PROCEEDINGS OF THE YEREVAN STATE UNIVERSITY

Chemistry and Biology 2025, **59**(1–2), p. 1–11

Chemistry

THE INFLUENCE OF PEAT ON NEUTRALIZATION PROCESSES OF ACID MINE DRAINAGE WATERS

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The effect of peat on the processes of neutralization of acid mine drainage (AMD) waters was studied using ICP-OES and Jar test methods. Untreated peat (from the Vardenis deposit, Armenia) and acid-activated peat were used. Peat activation means the elution of metals sorbed on the peat surface with mineral acid. In this case, active sorption groups (in particular, carboxyl) are released from metals, and acidic groups begin to act by the cation-exchange mechanism. The introduction of peat (untreated and acid-activated) into AMD leads to a decrease in the amount of alkali used in neutralizing AMD. With an increase in pH, schwertmannite and basaluminite are formed, which sorb copper, nickel, cobalt, zinc, etc. The sorption study was carried out in the range up to pH 6.15, since at higher pH values, against the background of more intensive precipitation of metal hydroxides, the sorption effect in terms of removing metals from water becomes less noticeable.

https://doi.org/10.46991/PYSUB.2025.59.1–2.001

***Keywords*:** peat-based sorbents, acid activation of peat, sorption of heavy metals, acid mine drainage.

Introduction. There is a great need to remove heavy metals from heavily metal-contaminated wastewater sludge. The use of chemical precipitation to remove dissolved heavy metals from wastewater involves high chemical costs. The disadvantages of the chemical precipitation process are related to the amount of chemicals, the final amount of waste, the difficulty of separating the sludge and liquid and, of course, the cost of the chemicals. However, these disadvantages are the reason for research in this area to improve heavy metal removal processes, as they are not yet applicable on a real scale [1–3].

Neutralization of acid mine drainage (AMD) results in the formation of either schwertmannite (at $pH<4$) or ferrihydrite (at $pH>4$). At higher pH values (~5), precipitates rich in Al are formed. Manganese compounds precipitate at pH~8. The removal of trace elements depends on the precipitation of these compounds, which act as sorbents [2].

It has been shown that Fe (III) and Al (III) can start to form hydroxide precipitates at pH 1.3 (Fe) and 3.5 (Al). It is suggested that adsorption and/or coprecipitation of Cr, Pb, and Zn with $Fe(OH)_3$ and $Al(OH)_3$ occurs as the dominant factors in the removal of these metals. Thus, the removal of Cr, Pb, and Zn depends

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on the precipitation of Fe and Al hydroxides, which act as sorbents [2] and co-precipitants [4].

AMD is perhaps one of the most serious due to its nature and scale. The mining industry is the main source of AMD. The AMD problem affects active and abandoned mines. AMD must be treated because it can pollute surface waters [5].

Acid rock drainage (ARD) is a natural process that occurs when sulfur-containing compounds in rocks are exposed to air and water. When this process occurs during mining operations, the process is called acid mine drainage. In this process, acidic waters are produced by the oxidation of sulfur-containing compounds, forming sulfuric acid. Typically, the pH of acidic water is between 2.1 and 3.5, which allows the water to leach metals such as As, Cd, Co, Cu, Pb, and Zn from the rock and soil in contact with them [6, 7].

Constructed Wetland System. AMD processing is carried out by passive and active Wetland methods. Active methods require constant input of chemical reagents. In practice, active AMD processing refers to the continuous use of alkaline materials to neutralize acidic mine waters and precipitate metals [8–10].

Wetlands (CW-constructed Wetlands) are capable of improving water quality through physical, chemical, microbial, and plant processes. In addition to other processes (oxidation, reduction, precipitation, sedimentation, complexation, filtration, chelation, active absorption of metals, etc.), adsorption also belongs to them [11, 12].

There are many treatment practices for removing pollutants from wastewater. Of these, bed filtration using adsorbents has been widely employed as an effective treatment strategy for removing organic and inorganic pollutants from stormwater [13].

Humic acids are a universal sorbent for all types of heavy metals in cationic form, as they form strong compounds with metal ions. It is known that a variety of compounds form complexes with metal ions, providing the potential to remove metal ions from the compositions they contaminate. The capacity of humic and fulvic acids to form complexes with metal ions is well established, and a process for removing metals from water using humic acid has been developed [14, 15].

The search for new cheap sorption materials is becoming more relevant, since the treatment of large quantities of wastewater (in particular, AMD) requires adsorbent materials, the amount of which will be counted not in kilograms, but in tons. In this regard, peat is becoming an indispensable sorption material.

Peat is an inexpensive, readily available and effective sorbent for a wide range of environmental pollutants and can be used either alone or as a component of a combined sorbent. Peat is an effective sorbent for heavy metals due to its high cation exchange capacity. Over the past decades, various types of peat have been used to produce cost-effective adsorbents for the treatment of municipal and industrial wastewater [16–19].

Peat is a complex polydisperse multicomponent system, including an organic part, moisture, and mineral impurities. Due to the presence of many biologically active compounds and several reactive functional groups in peat, interest in the development of peat sorbents is growing. Due to the presence of organic compounds containing polar functional groups, such as hydroxyl, carboxyl, carbonyl, etc., peat has a high complexation capacity [20, 21].

This work aims to study the influence of peat on the processes of neutralization of AMD waters.

Experimental Part.

Experimental Methodology. Jar Test Procedure. The equipment used for the jar tests was a Flocculator 2000 (“Kemira”, Finland), which has six adjustable paddles with rotating speeds between 0 to 250 *rpm*. AMD solution was poured into each of the six 1000 *mL* glass beakers for the test. Different doses of 2 *N* NaOH were added to the AMD samples. The experiments were conducted employing rapid stirring (250 *rpm* for 5 *min*).

Peat materials in the amount of 5 *g* were introduced into 800 *mL* of the solution before increasing its pH.

pH Measurement. The instrument pH/Ion 340i Meter (Weilheim 2004, WTW GmbH) was used to measure pH values. The equipment was calibrated with standard solutions of pH of 4.0 and 7.0 before use.

Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES). Perkin Elmer Optima DV 7000 ICP-OES instrument (USA) was used to determine the metals in the supernatant using *ppm* as the unit of measurement. It was calibrated with the standard solution between 2 to 50 *ppm* of the salts mentioned in this study.

Materials and Methods. Drainage water was used for AMD treatment experiments. The sorption materials were kept in water for 60 *min* and then filtered. The resulting supernatant was neutralized with a solution of 2 *N* NaOH.

Two types of peat were used: 1) untreated peat; 2) peat activated with mineral acids. To obtain activated peat samples, acid treatment was carried out. For this purpose, 1 *N* HCl solution was prepared, added to the sample and kept for 24 *h*. Then it was washed with distilled water until neutral pH, dried in the air, and then in a dryer at 105°C.

Acid Leaching of Metals from Peat. 3 samples of unprocessed peat, 12.5 *g* each, with a moisture content of 60% (based on 5 *g* of dry peat) were taken, 10 *mL* of 5 *N* solutions of hydrochloric, sulfuric, and nitric acids were added, and were left to stand during the day, the volume was brought up to 50 *mL* with distilled water and filtered. The chemical composition of the resulting solutions was determined by the ICP-OES method. The results obtained are presented in Tab. 1.

Table 1

The chemical composition of the solutions after acid treatment of the peat

Acid	Al, *mg/L*	As, *mg/L*	Ba, *mg/L*	Ca, *mg/L*	Cd, *mg/L*	Co, *mg/L*	Cr, *mg/L*	Cu, *mg/L*	Fe, *mg/L*	Mg, *mg/L*	Mn, *mg/L*	Na, *mg/L*	Ni, *mg/L*	P, *mg/L*	Pb, *mg/L*	S, *mg/L*	Ti, *mg/L*	Zn, *mg/L*	Total content, *mg/g*
10% H_2SO_4	1602	2.01	0.07	185.8	0.04	0.43	0.82	21.53	426.82	390	14.90	373	6.96	19.5	10.5	14783	23.39	104.0	31.72
10% HCl	1554	2.29	3.19	756.9	0.02	0.38	0.77	25.62	329.78	408	15.85	353	6.57	28.6	22.2	154	14.54	100.2	36.15
10% HNO_3	1652	2.67	2.52	258.2	0.02	0.43	0.34	24.43	417.78	442	16.42	371	7.33	31.2	24.3	53	17.50	110.0	31.00

Results and Discussion. Neutralization of AMD waters involves the use of active ingredients. Among such ingredients, sorption materials can occupy an important place, since they help remove heavy and other metals. In addition, after saturation of sorption materials (in particular, peat) during their regeneration (which implies acid treatment), it will be possible to extract several metals from regeneration solutions, which will make it possible to reduce the cost of purification processes of AMD. Among such sorption materials, peat occupies an important place, because it is an accessible, cheap, and effective sorbent material. However, the use of untreated peat for these purposes is ineffective. Therefore, peat is subjected to acid treatment before entering AMD. In addition, peat is well regenerated, as a result of which it can be used repeatedly.

Fig. 1 shows the neutralization curve of AMD with and without sorption material (acid-treated peat) as NaOH is added. It follows from Fig. 1, that the curve has 3 plateaus. We assume that the first plateau in the pH range of 3.2–3.6 corresponds to the precipitation of trivalent iron, i.e. until all the trivalent iron precipitates, the pH of the medium does not increase. After adding 0.32 *g* of alkali, a slight increase is observed (pH 3.6–3.96). Then the second plateau is observed, which reached 1.04 *g* NaOH. After adding the next portions of NaOH, the pH of the medium rises to pH 8.09. After which the 3rd plateau begins – 2.4 *g* NaOH in the pH range of 10.50–11.25.

It should be noted that when introducing acid-treated peat (Peat-AT) into AMD (pH 2.85), the AMD neutralization curve not only does not move upward from the AMD neutralization curve without introducing sorbent material into its composition (Fig. 1), but even in some of its sections is below this curve.

It follows from Tab. 1, that the maximal quantity of metals is eluted with hydrochloric acid (36.15 *mg*/*g*). However, in general, the quantity of metals eluted with the three acids is, on average, comparable. The values of the sorption capacity of peat, determined in this way, are 33.87 ± 2.87 *mg*/*g*.

Table 2

Precipitation of the elements from AMD in the presence and without it connected with rising pH

Element, *mg*/*L*	Initial Wt.	6 *L* AMD + 20 *mL* 2 *N* NaOH, pH 3.95	3 *L* AMD + Peat	3 *L* AMD + Peat-AT	6 *L* AMD + 70 *mL* 2 *N* NaOH, pH 4.85	6 *L* AMD + Peat	6 *L* AMD + Peat-AT	6 *L* AMD + 90 *mL* 2 *N* NaOH, pH 6.15	9 *L* AMD + Peat	9 *L* AMD + Peat-AT
Al	178.42	145.34	124.84	102.12	4.79	2.63	1.58	0.56	0.40	0.34
Ca	285.71	280.24	321.89	342.34	291.83	321.32	349.36	295.45	357.18	346.46
Cd	0.19	0.18	0.16	0.16	0.19	0.14	0.11	0.19	0.10	0.10
Co	1.05	0.96	0.96	0.90	0.93	0.80	0.72	0.91	0.64	0.70
Cr	0.03	0.02	0.02	0.02	0.03	0.02	0.02	0.02	0.02	0.02
Cu	2.84	2.43	1.47	1.01	0.79	0.16	0.05	0.09	0.01	0.01
Fe	162.95	93.78	49.07	109.18	88.50	80.89	64.67	69.05	50.47	50.96
Mg	284.37	264.19	277.12	268.32	263.36	259.92	255.45	267.92	260.40	257.61
Mn	169.31	159.57	157.05	154.21	158.06	148.75	139.44	159.24	144.72	140.02
Ni	0.44	0.41	0.42	0.40	0.37	0.32	0.27	0.36	0.24	0.25
S	1361.57	1158.85	1301.83	1174.12	1211.61	1198.30	1203.78	1250.70	1240.77	1086.19
W	1.72	1.59	1.55	1.46	1.66	1.41	1.20	1.35	0.82	0.93
Znl	67.12	62.54	61.10	58.29	59.75	51.24	45.38	48.94	31.07	35.35

When untreated peat was introduced into AMD with a pH of 2.85, some of the adsorbed elements (in particular, calcium and magnesium) passed into the solution (see Tab. 2).

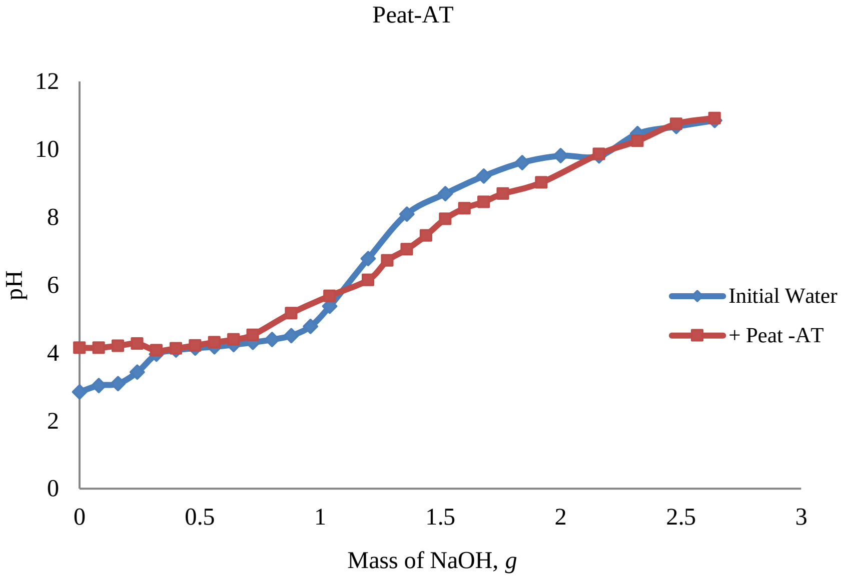

Fig. 1. Neutralization curves for the following systems:
1) initial AMD (pH 2.85); 2) AMD (pH 2.85) – Peat-AT, when adding NaOH.

It follows from Fig. 1 that after adding Peat-AT to AMD (at pH 2.85), the neutralization curve in the range of 1.2–2.3 *g* NaOH is located below the neutralization curve of the original AMD. Due to its nature, acid-activated peat has cation-exchange functional groups in the form of COO^- groups of humic acids present in the peat. Especially after acid treatment, such groups are released from adsorbed metals and, as a result, cation-exchange centers are obtained on the surface of such systems. After adding Peat-AT, cation exchange occurs, as a result of which H^+ ions appear in the system, for the neutralization of which an additional NaOH amount needed. These processes are reflected on the neutralization curves.

With increasing pH, the system for some time is on the so-called aluminum plateau up to 0.75 *g*/*L* NaOH, then the neutralization curve smoothly increases, slightly breaking away from the neutralization curve of the initial water, however, at the point of 1.1 *g* NaOH, the curves intersect and starting from 1.1 *g* to 2.3 *g* NaOH, the curve with Peat-AT is below the curve of the initial water, since this interval corresponds to the precipitation of different metals (Cu, Mn, Fe^{2+}, partly Zn, Ni, etc.) apparently as a result of cation exchange, H^+ is introduced into the system, which lowers the pH.

Therefore, it was advisable to first increase the pH to 3.95 and then introduce Peat-AT, and this is natural, since at low pH, the sorption by the cation exchange mechanism is inhibited or does not occur at all.

Fig. 2 shows the neutralization curve of AMD, in which the pH was preliminarily increased to pH 3.95, after which sorption materials (untreated and acid-activated peat) were introduced. In this case, the neutralization curves move above the neutralization curve of the original AMD, i.e., with the same amounts of NaOH, the pH of the medium is higher when peat materials are used. It should be noted that the further the neutralization curve is from the neutralization curve of the original AMD, the greater the sorption properties of the given sorption material.

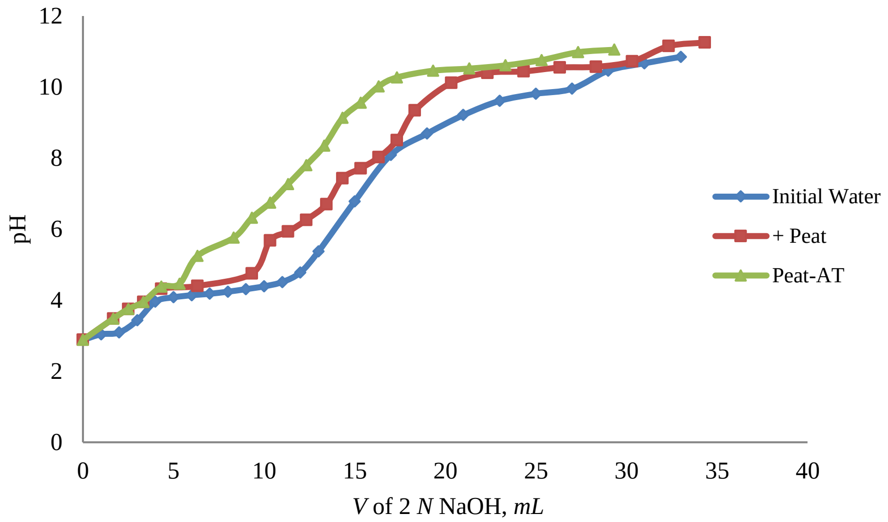

Fig. 2. AMD Neutralization curves (pH 3.95): 1) initial water; 2) with peat; 3) with Peat-AT.

Such behavior of the system with sorption material can be explained by the fact that the presence of sorption materials in AMD leads to the sorption of metals (in particular Fe, Al) (see Fig. 3, Tab. 3).

In the case of Peat-AT, the neutralization curve reaches saturation faster (1.2 *g* NaOH), pH is greater than 10, than in the case of using untreated peat (1.6 *g* NaOH) and the original AMD (2.0–2.4 *g* NaOH), which indicates that when using sorption materials, the amount of NaOH required to neutralize AMD decreases, which in turn reduces the cost of the AMD purification process.

Using acid-treated peat as an example of a sorption material, the efficiency of using such systems in the process of neutralizing AMD is shown depending on the pH of the environment at which the sorption material is introduced into the system.

The above-mentioned regularities (placement of neutralization curves AMD in the presence of sorption materials) will be considered from the point of view of which metal behaves how under different conditions.

To clarify the effect of acid activation of peat on its sorption properties, the values of the content of individual metals in AMD during its neutralization with sodium hydroxide are presented in Tab. 3.

It was noted above that when neutralizing AMD to pH 4.0, iron precipitates in the form of schwertmannite, then at pH>4, iron precipitates in the form of ferrohydride [2]. As follows from Fig. 3 the precipitation of manganese with an increase in pH to 6.15 is insignificant, therefore, manganese hydroxide practically does not participate as a sorption material. Of course, after adding peat and activated peat, the precipitation curves are located below the precipitation curve from AMD without adding sorption materials.

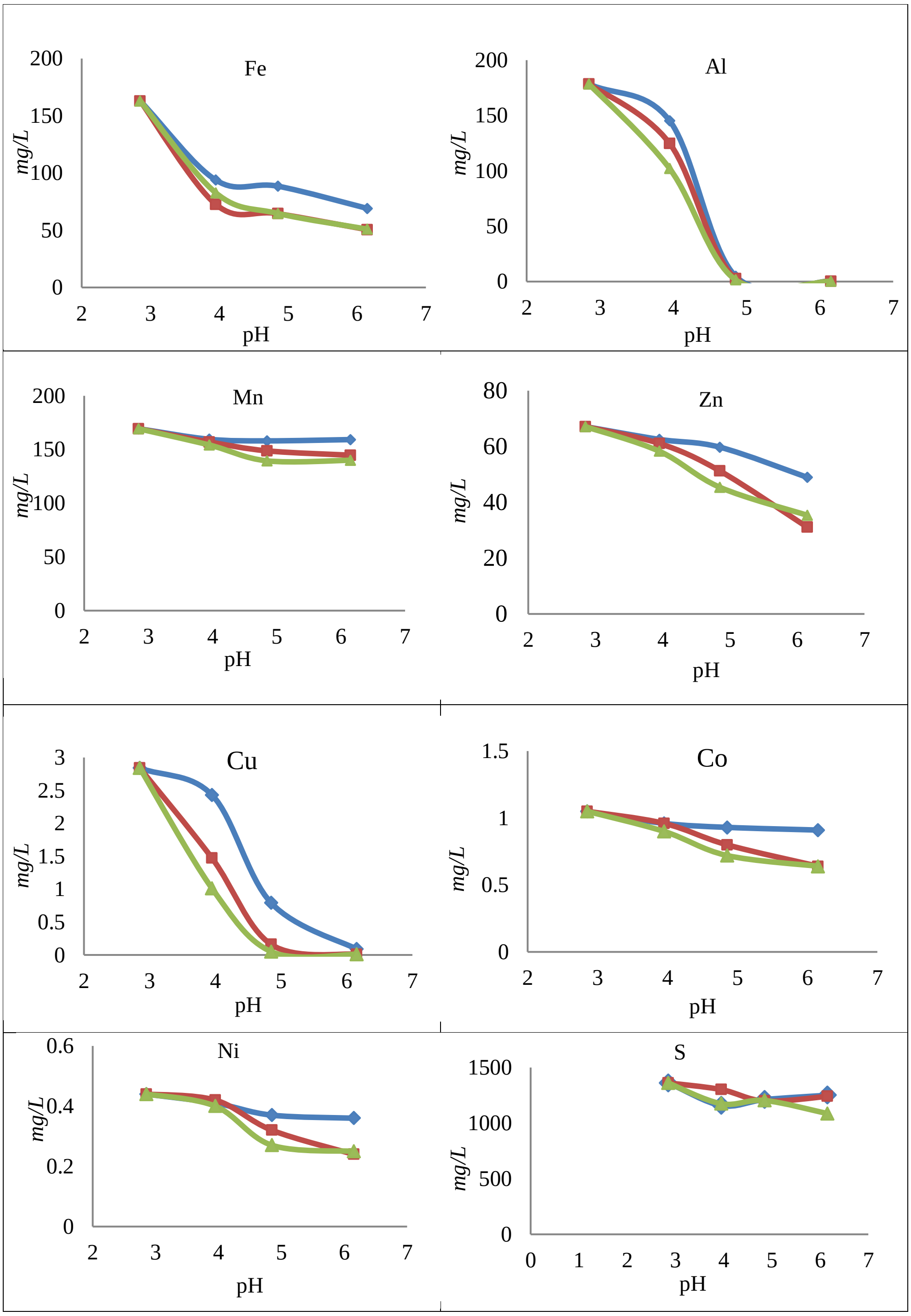

Fig. 3. The influence of pH on the metal precipitation:
1 – without using sorption material (blue line); 2 – non-treated peat (red line);
3 – Peat-AT (green line).

Table 3

Precipitation of the metals

pH	3.95			4.85			6.15		
	[Me]	%	Removal, %	[Me]	%	Removal, %	[Me]	%	Removal, %
Precipitation of Fe (initial concentration 162.95 *mg*/*L*)									
Without sorbent	95.78	57.53	42.45	88.50	54.31	45.69	69.05	42.37	57.63
Peat	72.51	44.50	55.50	64.52	40.21	59.79	50.47	30.97	69.03
Peat-AT	82.54	50.65	49.35	64.67	39.59	60.31	50.96	31.27	68.73
Precipitation of Al (initial concentration 162.95 *mg*/*L*)									
Without sorbent	145.34	89.19	10.81	4.79	2.94	97.06	0.56	0.34	99.66
Peat	124.84	76.61	23.39	2.63	1.61	98.39	0.4	0.25	99.75
Peat-AT	102.12	62.67	37.33	1.58	0.97	99.03	0.34	0.21	99.79
Precipitation of Mn (initial concentration 169.31 *mg*/*L*)									
Without sorbent	159.57	94.25	5.75	158.06	93.36	6.64	159.24	94.04	5.96
Peat	157.05	92.76	7.24	148.75	91.29	8.71	144.72	85.48	14.52
Peat-AT	154.21	91.08	8.92	139.44	82.36	17.64	140.02	82.70	17.30
Precipitation of Zn (initial concentration 67.12 *mg*/*L*)									
Without sorbent	62.54	93.18	6.82	59.75	89.06	10.94	48.94	72.91	27.09
Peat	61.10	91.03	8.97	51.24	76.34	23.66	31.07	46.29	53.71
Peat-AT	58.29	86.84	13.16	45.38	67.61	32.39	35.35	52.67	47.33
Precipitation of Cu (initial concentration 2.84 *mg*/*L*)									
Without sorbent	2.43	85.56	14.44	0.79	27.82	72.18	0.09	3.17	96.83
Peat	1.47	51.76	48.24	0.16	5.63	94.37	0.01	0.35	99.65
Peat-AT	1.01	35.56	64.44	0.05	1.76	98.24	0.01	0.35	99.65
Precipitation of Co (initial concentration 1.05 *mg*/*L*)									
Without sorbent	0.96	91.43	8.57	0.93	88.57	11.43	0.91	86.67	13.33
Peat	0.96	91.43	8.57	0.8	76.19	23.81	0.64	60.95	39.05
Peat-AT	0.90	85.71	14.29	0.72	68.57	31.43	0.70	66.67	33.33
Precipitation of Ni (initial concentration 0.44 *mg*/*L*)									
Without sorbent	0.41	93.18	6.82	0.37	84.09	15.91	0.36	81.82	18.18
Peat	0.42	95.45	4.55	0.32	72.73	27.27	0.24	54.55	45.45
Peat-AT	0.40	90.91	9.09	0.27	61.36	38.64	0.25	56.82	43.18

The following picture is observed during zinc precipitation. At pH 6.15, the zinc content decreases by 27.09%, i. e. zinc co-precipitates with aluminum and iron hydroxides: when using untreated peat 53.74% and Peat-AT – 47.33%. Apparently, mechanisms other than cation-exchange are involved in zinc sorption because otherwise sorption on activated peat should be greater than on untreated peat. With an increase in pH, a sharp decrease in the aluminum content is observed, of course, in the presence of sorption material such a decrease begins earlier, and at pH 6.25 in both cases the aluminum content reaches zero. An insignificant loop is observed. Already after reaching pH 4.85, the aluminum content reaches zero. In the case of iron, even at pH 6.25, some of the iron does not precipitate, which indicates that

AMD contains not only Fe (III) iron, but also divalent iron. Not all divalent iron has converted to trivalent iron, and the precipitation of divalent iron at pH 6.25 is only just beginning. In addition, metals present in AMD during its neutralization with sodium hydroxide co-precipitate with Fe (III) and Al (III) [1, 2], although their hydroxides precipitate at relatively high pH values. If Fe (III) precipitates completely at pH 4.0, and aluminum at pH 5.0–6.0, then the detection of metals such as copper, zinc, cobalt, nickel, etc., in the sediments can only be explained by the co-precipitation of these metals in adsorbed form with Fe (III) and Al (III) (Tab. 2).

Conclusion. Sorption materials accelerate the process of removing metals from AMD during its neutralization with sodium hydroxide, and the removal of metals occurs at relatively low pH values, which in turn means a reduction in the amount of neutralizing agent (alkali).

Sorption of trace (and other) metals occurs on the surface of freshly deposited schwertmannite and basaluminite, which, since they have just formed, have very active and developed surfaces. Such processes greatly affect the deposition curves of the indicated metals in the process of neutralization (increasing the pH) AMD. Against this background, the neutralization curves of these solutions look interesting, but already with sorption materials (in particular, peat). All these curves are located to the left of the neutralization curve AMD, but without sorption materials, which indicates that the amount of neutralizing agent (in this case, NaOH) is significantly reduced, which in turn makes the neutralization processes cheaper.

Received 25.03.2025
Reviewed 16.04.2025
Accepted 25.04.2025

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Մ. Ս. ՀԱՅՐԱՊԵՏՅԱՆ

ՏՈՐՖԻ ԱԶԴԵՑՈՒԹՅՈՒՆԸ ԹԹՎԱՅԻՆ ՀԱՆՔԱՅԻՆ ԴՐԵՆԱԺԻ ՋՐԵՐԻ ՉԵԶՈՔԱՑՄԱՆ ՎՐԱ

ICP-OES и Jar test եղանակների կիրառմամբ հետազոտվել է տորֆի ազդեցությունը թթվային հանքային դրենաժի ջրերի (ՀԴՋ) չեզոքացման վրա: Օգտագործվել են չմշակված տորֆ (Հայաստանի Հանրապետության Վարդենիսի հանքավայր) և թթվային մշակման միջոցով ակտիվացված տորֆ: Տորֆի ակտիվացում ասելով, նկատի ունենք հանքային թթվով տորֆի մակերևույթին կլանված մետաղների լվացումը: Այդ ժամանակ ակտիվ թթվային սորբումային խմբերը (մասնավորապես, կարբօքսիլային) ազատվում են մետաղներից և սկսում են գործել կատիոնափոխանակային մեխանիզմով: ՀԴՋ-ի մեջ տորֆի ներմուծումը հանգեցնում է չեզոքացման համար ծախսվող ալկալու քանակի նվազման: Թթվության նվազման արդյունքում առաջանում է շվերտմաննիտ և բասալյումինիտ, որոնք կլանում են պղինձը, նիկելը, կոբալտը, ցինկը և այլ տարրեր: Սորբումային գործընթացներն ուսումնասիրվել են մինչև pH 6.15, քանզի ավելի բարձր արժեքների տիրույթում, մետաղների հիդրօքսիդների ավել ինտենսիվ նստեցման ֆոնի վրա զուտ սորբումային գործընթացների ազդեցությունը մետաղների հեռացման գործում համեմատաբար քիչ նկատելի է դառնում:

М. С. АЙРАПЕТЯН

ВЛИЯНИЕ ТОРФА НА НЕЙТРАЛИЗАЦИЮ ВОД КИСЛОТНОГО ШАХТНОГО ДРЕНАЖА

Методами ICP-OES (индуктивно-связанной плазма-оптической эмиссионной спектроскопии) и Jar test исследовано влияние торфа на процессы нейтрализации кислых шахтных дренажных вод (ШДВ). Использовали необработанный торф Варденисского месторождения (Армения) и кислотно-активированный торф. Под активацией торфа подразумевалось элюирование минеральной кислотой сорбированных на поверхности торфа металлов. При этом активные сорбционные группы (в частности карбоксильные) освобождаются от металлов и начинают действовать как кислые группы по катионно-обменному механизму. Введение торфа (необработанного и активированного кислотой) в ШДВ приводит к уменьшению количества щелочи, используемой для нейтрализации ШДВ. С повышением pH образуются швертманнит и басалюминит, которые сорбируют медь, никель, кобальт, цинк и др. Исследование сорбционных явлений проводилось в интервале до pH 6,15, поскольку при более высоких значениях pH на фоне более интенсивного осаждения гидроксидов металлов эффект сорбции, в плане удаления металлов из воды, становится менее заметным.