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Original Research

In-situ Stability and Remediation of Typical Antimony (Sb) Tailings by Combined Solid Waste in Southwest China

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Abstract

To explore the feasibility of reusing solid waste to stabilize high-content arsenic (As) and antimony (Sb) tailings, red mud, fly ash, dried sludge, ferrous sulfate ($FeSO_4$), and rice husk ash (RHA) were used as the stabilizer to stabilize the Sb tailings in Qing Long and Du Shan. The combined treatment with 5% red mud, 10% fly ash, 5% dried sludge, 1% $FeSO_4$, and 1% rice husk ash had the best stabilization effects on As and Sb, the leaching concentrations of As and Sb decreased considerably under neutral conditions, this indicates that the leaching behavior of As and Sb is controlled by the alkaline and acid-retarding capacity of the materials. In addition, the leaching of heavy metals decreases with the formation of (C-S-H) and calcite ($CaCO_3$), indicating that heavy metals exist in the form of metal hydration or hydroxide and precipitate on the surface of calcium silicate hydrate (C-S-H) and calcite particles. Leaching of heavy metals in stabilized materials can be considered as a pH-dependent and control process of stabilization products.

Keywords: antimony tailings, *in-situ* stabilization, leaching toxicity, morphological of occurrence, bioavailability

Introduction

Tailings are solid wastes produced in mining activities. Some heavy metal elements (Cr, Cd, Pb, Zn, Cu and Ni) and radioactive elements are prone to oxidation and ion release under natural conditions [1, 2]. At present, tailings disposal methods mainly include surface tailings storage or underground filling. In the karst region, due to rich aquifer, fragile ecological environment [3], it is easy to be polluted by pipes, fractures, well drilling and falling caves [4]. Rapid hydrology flow through special karst structure with high permeability [5], resulting in more serious heavy metal pollution problems than in non-karst areas. The direct burial of tailings pond will lead to large numbers of heavy metals infiltrating into the groundwater along with the tailing slurry [6], and has a long time of continuous spreading [7]. Therefore, the vulnerability of karst areas makes the prevention and control of tailings

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pollution difficult and challenging (cost and complex investigation procedures etc.).

The Qing Long antimony mine is predominantly carbonate rock with underground karst pipelines [8], heavy metals can easily enter the surrounding water and soil media. The clay rock contained in the minerals is easily leached and the heavy metal is easily transported over long distances, resulting in serious pollution levels and high potential ecological risks [9]. The accompanying fractures of Du Shan antimony ore in Du Shan County, Guizhou Province not only activate, migrate, and enrich the metallogenic elements in the ore source layer but also antimony ore accumulation. Drainage-irrigated threatens farmland and inhabitants [10]. Stabilization is the process by which the waste is mixed with an appropriate additive to bind contaminants, convert the toxic waste into a more physically and chemically stable form, that is, to produce a less toxic or fluid form, although stabilization techniques have been used for a long time, the chemical morphology and binding mechanism of metal ions and composite stabilizers have not been fully characterized.

Previous studies have more focused on the stable adsorption of heavy metals such as lead (Pb), zinc (Zn), cadmium (Cd) and As in soil and water by modified stabilizer [2, 11], while there were few studies on combined pollution of As and Sb (presence of oxyanion) under long-distance migration, the existing studies were lacking: (1) the inorganic and organic polymer system of stabilizer-toxic tailings was used to verify the stabilization mechanism of As and Sb, (2) to evaluate the optimal proportion based on the occurrence form of metals to direct and potential ecological toxicity, (3) By making use of fly ash with high activity, strong adsorbability, and the stable, highly alkaline environment provided by red mud, to explore the redox release process of As and Sb controlled by Fe/Al hydroxide.

Based on the concepts of high efficiency, low cost, and waste treatment, the study took advantage of the characteristics of alkali, high activity and abundant Iron oxides of red mud and fly ash to stabilize the high concentration tailings. Screening the best stabilizer ratio, to achieve the removal and repair of Sb and As. The main objectives were to 1) investigate the influence of stabilization under different application rates, 2) gain insight into the stabilization effect of Sb and As in tailings with inorganic solid and organic waste and 3) elucidate the mechanisms of removal of Sb and As, this provides a reference for the long-term leaching mechanism and chemical leaching behavior of As and Sb of tailings in karst areas, and solves the problems of large tailing accumulation and heavy metal migration during mining.

Materials and Methods

Experimental Materials

Tailing samples were collected from the Qing Long and Du Shan antimony mines in Guizhou Province. Du Shan antimony mine is under exploitation and utilization, its tailings have been treated accordingly and vertical distribution is relatively uniform, so one tailing sample was collected by mixing point sampling method. Qing long antimony mine is currently out of production, there are differences of tailings accumulation. Therefore, 12 surface samples of tailings were selected by the five points method and evenly mixed.

Red mud, fly ash, dried sludge, and rice husk ash were purchased from the Internet. $FeSO_4$ was excellent grade pure. The physical and chemical properties of the stabilizers and tailings are listed in Tables 1-4.

Du Shan antimony tailing is weakly alkaline with a pH of 8.2. The content of As, Sb exceeded 5.8, 158.4 times of the soil background value of Guizhou Province, respectively. Qing Long antimony tailing is medium-weak alkaline with a pH of 7.4, As and Sb content exceed 20.26, 636.8 times of the local soil background value, respectively (Table 1).

The content of As, Sb of red mud and fly ash is lower, the pH are11.62, 10.27, respectively. The pH of dried sludge is weak acid-neutral, organic matter content is 421.34mg.kg^{-1} (Tables 2-3).

The contents of main components in red mud and fly ash are shown in Table 4. The main components of fly ash are Al_2O_3 and SiO_2, the red mud are Al_2O_3, Fe_2O_3, SiO_2.

Experimental Design

According to Table 5, add stabilizer and appropriate amount of deionized water into 100 g of dried and sifted tailings, stir thoroughly, stir evenly, seal the bottle with safety film, and maintain in a dry and ventilated place for 60 days. After curing, the tailings were air-dried, ground evenly and put into the sample bag for use.

Table 1. Basic physical and chemical properties of tailings.

Heavy metal content (mg.kg^{-1})	Sb	As	Pb	Cd	Zn	Cr	Organic matter	pH
Du Shan-D	354.39	111.76	18.42	0.82	90	58.6	6.56	8.2
Qing Long-Q	1426.43	405.26	62.17	0.55	60.17	69.76	26.84	7.4
Background values^1	2.24	20.00	36.5	0.08	88.8	95.9		

Background values^1 refer to the background values of heavy metals in soil from Guizhou province.

Table 2. Parameters of stabilization agent.

Stabilization agent	pH	Heavy metal (mg.kg^{-1})				
		As	Cd	Pb	Sb	Zn
Fly ash	10.27	39.39	1.614	71.63	0.58	113.9
Red mud	11.62	81.09	0.5118	44.401	1.63	89.03
Rice husk ash	7.63	-	-	-	-	-

Table 3. Basic physicochemical characteristics of dry sludge.

Material	pH	Heavy metal (mg.kg^{-1})				Essential nutrient (mg.kg^{-1})			
		As	Cd	Pb	Zn	Organic matter	TN	TP	TK
Dry sludge	6.63	22.5	1.53	70.23	98.87	421.34	23.65	11.67	13.02

Table 4. Main chemical composition (%).

Stabilization agent	Al_2O_3	Fe_2O_3	MgO	CaO	SiO_2	Na_2O	TiO_2	SO_3	P_2O_5	K_2O
Fly ash	34.86	2.52	0.779	2.63	54.94	0.475	-	0.313	0.232	1.76
Red mud	21.12	37.26	1.433	5.045	13.38	4.28	2.3	-	-	-

Table 5. Formulation of stabilization treatment.

Treatment groups	Red mud (%)	Fly ash (%)	Dry sludge (%)	Rice husk ash (%)	$FeSO_4$ (%)
CK	0	0	0	0	0
1	5	5	0	0	0
2	5	10	5	0	0
3	5	10	5	1	0
4	5	10	5	0	1
5	5	10	5	1	1

CK refers to treatment without adding stabilizer

After curing, the naturally dried tailings were weighed and carried out leaching experiment and morphological extraction experiment to explore the leaching behavior and morphological changes of metal, screen the best stabilization scheme. The leaching concentration and morphology of arsenic and antimony were determined by ICP MS (PerkinElmer Nex ION 350, US), the minimum detection limit is 0.08~0.12 μg·L^{-1}.

Determination Method

Physical and chemical properties: the pH value of tailings and stabilizers was determined with a water-soil ratio of 2.5:1, and the organic matter was determined by a high-temperature external thermal potassium dichromate oxidation-volume method. The chemical composition of the tailings and stabilizer are analyzed by X-ray fluorescence (XRF) analysis (AXS S4 Pioneer, Bruker Co., Germany).

Mineral analysis: mineral composition of tailings was conducted by X-ray diffraction (Germany-Bruker-Bruker D8 Advance). Measurement conditions: operating voltage: 40 kV, operating current: 40 mA, copper target, wavelength 1.5406 angstroms, light source voltage: 50 kV, incident Angle >0.5, scanning range 5-90 degrees, test sample shape: powder; Ambient conditions: room temperature; Qualitative analysis software MDI Jade software (6.5) was used for qualitative analysis of the results after sample determination.

Leaching toxicity test: the leaching method adopts the "Solid Waste Leaching Toxicity Horizontal Oscillation Method". The leaching solution was deionized water. The liquid-solid ratio was L/S = 10, the mixture was shaken for 8 h at room temperature (25°), allowed to stand for 16 h, the oscillation frequency was 110±10 times/min, and the sample particle size was ≤3 mm.

Morphological extraction experiment: the EU improved BCR sequence extraction process was used to extract the heavy metals under the conditions of acetic acid (CH_3COOH), hydrogen peroxide (H_2O_2) hydroxylamine hydrochloride ($NH_2OH·HCl$) and ammonium acetate (CH_3COONH_4) to measure the morphology of heavy metals before and after treatment. The contents of weak acid extraction state (F1), reducible state (F2), oxidizing state (F3) and residual state (F4) were determined.

Data Analysis

All experiments in this study were set up as controls and repeated, the obtained data are the average values of each replicate. Excel and Origin software were used to process the data, jade software was used to fit the XRD diffraction pattern. Pearson correlation analysis was used to analyze the relationship between the physicochemical properties of the leaching solution and the metal leaching concentration.

Results

XRD Analysis

D-3, D-5, Q-5 and Q-3 treatment groups were selected for XRD diffraction pattern analysis (in Fig. 1(c-d)). During the stabilization process, the mineral phase content of tailings has little change. The main mineral facies of tailings in Du Shan are dolomite ($MgCaCO_3$), quartz (SiO_2), and silicaite ($Ca_3SiO_2O_7$) (Fig. 1a). The formation of new substances calcite ($CaCO_3$) and calcium silicate hydrate (C-S-H) was detected in D-5 and D-3 (Fig. 1c). Compared with untreated tailings, the diffracted intensity of $MgCaCO_3$ in D-3 and D-5 is weakened, which may be the dissolution of a small amount of $MgCaCO_3$ to produce Ca and Mg [12].

The mineral facies of tailing composition in Qing Long are mainly $CaSiO_3$, SiO_2, $CaCO_3$ (Fig. 1b). The stabilization products including Al_2O_3, C-S-H and $CaCO_3$ are detected in Q-3 and Q-5 (Fig. 1d). Al_2O_3 has a certain adsorption effect on Sb and As, the adsorption mechanism is mainly determined by spherical and surface complexation, hydrogen bonding and oxidation [13]. The diffraction peaks of Fe-As and Fe-Sb precipitated phases are not detected, which may be their poor crystallizability or amorphous state, or relatively small amount of precipitated phases [14].

Physical and Chemical Properties of Leachate

The physicochemical properties of stabilized tailings are shown in Fig. 2. The pH of tailings in the two mining areas increases significantly, by 0.96% in D-1 and 1.14% in Q-1 compared with the CK, respectively (Fig. 2(a-b)), which may be attributed to Al content, Fe, Ca in alkaline substances of red mud and fly ash [15]. At treatment group 5, the pH is the lowest, which may be attributed to the neutralization of potassium (K), phosphorus (P), calcium (Ca), magnesium (Mg), silicon (Si) in rice husk ash (RHA) and dried sludge [16]. In addition, the pH value of the system reduces by $FeSO_4$.

The Eh of the tailings in Du Shan (Fig. 2c) and Qing long (Fig. 2d) increases in treatment groups 1 and 4, respectively. In treatment group 1, the Eh in Qing Long and Du Shan tailings increases by 47 mv and 71 mv. With the addition of dry sludge, the Eh of tailings in treatment group 2 decreases somewhat. The comparison of treatment groups 3, 5 and 2, 4 shows that the addition of 1% $FeSO_4$ is beneficial to the increase of Eh of tailings.

Conductivity of electricity (EC) characterizes the amount of salt in a solution and is often used to measure changes in ions in complex system. Changes in salinity associated with ion exchange can be estimated by measuring EC of the leachate. In Fig. 2(e-f), the EC value of tailings in the two mining areas after stable treatment is higher than that in the CK. The ion content of the leaching solution in D-3, D-5, Q-3 and Q-5 treatment groups was the highest. The rice husk ash and $FeSO_4$ increase the Fe content of the leachate.

There is no significant change in organic matter content when red mud and fly ash are added (Fig. 2(g-h)). The addition of dry sludge and rice husk ash is beneficial to the increase of organic matter content of tailings. In treatment group 5, the content of organic matter of tailings in Qing Long (Fig. 2h) and Du Shan (Fig. 2g) increase by 8.69 and 13.24 $mg.kg^{-1}$ compared with the CK, respectively, which is consistent with the determination results of higher organic matter content in dry sludge and RHA.

As and Sb Leaching Toxicity of Stabilization Products

Studies have confirmed that the leachability of metals in wastes is controlled by the solubility of metal compounds in the leachate [17]. pH affects and controls the dissolution and conversion of metal hydroxides and thus impacts the leaching behavior of heavy metals [18]. The leaching toxicity of As and Sb after stabilization is shown in Fig. 3. In the untreated tailings, the leaching concentration of Sb from Du Shan and Qing Long reaches 3811 and 320.2μg/L, respectively. The leaching effect of As and Sb is the best in treatment group 3 and 5, and the As content of in Du Shan is reduced by 7.167μg/L and 6.97μg/L, respectively, Sb content decreases by 3014.9μg/L and 2961.8 μg/L.

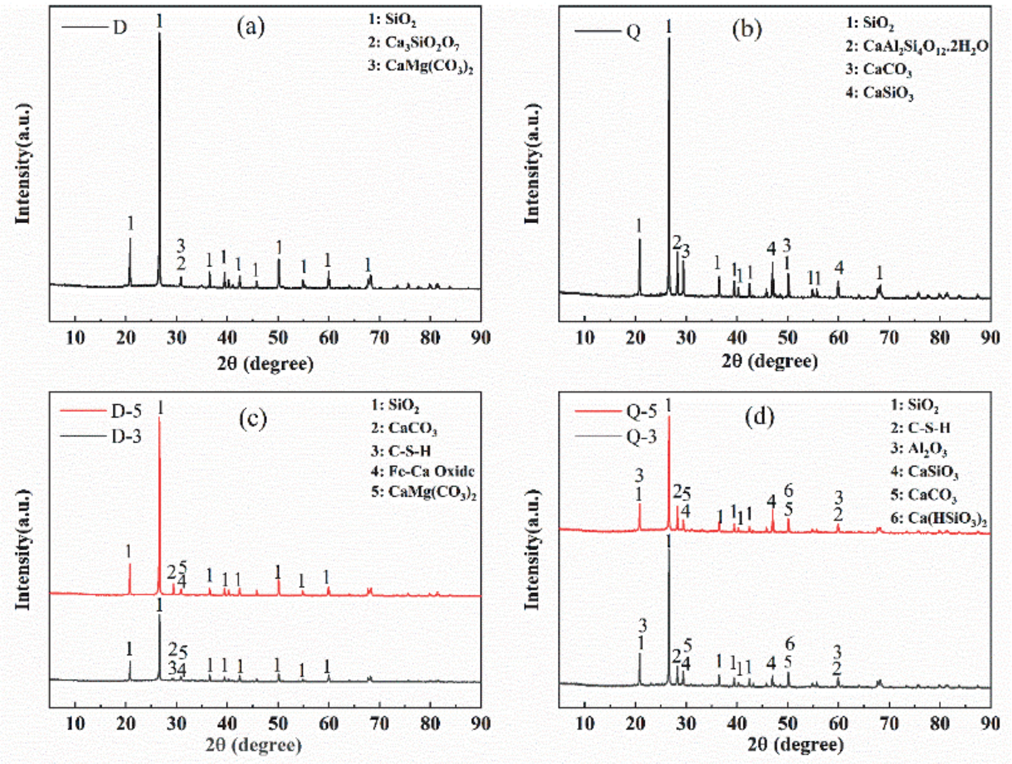

Fig. 1. XRD analysis in Qing Long and Du Shan antimony tailings (before stabilization: a) Du Shan b), Qing Long; After stabilization c) Du Shan, d) Qing Long).

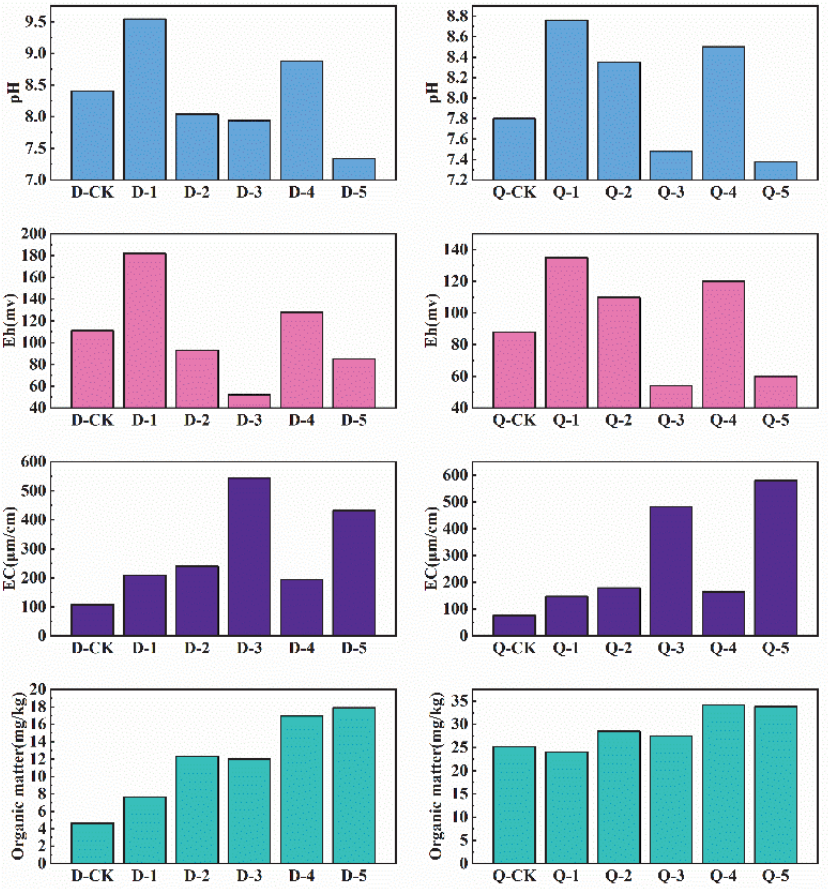

Fig. 2. Change of the stabilizer on physicochemical properties of leachate and organic matter content.

The content of As in Qing Long decrease by 54.88μg/L and 70.17μg/L, respectively, Sb content decreases by 168.4μg/L and 225.3μg/L, respectively. The mechanism may include: (1) the large surface area and porosity of fly ash and the adsorption of Fe and Al oxides in red mud; (2) Precipitation of Ca-As and Ca-Sb with low solubility [19]; (3) Ion exchange, physicochemical inclusion and adsorption of As and Sb by C-S-H and $CaCO_3$. Heavy metal anions can easily replace SO_4^{2-} and OH^-, C-S-H is the main mineral phase for adsorption of As [20]; (4) Chelation of organic functional groups in dried sludge and rice husk ash [21, 22].

The leaching concentration of As and Sb in Qing Long increase at treatment 1, 2 and 4, indicating that stabilizer activate As and Sb of the tailings. It may be that the rapid increase of pH of the leachate affects the stability of the hydration products C-S-H [23]. In addition, the special carbonate landform in Qing Long antimony mine results in the different pH values, which affects the release of Fe, Sb and As. The neutralization reaction of carbonate rocks can induce co-precipitation of Fe with Sb and As [24]. The leaching rate of As is determined by the unstable precipitation of amorphous arsenite [25] and more Sb [26] was released under alkaline pH condition [27].

As and Sb Morphology of Stabilized Products

Fig. 4 shows the transformation of As and Sb form after stabilization. In untreated contaminated tailings, the occurrence forms and proportions of As in Qing Long and Du Shan are similar, with residual As as the main form, followed by Fe-Mn oxide, organic matter binding phase, which is consistent with the research results of [28]. The main Sb forms of tailings in Du Shan are reduced and weak acid extraction state，Sb in Qing Long is mainly residual and reducible state. After the stabilizer is added, the stabilization effect of Sb is greater than that of As, which may be the interaction of As and Sb with iron oxides to reduce the mobility of Sb and moderately increase the mobility of As [29, 30]. Furthermore, the effects of different stabilizers on As and Sb content are significantly different. In the Du Shan antimony mine, treatment groups 1, 4 and 5 promote the conversion of weak acid extraction state, oxidized, reducible As to residual As, the content of weak acid extraction state of the Qing Long decreases at the treatment groups 3 and 5, which indicates that the single and combined addition of $FeSO_4$ and rice husk ash contribute to the stability of As to some extent.

The content of weak acid extraction state Sb is reduced in 5 treatment groups, Sb content in residual state is increased, which reduces the migration of Sb in bioavailable state. This could be that fly ash, as a product of highly active volcanic ash, has strong obligate adsorption on As and Sb. The fixation of As and Sb is mainly controlled by the formation of Ca-As and Ca-Sb precipitates [31, 32]. As and Ca can precipitate $CaHAsO_4$ and $Ca_3(AsO_4)_2$ at stable and moderate pH [33]. This is consistent with the best stabilization effect of As and Sb in treatment group 5 when pH is neutral. Calcium arsenite were deposited as aggregates in

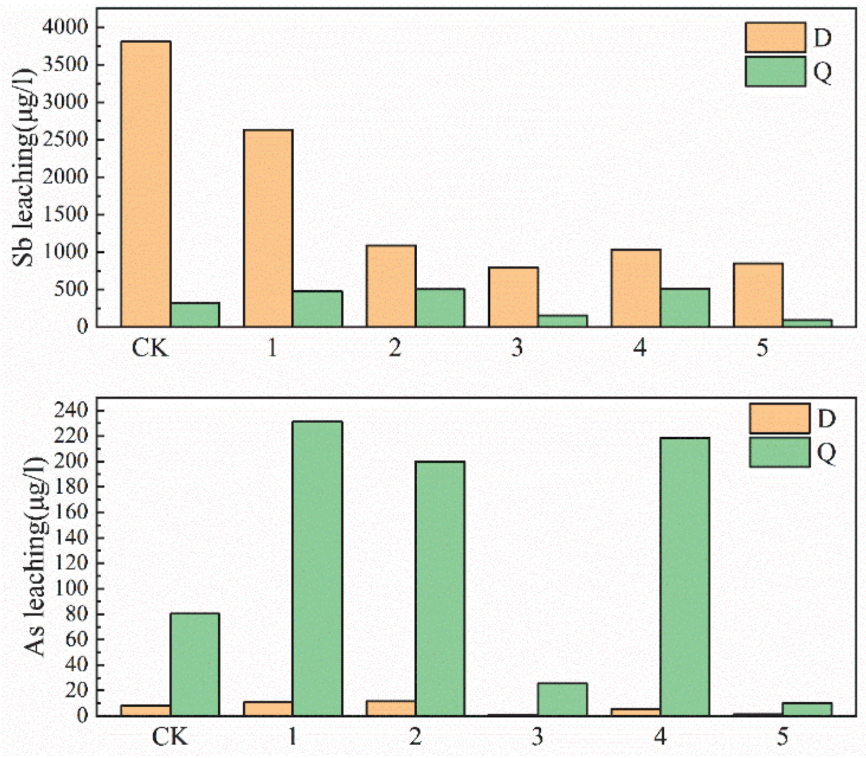

Fig. 3. Changes of leaching concentrations of As and Sb before and after stabilization.

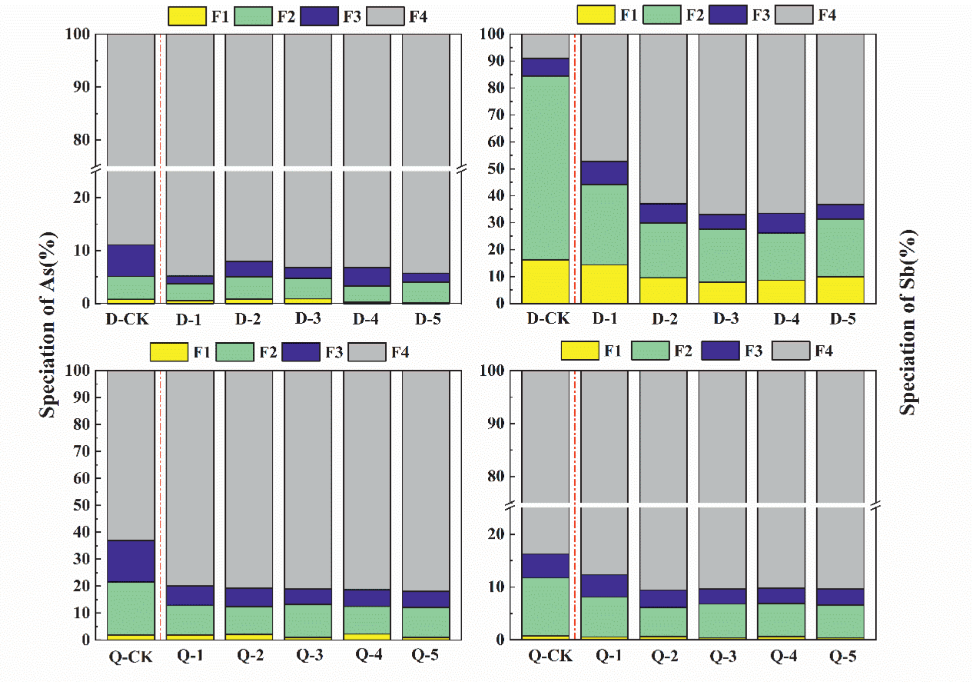

Fig. 4. Morphological transformation of As and Sb in Du Shan and Qing Long antimony mine before and after stabilization.

the pores of biochar, increasing the removal rate of As [34]. According to XRD analysis, the generated C-S-H gel and hydration products of aluminum-calcium hydrate have obvious adsorption and physical sealing effects on Sb [19, 35]. Active groups COO-, -NH, NH, -S- and -O- in dry sludge and rice husk ash enhance the chelation and complexation of heavy metals, promote the formation of organic complexes of heavy metals of tailings, and further enhance the adsorption capacity of heavy metals.

Discussion of Results

Influence of Stabilizer Ratio on As and Sb Leaching Concentrations

Compared with the treatment group 1 and 2, the leaching rate of Sb decreases and As does not change significantly, which is consistent with the research results of [36]. The reasons are speculated as follows: (1) the application of dry sludge has a significant impact on pH and organic matter, the combination of organic matter and tailings can promote the retention of metals. As a non-biomass material, dry sludge is usually used in adsorption studies to eliminate the toxicity of heavy metals, and the adsorption efficiency of metals increases first and then slows down; (2) the increase of fly ash provides more active SiO_2 and Ca^{2+}, which provides conditions for the generation of hydration products. In addition, when the red mud content is too much, fly ash particles will be wrapped, which is not conducive to the further activation of fly ash activity.

Influence of Organic Matter on As and Sb Leaching Concentrations

Tables 6-9 shows the correlation between the physicochemical characteristics of the leachate and the toxicity and occurrence of heavy metals. Compared with treatment groups 2 and 4, the contents of weak acid extraction state As and Sb in treatment groups 3 and 5 decreases in Qing Long, indicating that the stabilization effects of As and Sb were improved by drying sludge and rice husk ash. This is because organic matter contain large numbers of adsorption sites and functional groups such as hydroxyl, carboxyl and aromatic groups, which can be coordinated with heavy metals [37, 38] to control and influence the occurrence state of heavy metals. Compared with treatment group 1, the weak acid extraction state As content in Qing Long at treatment group 2 increases slightly, which may be related to the As content in the dry sludge.

The leaching concentration Sb of tailings in Qing Long is negatively correlated with the organic matter ($R = -0.18$), the weak acid extraction state Sb ($R = -0.34$), the reducible state Sb ($R = -0.57$), the oxidized state Sb ($R = -0.75$), and the residual state Sb ($R = 0.63$) (Table 6). The organic matter of tailings in Du Shan was negatively correlated with Sb leaching concentration and weak acid extraction state ($p<0.05$), and the correlation coefficients (R) were -0.88 and -0.83, respectively. It is positively correlated with reducible Sb ($R = -0.79$), oxidized Sb ($R = -0.36$) and residual Sb ($p<0.05$, $R = 0.82$) (Table 7). The results show that the organic matter is promoted the conversion of bioavailable to residue state. This is consistent with previous findings that weak acid extraction state As and Sb levels at treatment groups 3 or 5 reduced.

For As, the organic matter of tailings in Qing Long is negatively correlated with the leaching concentration of As (R = -0.14), the weak acid extraction state As (R = -0.14), the reduced state As (R = -0.47), the oxidized state As (R = -0.50), and the residual state As (R = 0.50) (Table 8). The organic matter in Du Shan is negatively correlated with the leaching concentration of As (R = -0.54), the weak acid extraction state As (R = -0.68), the reduced state As (R = -0.31), the oxidized state As (R = -0.5), and the residual state As (R = 0.56) (Table 9).

The possible reasons are as follows: (1) The larger specific surface area of rice husk ash and has adsorption and exchange effect; (2) The organic functional groups are complexed with As and Sb, which reduces the bioavailability. In addition, organic matter and heavy metal ions are adsorbed together on mineral surface, and their interaction in mineral-organic matter mix system may lead to antagonistic or synergistic effect on heavy metal adsorption [39, 40]. This is also the reason that the change of weak acid extraction state As and Sb is not obvious after adding dry sludge and rice husk ash.

Influence of $FeSO_4$ on As and Sb Leaching Concentrations

The addition of $FeSO_4$ reduces the leaching concentration of As and Sb of tailings in Qing Long mining area. This is consistent with the findings of [41] and [42]. Which may be that $FeSO_4$ has a certain reducibility, which can reduce Sb (V) to Sb (III), then generate $Sb(OH)_3$ precipitation. In addition, due to the neutralization in Qing Long antimony ore carbonate rocks (natural substances in karst areas), iron flocculates containing large amounts of iron oxides or iron hydroxides at different pH values can effectively remove heavy metal ions Sb and As [24].

In treatment groups 1, 2 and 4, it is found that the content of weak acid extraction state As in Qing Long increased. This may be caused by the precipitation of

Table 6. The correlation between the physicochemical properties of the leachate, toxicity and occurrence of Sb in Qing Long.

Q-Sb	Sb leaching	F1	F2	F3	F4	Organic matter	pH	EC	Eh
Sb leaching	1								
F1	0.59	1							
F2	-0.05	0.69	1						
F3	0.28	0.69	0.83*	1					
F4	-0.07	-0.74	-0.99**	-0.90*	1				
Organic matter	-0.18	-0.34	-0.57	-0.75	0.63	1			
pH	0.46	0.20	0.12	0.63	-0.26	-0.8	1		
EC	-0.85*	-0.90*	-0.48	-0.66	0.57	0.46	-0.45	1	
Eh	0.94**	0.51	0.014	0.41	-0.14	-0.2	0.55	-0.8	1

Note: Significant level *p<0.05, **p<0.01.

Table 7. The correlation between the physicochemical properties of the leachate, toxicity and occurrence of Sb in Du Shan.

D-Sb	Sb leaching	F1	F2	F3	F4	Organic matter	pH	EC	Eh
Sb leaching	1								
F1	0.97**	1							
F2	0.92**	0.87*	1						
F3	0.39	0.42	0.05	1					
F4	-0.96**	-0.92**	-0.99**	-0.16	1				
Organic matter	-0.88*	-0.83*	-0.79	-0.36	0.82*	1			
pH	0.64	0.66	0.32	0.79	-0.41	-0.73	1		
EC	-0.70	-0.66	-0.57	-0.72	0.63	0.48	-0.48	1	
Eh	0.52	0.58	0.18	0.90*	-0.29	-0.34	0.79	-0.69	1

Note: Significant level *p<0.05, **p<0.01.

Table 8. The correlation between the physicochemical properties of the leachate and the toxicity, occurrence of As in Qing Long.

Q-As	As leaching	F1	F2	F3	F4	Organic matter	pH	EC	Eh
As leacing	1								
F1	0.88*	1							
F2	-0.37	0.01	1						
F3	-0.12	0.27	0.95**	1					
F4	0.18	-0.22	-0.97**	-0.10**	1				
Organic matter	-0.14	-0.14	-0.47	-0.50	0.50	1			
pH	0.50	0.31	-0.04	0.13	-0.07	-1	1		
EC	-0.75	-0.93**	-0.33	-0.55	0.51	0.5	-0.4	1	
Eh	0.97**	0.88*	-0.24	0.02	0.04	-0	0.55	-0.8	1

Note: Significant level *p<0.05, **p<0.01.

Table 9. The correlation between the physicochemical properties of the leachate, toxicity and occurrence of As in Du Shan.

D-As	As leaching	F1	F2	F3	F4	Organic matter	pH	EC	Eh
As leaching	1								
F1	0.35	1							
F2	-0.04	0.59	1						
F3	0.26	0.26	0.44	1					
F4	-0.24	-0.5	-0.69	-0.95**	1				
Organic matter	-0.54	-0.68	-0.31	-0.5	0.56	1			
PH	0.67	0.31	-0.35	-0	0.07	-0.73	1		
EC	-0.78	0.004	0.12	-0.7	0.49	0.48	-0	1	
Eh	0.65	-0.27	-0.65	-0	0.22	-0.34	0.8	-0.7	1

Note: Significant level *p<0.05, **p<0.01.

Fe(II) and the inhibition of the formation of ferrous arsenate minerals [38]. The interaction of As, Sb and iron oxides reduced the mobility of Sb and increased the mobility of As in soil to a small extent [29]. Compared with treatment group 3, the concentration of Sb in Du Shan increases in treatment group 5, which indicates that the addition of a certain amount of $FeSO_4$ may promote the release of a small amount of Sb. This is consistent with the result that the content of weak acid extraction state Sb at treatment group 5 is slightly increased compared with that in treatment group 3. It is speculated that the oxidation and reducibility of Fe affect the adsorption and precipitation of Sb, which is consistent with the experimental results of [43] that after curing for more than 5 days, the stability effect of ferrite, $FeSO_4$ on Sb all decreased slightly. The results of [44] have showed that the presence of inorganic and organic matter changed the physicochemical properties of the iron salt precipitation, in turn affected the adsorption and precipitation of Sb during the stabilization process.

Effect of pH-Eh on Leaching of Heavy Metals

pH-Eh has great influence on the solubility, migration and transformation of metals [45]. The increase of pH value of tailings induces metal solidification/stabilization, facilitates metal precipitation, reduces the solubility of heavy metals, and promotes metal adsorption by increasing the net negative charge of variable charge in environmental media [46, 47]. In addition, the pH of the system can affect the reaction rate and the formation and crystallization of stabilized products [48].

According to Fig. 3, the leaching rate of Sb of the tailings in two mining areas is the highest in treatment groups 1,4. For As, the two antimony mine have the highest leaching rate in treatment group 1, which may be related to the higher pH. The high alkalinity of red mud and fly ash makes the pH of the system increase rapidly, the adsorption and complexation of As and Sb in Fe and Al(hydrogen) oxides decrease, and the concentration of As and Sb increases. For As, the reduction process of Fe(hydrogen) oxide affects the precipitation of amorphous Fe-As. [49] showed that when pH = 7, the removal rate of As was as high as 99.9%. This indicated that when pH was alkaline, the leaching rate of As increased. With the addition of $FeSO_4$, rice husk ash and dried sludge, the maximum leaching rates of As and Sb are reduced, which indicates that the dried sludge, rice husk ash and $FeSO_4$ reduce the pH of the system to a certain extent and promote the stability of As and Sb.

Studies have shown that pH-Eh controls the morphologic transformation and mobility of As and Sb as well as the properties of adsorbents [50-52]. The main forms of As and Sb exist are As (III), As (V), Sb (III) and Sb (V), and Sb (III) has poor liquidity and availability [53]. The adsorption strength of Fe-Al (H) oxides on As and Sb is affected by pH-Eh, As and Sb are strongly retained on iron (hydrogen) oxides. Oxidized metals are generally more readily retained at acidic pH, while reducing substances are easier adsorbent at higher pH [54, 55]. The results showed that pH was positively correlated with leaching concentrations of Sb and As of tailings, which further indicated that when pH increased, the system was in a weak reducibility, because Sb(V) was relatively stable, causing Sb not easy to be released. While As exists as a mixture of As(III) and As(V) with increased mobility [51, 56]. Therefore, for Sb, this better compensates for the release of adsorbed on the surface of Fe-Al (hydrogen) oxidized minerals under reduction conditions.

Influence of Stabilized Products on As and Sb Leaching Concentrations

As a cementitious material of alkaline activator, the pozzolash activity of fly ash may be excited. In the hydration process, the hydrolysate of red mud generated OH^-, which provided alkaline conditions. OH^- gathered around fly ash particles and wrapped them, due to the polarization of OH^-, Si-O bonds and Al-O bonds in fly ash particles were broken. With the increase of pH in the system, the vitreous of fly ash was gradually corroded, the internal active SiO_2 and Al_2O_3 were gradually released. The hydration reaction occurred with OH^- to form C-S-H.

$$SiO_2 + mCa(OH)_2 + nH_2O - mCaO \,.\, SiO_2 \,.\, nH_2O \quad (1)$$

$$Al_2O_3 + mCa(OH)_2 + nH_2O - mCaO \,.\, Al_2O_3 \,.\, nH_2O \quad (2)$$

The main hydration products produced by C-S-H gel, calcite and other substances, are mixtures of crystalline minerals and disordered phases [57], with low permeability and containing larges number of micropores, which can wrap As and Sb, prevents the dissolution of metal ions. Due to the large specific surface area of these gels, the metal binding ability is strong and the stability is good.

Distribution of C-S-H enhances the compactness between tailings and stabilizers. By substituting Ca^{2+}, Ca, and Si of C-S-H surface, the metal can be fixed in a granular lattice. The replacement reaction of SO_4^{2-} and Ca^{2+} in C-S-H, the formation of precipitation of Ca-As and Fe-As, the physical encapsulation and chemisorption of C-S-H gel were the main mechanisms to prevent the release of As [58], which forms substances with low mobility and solubility. Comparing with tailings not stabilized, the generation of calcite ($CaCO_3$) promotes the stability of S and Sb. It is speculated that the formation of a small amount of carbonate precipitation, which is consistent with the research result that the weak acid extraction state of As increased slightly in this study. The continuous $CaCO_3$ structure adsorbed and encapsulated fly ash, heavy metal was transformed from the free state to carbonate precipitation through carbonation, was easier to be physically solidified by $CaCO_3$ [59]. In addition, the precipitation of metals on the surface of silicate phase reflects the degree of solidification of metals, $CaCO_3$ can accelerate the formation of silicate, promote the solidification/stability of waste. The $CaCO_3$ produce a larger proportion of hydrated crystals, fills the void of the tailings particles and cemented the adjacent particles, improve mechanical properties, reduce the permeability and leaching rate of the metal [60]. With the precipitation of $CaCO_3$, heavy metal ions can enter the $CaCO_3$ crystal or fill the vacancy in the crystal by replacing Ca^{2+}, and be fixed in the $CaCO_3$ lattice in the form of carbonate or co-precipitation [61-63].

In summary, for carbonate tailings, stabilizer content, iron oxide, pH and redox conditions affect the leaching behavior of As and Sb, and the release of metals in tailings is affected by acid-base buffering ability. The reducing property of $FeSO_4$ inhibits the release of As and Sb. However, the interaction of Fe with As and Sb promotes the stabilization of Sb and a small amount of As release. The resulting stabilized products contribute a lot to the fixation and cementation of metals. Stabilization technology has been widely accepted as a method to treat heavy metals. The leaching behavior of metals under long-term conditions is affected by freeze-thaw conditions, acid rain and other conditions, which has potential threats to the surrounding ecological environment. Therefore, the variability and complexity of mine tailings make this threat possible. In addition, there are significant differences in the properties of stabilizing materials, and a thorough mix of optimization work is required to determine the optimal stabilization method (thermal treatment, material strength etc.) [64], taking into account technical constraints, environmental factors, and cost management in stabilizing operations.

Conclusions

The high alkalinity of red mud and fly ash makes the pH of the system increase rapidly. Dried sludge and rice husk ash increased the organic matter content of the tailings. The addition of $FeSO_4$ buffered the rise of pH of the system, and its reducibility promoted the formation of precipitation of As and Sb. The interaction of As, Sb and iron oxides reduced the mobility of Sb, the mobility of As increased to a small extent. The addition of 5% red mud, 10% fly ash, 5% dried sludge, 1% rice husk ash, and 1% $FeSO_4$ reduced the leaching concentrations of As and Sb. When the pH was neutral, the stabilization effects of As and Sb was the best, promoting the conversion of As and Sb from the weak acid extraction state, oxidizable, and reducible states of the polluted tailings to the residual state, effectively reducing the bioavailability of As and Sb. Fly ash promotes the formation of stabilization products under the condition of OH^- provided by red mud. The main products C-S-H promoted the precipitation of metal ions and reduced their mobility through physical encapsulation, chemisorption and ion exchange. The formation of $CaCO_3$ promoted the close binding of tailings particles with stabilizers and reduced the metal leaching rate. The presence of inorganic and organic matter changed the physicochemical properties of the iron salt precipitation, in turn affected the adsorption and precipitation of Sb during the stabilization process. Excessive red mud is not conducive to the activation of fly ash and affects the solidification and stability of metals.

Therefore, on the basis of using industrial solid waste to treat soil stably in the past, this study mixed agricultural solid waste to realize waste treatment, effectively reduce costs, reduce environmental pressure, and provide a reference for future research on a variety of different composite stabilizer materials to solve the problem of large amounts of environmental media tailings accumulation and land occupation. It also provides a theoretical basis for the long-distance migration and chemical leaching behavior of antimony elements in antimony tailings. The follow-up tailings treatment in this study is applicable to metal leaching behavior in the short term, and the determination of metal release under long-term complex environmental stimulation requires outdoor simulation combined with the characteristics of the study area. In addition, this study is limited to indoor experiments, and fails to fully consider metal migration caused by changes in the external environment.

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Conflict of Interest

The authors declare no conflict of interest.

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