sb, such as mining, ore-dressing, smelting, polyethylene terephthalate (PET)-production (Vengris and Selskis, 2010; He et al., 2012). Anthropogenic activities lead to elevated Sb concentrations in surrounding local water bodies (Hu et al., 2017). In summary, it is imminent to remove Sb in contaminated water.

2. Current removal technologies for Sb from aqueous solution

Due to increasingly stringent emission standards, many scholars have developed different technologies to remove Sb from contaminated water. Promising technologies mainly comprise physicochemical methods, which include adsorption, coagulation/flocculation, electrochemical technology,

membrane technology and ion exchange, which depicted in Fig. 1.

2.1. Adsorption

During the course of the past few decades, adsorption played a key role in heavy metal removal. Recently, a considerable number of adsorbent materials have developed for removal of Sb from contaminated water. Adsorption is considered as one of the most attractive techniques because of its widely used, cost-effectiveness, easy operation and handling, minimal sludge production, reusability, high removal efficiency, etc. (Rahaman et al., 2008; Singh et al., 2015).

The adsorbents of Sb are broadly classified into inorganic adsorbents, organic adsorbents, compound adsorbents and so on. Some minerals comprise the most important environmental sorbents, controlling the mobility of dissolved Sb species in surface water, groundwater, soil and sediments. Different kinds of materials mainly based on Fe-based sorbents, Mn-based sorbents, aluminum oxides, titanium oxides, carbon material, biosorbents, and compound adsorbents, which possess a large surface area and small particle size (Guan et al., 2015; He et al., 2012; Ungureanu et al., 2015; Yan et al., 2016).

In this section, we discuss and summarize properties of these adsorbents, adsorption capacity, adsorption isotherms, contact/equilibration time, regeneration performance and environmental factors, etc. The environmental factors controlling adsorption are particularly include the Eh-pH, HAS, organic and inorganic ligands (e.g. hydroxyl, carboxyl, amino), temperature and so on, all of these factors directly affect the adsorption. The adsorption capacity and adsorption rate highly depends on the characteristics of the adsorbents (e.g. particle size, surface area, isoelectric point), speciation and initial concentration, optimum pH value, temperature, and interference ions (e.g. HAS, PO_4^{3-} , SO_4^{2-} , SiO_3^{2-} , CO_3^{2-} , Ca^{2+} ,

Mg^{2+} , Mn^{2+}) (Ungureanu et al., 2015). The attributes of different adsorbents are more specifically discussed in the following text and the adsorption capacity and performance of different adsorbents were list in supporting materials (Appendix A Table S1).

2.1.1. Inorganic adsorbents

2.1.1.1. Fe based oxides. Over the past decades, Fe based oxides have been extensively applied to remove contaminants, due to their intrinsic properties of easy production, low cost, non-toxicity, eco-friendliness, abundance and low maintenance (Sun et al., 2016). Iron-based oxide was successfully developed as a novel adsorbent. The Fe oxide used to adsorb Sb is predominantly composed of goethite (Xi et al., 2013; Xi and He, 2013), akaganeite (Kolbe et al., 2011), hematite (Wu et al., 2010), amorphous Fe (FeOOH) (Xu et al., 2011), magnetite (Verbinnen et al., 2013), zero-valent Fe (ZVI) (Guo et al., 2016), etc. These Fe-based oxides are widely distributed in a variety of aquatic systems, soils and sediments. They play a vital role in immobilizing toxic-elements through redox reactions, adsorption and precipitation processes.

The large specific surface area and outstanding performance aids the Fe oxides to act as a sorbent, precipitator to immobilizing the contaminants. Generally, it is accepted that the mechanism of Sb removal by Fe oxides involve ion exchange, reduction, specific adsorption, surface precipitation and co-precipitation (Guan et al., 2015). Abundant studies had demonstrated that Fe oxides or hydroxides have higher affinities to Sb(III)/(V). Previous study indicated Sb(III) adsorbed was slowly oxidized to Sb(V) (Guo et al., 2014). The mechanisms of Fe-based oxide removal of Sb(III)/(V) is presented in Fig. 2.

Goethite was discovered to be a prominent sorbent for the immobilization of Sb(III)/(V). Xi and He (2013) used goethite to remove Sb(III) and Sb(V) from aqueous media, the results

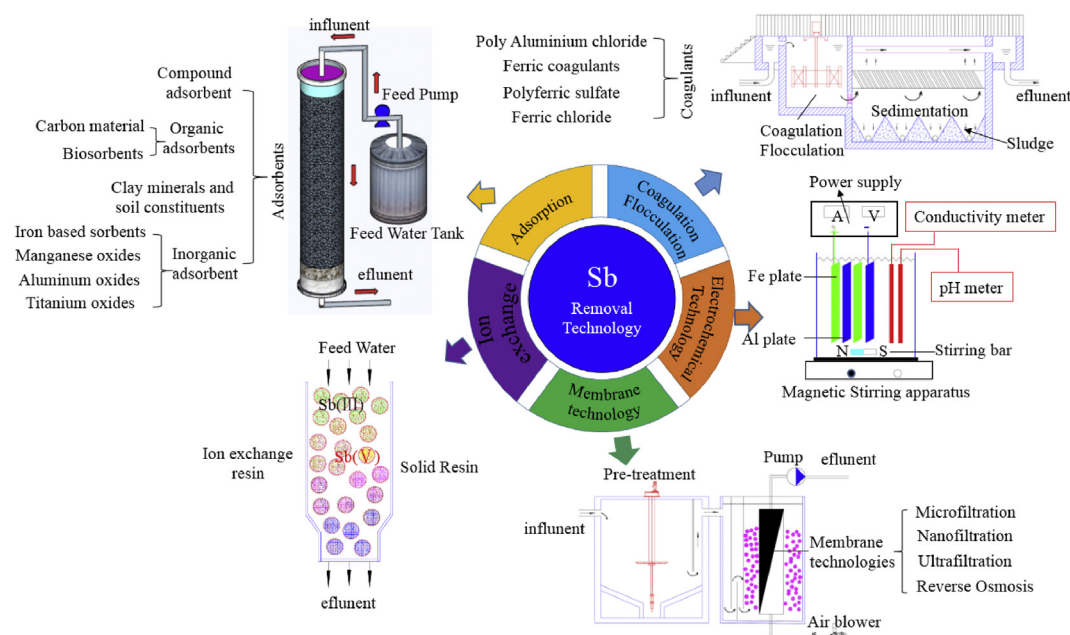


Fig. 1 – Schematic diagram showing various technologies used for removal of Sb from polluted water.

show that adsorption equilibria for both Sb(III) and Sb(V) are achieved within 24 hr. The pH values were the main factors influencing adsorption performance. Leuz et al. (2006) found that both Sb(III) and Sb(V) form inner-sphere surface complexes at the goethite surface. Sb(III) strongly adsorbs on goethite over a wide pH range (pH 3–12), yet the maximum adsorption capacity for Sb(V) is below pH 7.

Kolbe et al. (2011) showed that akaganeite could adsorb Sb(V) and As(V) in acidic conditions. Belzile et al. (2001) confirmed that amorphous Fe- and Mn-oxyhydroxides present in natural waters and sediments could play a detoxifying role by adsorbing and oxidizing Sb(III) to Sb(V).

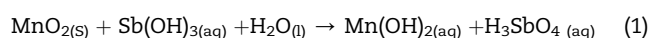
Some magnetic Fe oxides have attracted much attention because of convenient solid-liquid separation properties and their effective adsorption performance. Qi et al. (2017) synthesized Ce^{III}-doped Fe₃O₄ magnetic particles for efficient removal of Sb from aqueous solution. Wang et al. (2016) synthesized magnetic sludge (MS) by a facile co-precipitation method, which efficiently and inexpensively simultaneously removed both As and Sb. Shan et al. (2014) synthesized hematite-coated magnetic nanoparticles which showed great adsorption performance for Sb(III) (36.7 mg/g) with high adaptability and reusability.

Zero-valent Fe (ZVI) usually found in waterlogged or flooded soils, which converted to the activated zero-valent Fe containing both ferrihydrite and goethite distributed on its surface (Wilson et al., 2010). Mishra et al. (2016) synthesized zero-valent Fe decorated functionalized carbon nanotubes to remove Sb(III) and Sb(V). The carbon nano-tubes CNT-Fe(0) possess high surface area, and the maximum adsorption capacity of Sb(III) and Sb(V) at 250 mg/g at pH 5. Dai et al., 2014 synthesized nanoscale zero-valent Fe (nZVI) to remove Sb(III) and Sb(V). Sb could be thoroughly removed with appropriate nZVI dosage in a duration of 90 min. Guo et al., 2016 used a simple combination of oxidants (i.e. KMnO₄,

NaClO₄, H₂O₂) with ZVI to successfully and rapidly remove of Sb from wastewater.

2.1.1.2. Manganese oxides. Manganese oxides (hydroxides and oxyhydroxides) widely exist in the natural environment, and are considered to be one of the strongest oxidizing agents. Their presence exerts control over speciation, mobility and bioavailability of many toxic elements (Wang et al., 2012). Manganese oxides play an important role in reducing the mobility of Sb via oxidation and adsorption mechanisms. Several previous studies have revealed the relevance of Mn oxides in immobilizing the Sb present in water, soil and sediments (Wang et al., 2012; Basu et al., 2014).

Manganese oxides include MnO₂ (Xu et al., 2011), manganite (Wang et al., 2012) and Fe–Mn binary oxide (Liu et al., 2015; Xu et al., 2011), which possess excellent oxidation and adsorption capacity. Belzile et al. (2001) reported the adsorption mechanism of Sb(III) by synthetic MnO₂ in anoxic environments. The reaction 1 is given as follows:



The mechanism of the adsorption process is similar to the Fe oxyhydroxides. However, the authors demonstrated that the capacity of Mn oxides in oxidizing Sb(III) was much higher than Fe oxyhydroxides. The adsorption of Sb(III) and the surface complexation reaction with Mn often take place through one Sb(III) on one Mn(IV) binding side.

Liu et al., 2015 synthesized Fe–Mn (3:1) binary oxide (FMBO) and investigated the potential of an outer sphere complex of Mn-oxide and FMBO incorporated with Mn²⁺. The mechanisms of Mn-based oxide and FMBO removal of Sb is presented in Fig. 3. The adsorption of Sb(V) on the outer-sphere surface of Mn-oxides complex occurred through the substitution of –O–Mn groups within ≡Mn–O–O–Mn by Sb(V),

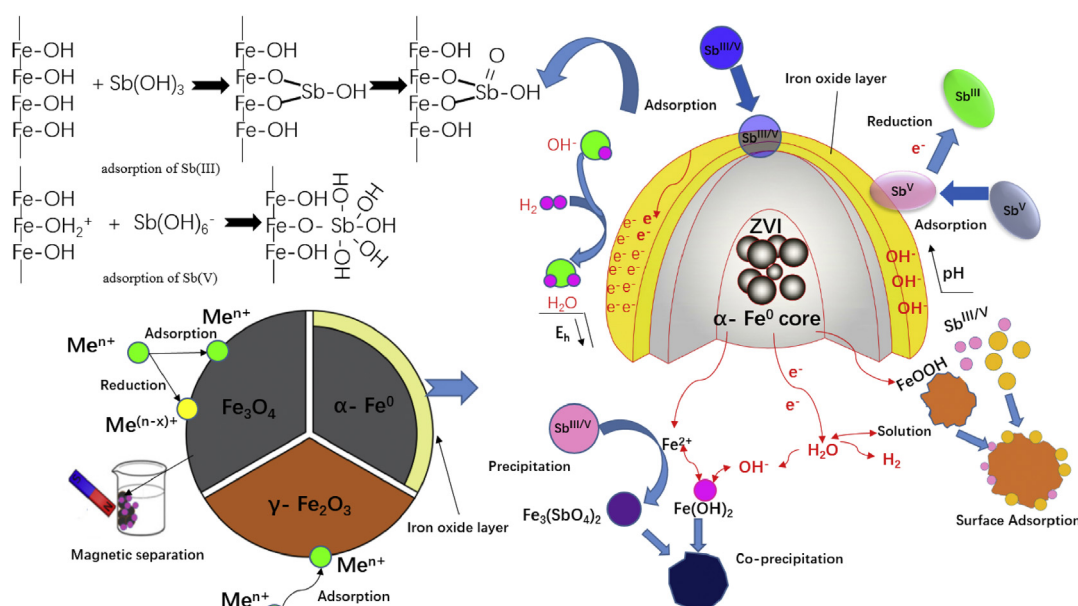
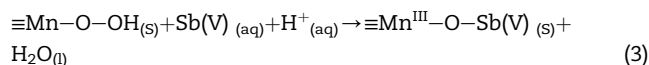
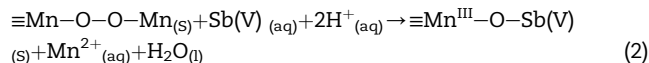


Fig. 2 – Schematic mechanisms of Sb reduction/adsorption/co-precipitation with iron based oxides (Belzile et al., 2001; Guo et al., 2014; Habuda-Stanić and Nujic, 2015; Liu et al., 2015).

thereby releasing adsorbed Mn^{2+} into the solution due to the competition between Sb(V) and Mn^{2+} ions for binding sites (reaction 2). Furthermore, the adsorption is attained by the formation of inner surface complexes and the replacement of hydroxyl groups, which attached on the Mn-oxide surface (reaction 3).



Sb(V) also formed inner-sphere complexes at the surface of manganite. Moreover, adsorption performance was independent of some common interfering ions in natural water, such as phosphate, sulfate, silicate, and carbonate, which made it superior to the Fe-based material (Wang et al., 2012). Zou et al., 2016 synthesized reduced graphene oxide coupled with Mn_3O_4 ($\text{RGO-Mn}_3\text{O}_4$) for the highly efficient removal of Sb(III) and Sb(V) from wastewater. The maximum adsorption capacity measured for $\text{RGO-Mn}_3\text{O}_4$ toward Sb(III) and Sb(V) are 151.84 and 105.50 mg/g at pH 2.5–11.5, respectively.

Wang et al. (2012) demonstrated that the adsorption capacity decreased along the sequence $\text{MnOOH} > \text{Al(OH)}_3 > \text{FeOOH}$. Xu et al., 2011 synthesized a novel Fe–Mn (3:1) binary oxide material (FMBO), which exhibited a much higher adsorption capacity (1.76 mmol/g) for Sb(III) than FeOOH (0.83 mmol/g) and MnO_2 (0.81 mmol/g).

2.1.1.3. Aluminum oxides. Few studies have reported interaction mechanism of Sb species with aluminum oxides, because Fe oxides are more efficient than aluminum oxides (Guo et al., 2009). In general, aluminum oxides used to adsorb Sb usually contain macro-/mesoporous amorphous alumina, activated alumina (AA), gibbsite, and Fe–Al double hydroxide, etc.

Activated alumina was usually used in fixed reactors to remove waste contaminants such as fluoride, selenium, organics and arsenic. Xu et al. (2001) used commercially available Activated Alumina to investigate the adsorption of Sb(V) . Sb(V) ions were likely adsorbed through electrostatic attraction or specific adsorption mechanisms. The optimum pH reported was 2.8–4.3. The addition of nitrate, acetate, arsenite, chloride, and silicate ions was resulted in negligible effects to the adsorption of Sb(V) , however, the coexisting ions (e.g. ascorbate, arsenate, phosphate, sulfate, EDTA, tartrate,

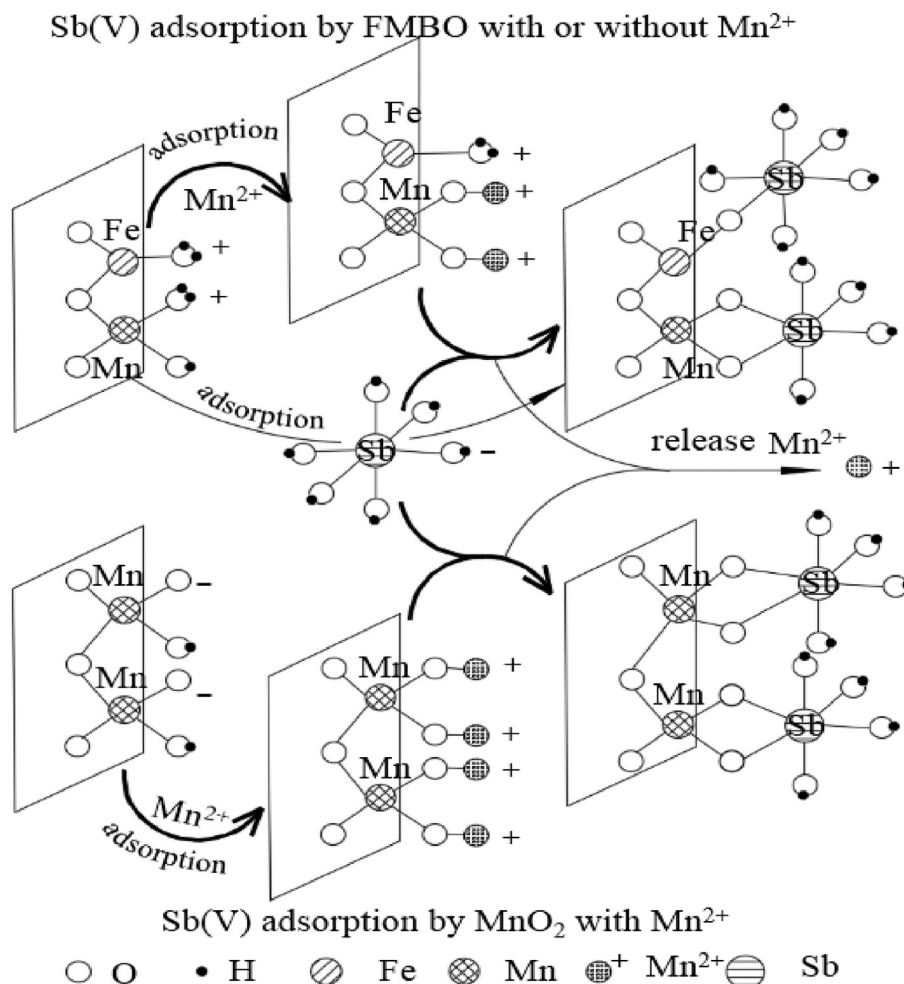


Fig. 3 – The adsorption of Sb(V) onto FMBO and MnO_2 by involved Mn^{2+} (Liu et al., 2015).

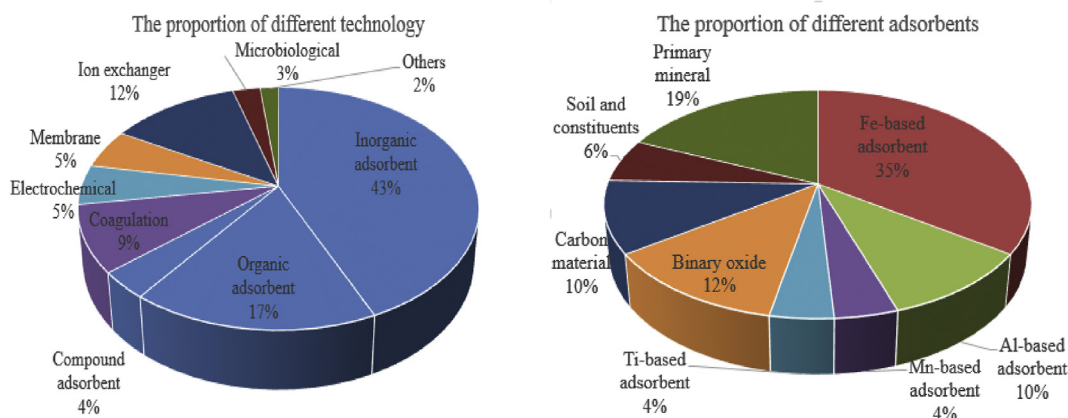
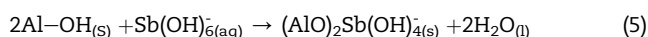
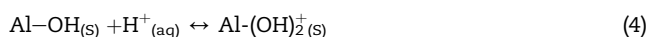


Fig. 4 – The proportions of reports on different technologies and adsorbents.

citrate) severely depressed the adsorption of Sb(V). Bullough et al. (2010) attempted to study the AA adsorption in the competitive system with Sb and As, and found that As(III) could promote the adsorption of Sb(III).

Gibbsite (Al-oxide) is a common mineral in soils, which has a strong interaction with Sb(V) by surface adsorption mechanisms (Rakshit et al., 2011). The surface complexation reaction for the sorption of Sb(V) on gibbsite can be formally expressed as the following reactions (Xu et al., 2001; Rakshit et al., 2011).



Moreover, a commercially available alumina has shown increased adsorption capacity for Sb(V). In acidic solutions, Al-oxides can be protonated, making the surface positively charged and generating electrostatic interactive forces with negatively charged Sb(V) species on the surface (Xu et al., 2001).

Kameda et al. (2009) synthesized the Fe–Al double hydroxide through the co-precipitation of a mixed solution of FeCl_3 and AlCl_3 . The reaction mechanism can be attributed to the ion-exchange ability of the Fe–Al double hydroxide and the adsorption of SbO_3 on the FeO(OH) surface. In addition, the authors also studied the adsorption of Sb on the Magnesium-aluminum (Mg–Al binary oxides) prepared from the thermal decomposition of Mg–Al double hydroxide at pH 10.5. The results show that the hydration of this material and Sb(V) formed a mineral with a structure similar to brandholzite. High concentrations of Cl^{-} and SO_4^{2-} had little effect on the removal of Sb(V). However, high concentration of CO_3^{2-} had a significant impact on the removal of Sb(V) (Kameda et al., 2011).

2.1.1.4. Titanium oxides. Titanium dioxides have many merits due to their unique properties, such as small particle size, large surface area, and strong adsorption ability (Yan et al., 2016). However, the adsorption of Sb by titanium oxide is limited and there are only few research articles. Titanium dioxides only deal with lower concentration Sb in wastewater

by batch and kinetic experiments performed under laboratory conditions.

Yang et al. (2017) used nanocrystalline titanium dioxide (TiO_2) to adsorb Sb(III) and Sb(V). The sorption studies showed that the Sb(V) sorption capacity and rate was greater than Sb(III). This tendency was opposite to that of the adsorption of Sb(III)/(V) by most Fe and Mn oxides. The maximum adsorption capacities of Sb(V) and Sb(III) were 588 and 333 mmol/kg, respectively. In addition, the reaction was found to be highly dependent on the presence of HAs and the pH of the aqueous solution. The paper provides insight into the adsorption of Sb(III)/(V) on {001}- TiO_2 . The kinetics results show that Sb(III)/(V) adsorption follows the pseudo-second order reaction, the adsorption capacity is 200 mg/g for Sb(III) and 156 mg/g for Sb(V). EXAFS results suggest that Sb(III)/(V) adsorption exhibited a bidentate binuclear surface complex. Nishad et al., 2014 synthesized a superior sorbent (nano-titania-crosslinked chitosan beads) to remove Sb(III)/(V). The composite beads exhibited high sorption efficiency, which demonstrated complete sorption of Sb from aqueous solutions with Sb concentrations ranging from 150 $\mu\text{g/L}$ to 120 mg/L at pH 2–10.

2.1.2. Clay minerals and soil constituents

Naturally occurring kinds of materials with respect to Sb adsorption in soil, such as kaolinite (Xi et al., 2010), bentonite (Xi et al., 2011), diatomite (San et al., 2010), the grey and red Erzurum clay (Targan et al., 2013), montmorillonite (Anjum and Datta, 2012), and Florisil (Zhang et al., 2011). Adsorption was predominately governed by a ligand exchange mechanism with surface structural OH_2 or OH^{-} groups at the adsorption sites (Jain et al., 1999). Such naturally occurring processes, particularly in water and soil environments, tend to control the transport of toxic contaminants. The adsorption of Sb by low-cost clay minerals from aqueous solution proved feasible, however, inefficient.

Xi et al. (2016) investigated adsorption behaviors of Sb(III) and Sb(V) on kaolinite by means of varying solution properties, such as the pH, temperature, ionic strength, initial concentrations and HAs content, etc. The authors found that adsorption of both Sb(III) and Sb(V) strongly dependent on the pH. The presence of competitive anions and HAs does not

have any obvious effect on the adsorption of Sb(III) on kaolinite. However, the adsorption of Sb(V) was strongly affected by ionic strength (Xi et al., 2010). The laws governing adsorption of Sb(III) and Sb(V) on bentonite were explored, the temperature between 278 and 323 K, pH 6.0, delivered adsorption capacities of Sb(III) and Sb(V) on bentonite in the range of 0.37–0.56 mg/g and 0.27–0.50 mg/g, respectively (Xi et al., 2011). Sari et al. (2010) found that the adsorption capacity of Sb(III) on diatomite reached 35.2 mg/g at pH 5.

Targan et al. (2013) studied the Sb(III) removal rate of Gediz River samples by using Grey and Red Erzurum clay. Results showed that the maximum removal rate of Sb(III) attained 72.6%. Some researchers studied the adsorption of Haro river sand, natural diatomite, and raw perlite to remove Sb. All of these are cheap adsorbent materials which exhibit selectivity for Sb (Sari et al., 2010).

2.1.3. Organic adsorbents

2.1.3.1. *Carbon material.* Activated carbon (AC), a commercial adsorbent, is most commonly applied to wastewater purification. AC has a microporous structure, huge surface area, and a high degree of surface reactivity, forming a versatile and efficient adsorbent. However, research papers about AC as an adsorbent to remove Sb present limited relevant data. The potential adsorption ability, mechanism, and reaction conditions is still unclear.

Navarro and Alguacil (2002) studied the adsorption performance of activated carbon for Sb in the copper electrolyte. The study shows that this adsorption reaction is exothermic. The mechanism suggests that the activated carbon loses electrons, becoming positively charged when interacting with water, the activated carbon is capable of adsorbing metal ions by electrostatic interaction.

Salam and Mohamed (2013) synthesized multi-walled carbon nanotubes (MWCNTs) to deal with real environmental samples containing Sb(III). The authors studied interference factors and kinetic models (pseudo-second-order kinetics). The adsorption of Sb(III) on the MWCNTs surface includes mechanisms of liquid film diffusion and further intra-particle diffusion. Leng et al. (2012) used graphene to remove Sb(III) from aqueous solution. The calculated adsorption capacity of graphene was 8.056 mg/g at pH 11.

The modified activated carbon is better than pure activated carbon in removing heavy metals. Thus, researcher attempted to improve the adsorption performance through metal-based materials and other modifications.

Yu et al., 2014 synthesized a novel materials Fe³⁺ modified granular activated carbon (GAC). The adsorption capacity of Sb(III) was 3.5 times than the original AC at pH 7.0 and 25°C. Thus, modification with Fe³⁺ increases adsorption capacity. Moreover, Yang et al., 2015 use graphene oxide (GO) to remove Sb from the aqueous solution. The result shows that GO is particularly effective for the adsorption of Sb(III) within a wide range of pH 2.0–10.0. Luo et al., 2015 synthesized carbon nanofibers decorated with zirconium oxide (ZrO₂) to remove Sb(III) and Sb(V) from aqueous solution. Zirconium oxide (ZrO₂)-carbon nanofibers (ZCN) were found to have maximum Sb(III) and Sb(V) adsorption capacities of 70.83 and 57.17 mg/g at pH 7, respectively. Moreover, it's more difficult to remove Sb(V) than Sb(III) from aqueous solution, because of its

negative charge in almost the entire pH range. Co-existing anions including nitrate, chloride and sulfate have no obvious influence on the removal of Sb(V), however, silicate, carbonate and phosphate present limitations at high concentrations (Zou et al., 2016).

2.1.3.2. *Biosorbents.* Biosorption is a potential technology for the removal of heavy metals from wastewater. Biosorbents are cheap, eco-friendly, and effective (Gadd, 2009). Their application for heavy metal removal had been adopted for many years, which divided into plant curing, plant fixation and plant accumulation (Bhatia and Goyal, 2014). Chitosan, cellulose, husk ash, chitin and starch have been used to remove Sb in wastewater (Li et al., 2018).

Agricultural waste is widely available at low cost (Sud et al., 2008). Many crops in mining areas accumulate high concentration of Sb in their edible parts (Wang et al., 2017). Some research showed that their adsorption capacity for heavy metals was better than that of activated carbon (Kurniawan et al., 2006). Rice husk contains hydrated amorphous SiO₂, which is similar to silicone, and has a good adsorption performance. Khalid et al. (2000) verified the rice husk's potential removal ability of Sb. Pyrochars was extracted from animal manure, which proved to effectively remove Sb(III) from aqueous solution (Han et al., 2017). Iqbal et al. (2013) carried out batch and fixed-bed continuous flow bioreactor experiments, which were more suitable for practical application, using the green bean husk for removal of Sb(III). Most biosorbents have the specific surface area and possess the relevant functional group (carboxyl, hydroxyl and amino groups), which plays a major role in the process of adsorption.

Biosorbent can be reused, and their mass loss was small during the desorption process, the regeneration performance of biosorbents was excellent. The main removal mechanism was the electrostatic and complexation interaction of Sb on the hydroxyl, carboxyl, and amino groups on the surface of the biosorbent. In addition, some biosorbents could oxidize Sb(III) to Sb(V), reducing the toxicity of Sb (Wu et al., 2012). Moreover, studies that attempted modifications showed that acidic treatment could improve Sb(V) adsorption significantly (Sun et al., 2011).

2.1.4. Compound adsorbent

Compound adsorbents based on modification techniques use inorganic solvent to modify the biosorbent or organic solvent to modify the inorganic adsorbent. Fan et al. (2014) pursued selective adsorption of Sb(III) from aqueous solution by an ion-imprinted organic-inorganic hybrid sorbent. The optimal pH range for adsorption of Sb(III) was 3.5–3.6. The imprinted sorbent had rapid kinetics for the adsorption of Sb(III) with saturation time of 20 min. The authors also synthesized an efficient mercapto-functionalized silica-supported organic-inorganic hybrid sorbent to remove Sb(III) from aqueous solutions by the batch technique. The adsorption of Sb(III) onto the mercapto-functionalized hybrid sorbent was independent of pH in the range from 3 to 8. The sorbent had a tendency toward higher adsorption affinities for Sb(III) compared to Sb(V). The resulting value for maximum adsorption capacity for Sb(III) was 108.8 mg/g. Thus, the functional groups on the

mercapto-functionalized hybrid sorbent provide good affinity for facile and fast adsorption to Sb(III) (Fan et al., 2016). Biswas et al. (2009) synthesized saponified orange waste (SOW), common anions (e.g. CO_3^{2-} , Cl^- , NO_3^- , SO_4^{2-}) had little influence on the adsorption, the adsorption mechanism was ligand exchange. Anjum and Datta (2012) synthesized montmorillonite modified by cetylpyridinium chloride (CPC) and cetyl trimethylammonium bromide (CTAB) to remove Sb(III). Miao et al. (2014) studied the removal of Sb(V) by HFO-201, hydrated ferric oxide (HFOs) was loaded on a polymeric anion exchanger (D201). Results showed that HFO coating on calcite was more effective than HFO-201, because the Ca^{2+} in the calcite was capable of enhancing the adsorption. Wang et al., 2015 used Fe-modified aerobic granules to remove Sb(V) from aqueous solution. Above all, the compound adsorbent has broad prospects, but many studies remain to be done, such as investigation of support materials, regeneration performance, and practical application.

2.2. Coagulation/flocculation

The mechanism in the coagulation/flocculation process is the destabilization of colloidal particles by coagulants due to changes in surface charge properties of the particles. Neutralization of charge in the particles eliminates the repulsive force acting among them and thus results in the particle agglomeration and enmeshment, and eventual precipitation due to gravity as the weight of the clumps overcomes the buoyant force acting on it. Flocculation is the capability of polymers to form bridges between larger mass particles, or flocs, and bind these together. Hydrolysis of the coagulant followed by the formation of a floc is a potential method for Sb removal from wastewater. Soluble Sb(III)/(V) could transformed into insoluble products by coagulants and precipitated, in a process that includes adsorption, interparticle bridging, precipitation and coprecipitation, etc. The solids are then removed by sedimentation or filtration (Inam et al., 2018).

At present, most commonly applied wastewater treatment is with aluminum and Fe-based salts as common coagulants, which are widely used in water treatment plants. These include aluminum sulfate [$\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$], and ferric chloride [$\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$] or ferric sulfate [$\text{Fe}_2(\text{SO}_4)_3 \cdot 7\text{H}_2\text{O}$]. All of them is considered to be the most important step in the water treatment process for contaminant removal owing to their efficiency and low-cost. Moreover, ferric coagulants are found to be more effective than aluminum coagulants in removing Sb on a weight basis over a wide pH range (Guo et al., 2009; Kang et al., 2003; Wu et al., 2010).

Kang et al. (2003) investigated polyaluminum chloride (PACl) and ferric chloride (FC) for Sb removal. Results showed an obviously higher Sb removal rate with FC than with PACl. The removal efficiency of FC could reach 94%. Guo et al. (2009) systematically studied the removal of Sb(III) and Sb(V) by coagulation/flocculation/sedimentation (CFS) by FC and aluminum sulfate (AS). The mechanism pointed out the removal of Fe and aluminum salts based on coprecipitation. The lattice structure of aluminum hydroxide has a low adsorption enthalpy resulting in lower removal rates of Sb(V) by aluminum salts.

Compared to Sb(V), effective Sb(III) removal was achieved even with less ferric coagulant over a wider pH range [the optimum pH for Sb(V) is (4.5–5.5) and Sb(III) is (4–10)]. Removal rate generally decreased with increase of pH. Guo et al. (2009) compare the behaviors of Sb and As during ferric coagulation process. The authors found that the removal of Sb(V) is more difficult than that of As(V), yet the removal of Sb(III) is more preferable to that of As(III). The removal efficiency order of various As and Sb species during FC coagulation at ambient pH values is $\text{As(V)} > \text{Sb(III)} > \text{As(III)} > \text{Sb(V)}$. Contrary to the removal of As, pre-oxidation is not suitable to Sb, which can lead to the opposite result. Therefore, keeping the water under reducing or anoxic conditions is more favorable for Sb removal (Kang et al., 2003; Guo et al., 2009; Ungureanu et al., 2015). Sb(III) removal is rarely affected by the presence of interfering ions such as phosphate and HAs (Guo et al., 2009), however, the removal rate of Sb(V) was markedly impeded by an increase of competing ions, including bicarbonate, sulfate, phosphate and HAs (Wu et al., 2010). The removal mechanism of Sb(III) included the formation process of hydrous ferric oxide (HFO) during coagulation and the anomalous incorporation of Sb(III) into HFO nanocrystalline and then coprecipitation. The mechanism of Sb(V) adsorption onto HFO occurred both at the surface and internally, which was the reasons why the removal of Sb(III) was effective under various suboptimal conditions, while the Sb(V) was highly sensitive to the presence of competing ions. Sb(III) removal efficiency increases with the increase of coagulant in the solution, among which particularly Fe increased precipitation. Sb(V) removal is influenced by the solution pH due to a change in Fe solubility" (Inam et al., 2018).

The coagulation/flocculation method remains the most effective treatment employed in experimental and field application. Coagulation/flocculation along with sedimentation-filtration was a more suitable method for Sb removal at higher concentrations in wastewater than other techniques. Gannon and Wilson (1986) found that Sb(III) is easily removed by co-precipitation with ferric hydroxide and the simultaneous adsorption of suspended colloid particles of ferric hydroxide and lauryl sodium sulfate. Vengris and Selskis (2010) use the coagulation technique to treat Sb-containing wastewater produced from the polyethylene terephthalate resin factory in Klaipėda city.

2.3. Electrochemical technology

The electrochemical method (EC) as an efficient treatment technology has been applied to treat numerous wastewaters, especially wastewater with high concentrations of pollutants, such as copper electrolytic refining, landfill leachate, textile, tannery, and petroleum refinery wastewater for removal of arsenic, fluoride, heavy metals and so on (Bazrafshan et al., 2015). Electrochemical technologies mainly have functions of coagulation, flocculation, flotation, oxidation and micro electrolysis, the sacrificial anodes form a range of coagulant species or metal hydroxides, which adsorb contaminants effectively through particle bridging and co-precipitation (Li et al., 2018; Vasudevan and Oturan, 2014).

Under optimal conditions (current density 2.58 mA/cm², pH 5.24, standing time 89.17 min, initial concentration

(521.3 µg/L) more than 99% Sb removed by electrocoagulation with hybrid Fe–Al electrodes (Song et al., 2015). Bergmann and Koparal, 2011 studied the removal of Sb (1500 mg/L and 3500 mg/L) from accumulator acid using electrochemical technology. Results showed that removal efficiency with a copper-graphite electrode was the highest. Zhu et al. (2011) treated the flotation wastewater, with high concentration of Sb (1500 mg/L), using electrocoagulation with an aluminum anode. The Sb concentration met local emission standards after treatment. However, the standing time was long: it took approximately 18 h to reach steady state. Moreover, Fe–Al electrodes exhibited higher removal efficiencies than Al–Al electrodes (Song et al., 2014). Di et al. (2017) found that Mn assisted the electrochemical generation of two-dimensional Fe–Mn layered double hydroxides (LDHs), which possess large internal surface areas with amphoteric surface hydroxyl groups. The LDHs removes Sb-pollutants efficiently.

The removal performance of Sb is influenced by many factors in electrochemical method, such as electrode materials, initial concentrations, pH, current density, duration, aeration intensity, interfering ions, etc. Many papers form consensus to conclude that higher current density, longer standing time, and lower pH values lead to greater removal rate (Bazrafshan et al., 2015; Bergmann and Koparal, 2007; Song et al., 2014; Zhu et al., 2011). The detailed performance of Sb removal by EC is summarized in Table 1.

2.4. Membrane technology

Membranes are selective barriers that block contaminants while allowing some selective constituents to pass through membranes. Membranes play an important role in chemical technology and used in a broad range of applications, especially the advanced treatment of potable water. However, the quality of membrane, constituent concentration, membrane electrical potential, flow rate, pressure gradient force, and chemical properties of the influent (temperature, pH, dissolved organic matter, etc.) influence removal efficiencies in membrane technology. MF and UF technologies function at low transmembrane pressure was not suitable for Sb removal, because Sb compounds may have smaller sizes than the membrane pores. But, hybrid system of coagulation-flocculation-ultrafiltration can increase the efficiency of the filtration process (Du et al., 2014). The integrated technology shows potential for application in water treatment (Ma et al., 2017).

Nishiyama et al., 2003 recovered Sb using a porous hollow-fiber membrane with a chelate-forming group iminodiethanol (IDE). Saito et al., 2004 investigated the removal of Sb(III) using polyol-ligand-containing porous hollow-fiber membrane which functionalized by N-methylglucamine (NMG) and 3-amino-1,2-propanediol (APD). The removal mechanism of Sb ions was described as when the solution containing Sb flows through the membrane, metal ions migrate faster than water molecules from the solution to the chelating group. These results also showed that the effect on pH depended on the polyol ligands, the maximum dynamic adsorption capacity reached 54 mg/g.

Du et al. (2014) used a hybrid process (CF-UF) high efficiency removal Sb(III) from contaminated surface water.

Table 1 – Summary of the remove performance of Sb removal by EC technology.

Oxidation state	Concentration	Electrode materials	Optimal pH	Optimum current density (mA/cm ²)	Interfering factors	Standing time	Removal rate	Ref.
Sb(V)	10–120 mg/L	Anode: cast iron Cathode: stainless steel	4.0–7.0	5.0–10.0	Mn ²⁺ (without) 0.5 mM Mn ²⁺	30 min	72%	Di et al. (2017)
Sb(III)	500 µg/L	Fe–Fe Al–Al	5.0–7.0	13.89	NO ₃ ⁻ SO ₄ ²⁻ PO ₄ ³⁻	40 min	99% 96.93%–99.31% 80.03%–89.31%	Song et al. (2014)
Sb(III)	521.3 µg/L	Fe–Al	5.24	2.58	–	89.17 min	98.47%–99.82%	Song et al. (2015)
Sb(V)	28.6–10.4 mg/L	Al–Al	2	166.7	–	5 hr	>99%	Zhu et al. (2011)
Sb(III/V)	3500 mg/L	Copper-graphite	–	1 2	–	5 hr/ 7 hr	96.7%–97.7% 93%/ 99%	Koparal et al. (2004)

Under optimized conditions, the removal rates of over 90% and residual Sb(III) concentrations less than 4.0 µg/L were achieved. The mechanism is described as the solutes (H_3SbO_3 or $\text{Sb}(\text{OH})_3$) being smaller than the membrane pore, most of Sb(III) diffuses across the membrane. During the CF-UF process, HFO–Sb particles formed and aggregated, rejected by UF. The synergistic effects of Fe-hydrolytic flocs and ultrafiltration membrane enhance Sb(V) removal. The authors found that Fe-based flocs integrated with an UF membrane showed a large potential advantage in removing Sb(V).

2.5. Ion exchange

Ion exchange is an important method for removal of heavy metals from wastewater. In most cases, ion exchange is used to denote processes of purification, separation, and decontamination of water and other ion-containing solutions with solid polymeric. Typical ion exchangers are ion exchange resins (functionalized porous or gel polymer), zeolites, montmorillonite, clay, and soil humus. Ion exchangers are either cation exchangers that exchange positively charged ions (cations) or anion exchangers that exchange negatively charged ions (anions).

Ion exchange was applied in related areas of Sb, such as the separation of Sb from copper electrorefining (Riveros, 2010), the analysis of different Sb oxidation speciation (Ozdemir et al., 2004). The study of Sb exchange behavior on anion exchange resins showed that the removal rate of Sb(III) was higher than Sb(V), while it was more difficult to elute Sb(III) than Sb(V) (Guin et al., 1998). The ability of elution is very important because it relates to the regeneration performance of the exchanger. Riveros (2010) focused on the elution of Sb(V) from amino-phosphonic resins. The result showed that adding thiourea to HCl could significantly increase the elution efficiency of Sb from the resin, because thiourea could reduce Sb(V) to Sb(III), and Sb(III) was easier to elute.

Cunningham et al. (1977) proposed and patented a method for the removal or recovery of dissolved Sb from an industrial stream using a strong acid or a strong base ion exchange resin. The authors found that a strong acid cation exchange resin in the hydrogen form is highly effective in removing Sb from an acidic solution, while a strong base anion exchange resin in the hydroxyl form is highly effective for removing Sb from a less acidic or basic solution. The resin can be regenerated using strong bases and acids.

Some researchers removed Se(IV), Se(VI), Sb(III) and Sb(V) from solutions using Duolite A7 (a type of commercially available weakly basic anion exchanger) functionalized by secondary amino groups. In addition to the organic ion exchanger, inorganic ion exchangers, such as magnesium–aluminum and copper–aluminum layered double hydroxide were also interested to other researchers (Kameda et al., 2012). Their studies were mostly focused on the mechanisms. Results showed that $\text{Sb}(\text{OH})_6^-$ could exchange with the NO_3^- in the interlayer of Mg–Al and Cu–Al LDH. Compared to Cl^- , CO_3^{2-} , and SO_4^{2-} , NO_3^- as the interlayer ion was the most effective for removal of Sb(V).

2.6. Other technologies

Presently, researchers focus on extraction methods for Sb removal. The liquid/liquid extraction technique is used to separate Sb from another metal in a mixture solution. Sb is extracted by repeated extraction, however, which is not sufficiently documented. Hu et al. (2016) used a pH-static leaching method, leaching of Sb from three Sb ore (Tongkeng Sb Mine, Muli and Banxi), which proved effective. Then, toxic-Sb was extracted from Sb minerals, which showed that Sb extraction results are related to the type of ore, species of Sb and dependent on the pH value. Moreover, Stewart and Mosheim (1985) used methyl isobutyl ketone as solvent to separate Sb from the columbium fraction.

2.7. Summary of the above technologies

Several types of technologies are reported in the literature reviewed above. Clearly, researchers focus on the removal technology of inorganic adsorbent. The Fe- or Mn-based adsorbent makes up large proportion of inorganic adsorbent, which depicted in Fig. 4.

At present, these adopted technologies are used in varying degrees. All of them have their salient features, advantages and disadvantages. A comparative discussion on conventional Sb removal technologies was summarized in Table 2.

3. Conclusion and prospectations

Present review shows that removal of Sb from aqueous solution have been considered of particular interest and concern for scientist. Nowadays, adsorption, coagulation/flocculation, electrochemical technology, membrane technology, ion exchange are used in varying degrees. Nevertheless, due to complex properties of metalloid-Sb including variable valence states and easily mobility, it is not possible to select one best technique, since each one has advantages and drawbacks. Adsorption technique is considered quite efficient in the removal of trace contaminants but the regeneration, recycling and reuse prevent its application. Coagulation/flocculation is considered quite efficient in the removal of high concentration contaminants but the toxic by-products and large amount of coagulant as the disadvantages. The blocking of membrane is the main disadvantage of the membrane technology. Economic investment and sludge discharge are important problems to be overcome for the electrochemical technology in field application. Therefore, it is urgent to develop the hybrid and efficient technologies simultaneous removal of high or low concentration wastewater of Sb. From the recently extensive literature, we would like to emphasize the following aspects: (1) Sb is a toxic element, which causes acute and chronic diseases, even cancer, but the specific bioavailability and toxicokinetics still unclear. (2) Many comprehensive papers focused on the adsorption isotherm and kinetics, dynamic search, equilibrium, kinetics in simulated water. However, it is unquestionably important to assess the real performance of the adsorbents in wastewater, and researchers should pay more attention to commercial-scale wastewater treatment.

Table 2 – Summary of salient features, advantages and disadvantages of Sb removal techniques.

Removal techniques	Salient features	Advantages	Disadvantages	Ref.
Adsorption	(1) Antimony species are adsorbed either physically or chemically onto the solid surface of adsorbents; (2) Sb compounds can be attracted by surface charge through electrostatic action, and then form monolayer or multilayer adsorption on the surface, some would form adsorption complexes accompanied with oxidation-reduction reaction; (3) Adsorbents provide Sb binding sites of amine, hydroxyl, thiol, methylated pectin, –SH.	(1) Large specific surface area; (2) Simple to use and easy operation; (3) Commercially available sorbents; (4) Cost-effective and low secondary pollution risk; (5) Excellent handle in low level of wastewater	(1) Regeneration, recycling and reuse of adsorbents is a significant problem; (2) Sensitivity to the water quality, such as Eh, pH, HA, interference ions, etc.; (3) Not extensively used in actual wastewater treatment	(Fan et al., 2016; Ungureanu et al., 2015)
Coagulation/flocculation	(1) Coagulants destabilize Sb compounds in water, which leads the Sb compounds to aggregate; (2) Coagulation is mainly based on the internal and surface adsorption coupled with coprecipitation; (3) The aggregated mass precipitates into the water and filtered.	(1) Low cost; (2) Simple to use and easy operation; (3) Readily available chemicals; (4) Efficient; (5) Mature and widely used in field application; (6) Applied on the treatment of high concentration wastewater	(1) Requires long time and extensive setup; (2) Ineffective for low concentrations of Sb; (3) Concentrated sludge production, generates large amounts of Sb sludge; (4) Secondary pollution risk	(Guo et al., 2009; Song et al., 2015; Inam et al., 2018)
Electrochemical technologies	Electrochemical method sacrifices anode to form active coagulant precursor, and then forms coagulation body, finally removes Sb by coagulation body's adsorption and co-precipitation.	(1) Easy to operate; (2) Generate less waste residue; (3) The secondary pollution risk was low. (4) Treated wastewater contained high concentration of Sb	(1) Power consumption; (2) Financial cost; (3) Operation parameters need to optimized	(Di et al., 2017; Song et al., 2014, 2015)
Membrane technologies	Membranes are selective barriers which block contaminants while allowing some selective constituents to pass through them to deliver purified water.	(1) Highly efficient; (2) No chemical required; (3) Ability to completely purify water; (4) Applied in depth treatment	(1) High initial investment; (2) High operating cost; (3) High pollutant concentration water disposal; (4) Membrane contamination, clogging and decay of membrane; (5) Risk of sudden failure	(Du et al., 2014; Ma et al., 2017)
Ion exchange	The mechanism of ion exchange is similar to adsorption, oxyanions of Sb are adsorbed or replaced with the ions of synthetic resins.	(1) High efficiency; (2) Less generated sludge	(1) High initial cost; (2) Sensitivity to water quality; (3) Contamination of resins, blogging and the risk of sudden failure	(Riveros, 2010, Yangyang et al., 2014)

(3) Most current researchers focused on the basic static behavior of adsorbents, such as high adsorption capacity, short equilibrium time, low-cost, low sensitivity to influence factors, and excellent regeneration performance. The leachability studies should be tested under different experimental conditions. (4) The Fe- or Mn-based material, binary (multi-) oxides, modified and composite adsorbents, and easy solid-liquid separation, may become the promising technique. (5) Urgently to developing portable and accurate testing kits for detecting environment Sb, develop hybrid and efficient technologies to fill the gap between theoretical research and practical applications.

Declaration of interests

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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Appendix A. Supplementary data

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