Original Research

Technogenic Soils Developed on Mine Spoils Containing Iron Sulfides in Select Abandoned Industrial Sites: Environmental Hazards and Reclamation Possibilities

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Abstract

Environmental hazards related to the weathering of iron sulfides in technogenic soils (*Technosols*) developed on sulfide-bearing mine wastes were investigated. The soil profiles studied were located on mine spoils of three abandoned industrial sites in Poland: the "Siersza" hard coal mine in Trzebinia, the "Staszic" pyrite mine in Rudki, and pyrite mines in Wieściszowice. The soils investigated were weakly developed and strongly acidic, unless neutralizing agents were present, and they contained large amounts of total sulfur, also in the form of sulfides. In some horizons of young soils from Trzebinia and Rudki, sulfide sulfur was the predominant form of sulfur. Iron sulfides in these soils were relatively poorly weathered, whereas in old soils from Wieściszowice sulfides were almost entirely weathered. The presence and weathering of sulfides in all of the soils investigated causes a risk of long-term acidification process. The study revealed the occurrence of high amounts of heavy metals and radioactive elements in investigated soils. Such properties of soils like strong acidity, as well as high contents of trace elements and sulfur (also occurring as sulfides), should be considered during reclamation and management of mine spoils containing iron sulfides.

Keywords: acid soils, *Technosols*, iron sulfides, acid sulfate weathering, heavy metals, uranium

Introduction

As a result of mining activity, vast amounts of mining wastes are deposited on land [1]. Some of them contain iron sulfides (pyrite and marcasite mainly) that intensively weather when exposed to aerobic conditions. The effect of sulfide weathering is strong acidification of waters and soils developing on mine wastes [2-4].

Mining activity causes the formation of technogenic soils. The problem of the occurrence of such soils was addressed in soil classification systems, e.g. in the World Reference Base for Soil Resources [5]. As a consequence, a new soil group – *Technosols* – was introduced in the 2nd edition of WRB soil system in 2006 [5]. *Technosols* comprise soils strongly influenced by human-made materials (e.g. mine spoils), and their properties and pedogenesis are dominated by their technical origin.

In Poland, technogenic soils (*Technosols*) strongly acidified owing to weathering of sulfides occurring on mine spoils of hard coal mines in the areas of the Upper Silesian, Lublin, and Wałbrzych coal basins [2, 3, 6, 7], as well as on mine spoils of brown coal mines, among others in the area of Bełchatów and Bogatynia [1]. Acid soils also occur in the abandoned sulfide mine districts in

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Wieściszowice, Lower Silesia [8], and Rudki (Holy Cross Mts.) [9-11], as well as in the area of the "Wiśniówka" quartzite mine near Kielce, where sulfides occur as inserts in quartzites [12].

Strong acidity caused by sulfide weathering is the main factor affecting the properties of technogenic soils developing on these mine spoils. The negative influence of strong acidity on soils is obvious, and manifested by e.g. leaching Ca, Mg, and K from soil profiles, and the increase of toxic elements in the soil solution [13, 14]. For this reason, acidic technogenic soils containing sulfides are reclaimed with the use of neutralizing agents, e.g. lime or Ca and Mg carbonates [15, 16]. Reclamation of sulfide-containing wastes may also be carried out by isolation process, i.e. covering toxic materials with a layer of fertile soil material [9].

Large amounts of sulfur and high content of toxic trace metals are another adverse property of technogenic soils containing sulfides. Elements such as Cu, Zn, Pb, Cd, and As occur in nature mostly as sulfides or sulfosalts, and may constitute a secondary component of the most common sulfide, which is pyrite. As a result of sulfide weathering, sulfur and metals are released to soils [17-20]. Along with coals and sulfide ores, also radioactive elements may occur [21-23].

The objective of the study was to determine the properties of technogenic soils containing iron sulfides in order to draw more attention to environmental hazards related to the weathering of sulfides in soils. This paper suggests certain solutions concerning the application of the results obtained in the reclamation of investigated sulfide-bearing technogenic soils.

Materials and Methods

Site and Material Description

Soil profiles developed on mine wastes (mine spoils, post-flotation sludges) were investigated (Table 1). The study areas were located in three abandoned industrial sites in Poland:

Table 1. Location and description of soils investigated.

Location	Vegetation	Description of soil profiles
Profile 1, *Spolic Technosol* (Toxic, Humic, Skeletic) Trzebinia, surface of a dump	**None**	Very weakly developed soil profile containing large amounts of crashed hard coal in the topsoil (C1 horizon) and grey loamy material in the subsoil (C2 horizon). Age: more than a dozen years.
Profile 2, *Spolic Technosol* (Toxic, Humic) Trzebinia, northern slope of a dump	**None**	Very weakly developed soil profile. The black layers (C1 and C3 horizons) contain large amounts of crushed hard coal. C2 horizon consists of grey loamy material, and C4 horizon consists of orange and rusty sandy loam. Age: more than a dozen of years.
Profile 3, *Spolic Technosol* (Calcaric, Toxic) Rudki, former flotation tank	**Meadow** the main plants: grasses (Small-reed), Creeping Thistle, *Euphorbia*	Very weakly developed and bipartite soil profile. In the subsoil (C2 horizon), brown post-flotation sludge containing carbonates and iron sulfides occurs. The topsoil (A and C1 horizons) consists of brown and orange loamy material deposited in flotation tanks during reclamation. Age: approx. 40 years.
Profile 4, *Spolic Technosol* (Toxic) Rudki, even, not reclaimed surface of the former mine waste dump Serwis	**Meadow** the main plants: grasses, mosses, lichens, and young birch	Very weakly developed soil profile developed on the border of an extremely acid area with no plant cover. The entire profile is brown and rusty, and the separated horizons (from C1 to C4) differ slightly in color. In C4 horizon, several-centimeter large rock fragments composed of iron sulfides were found. Age: approx. 40 years.
Profile 5, *Spolic Technosol* Rudki, even, reclaimed surface of the former mine waste dump Serwis	**Meadow** the main plants: grasses, mosses, lichens, and young birch	Very weakly developed soil profile developed within a neutralized area covered by grasses. The topsoil (A horizon) is characterized by near neutral reaction, whereas the subsoil is acid. Well structure and content of approximately 2% of organic carbon is characteristic for the A horizon. Age: approx. 40 years.
Profile 6, *Spolic Technosol* (Skeletic) Wieściszowice, surface of a stony dump	**Spruce and beech forest** the main plants in the groundcover: Bilberry, mosses, young beech	Weakly developed stony soil profile. In the topsoil (A1 and A2 horizons), relatively large amounts of soil organic matter (10% or more) characterized by various stages of decomposition were accumulated. In C horizon, rusty and brown spots, not forming a continuous horizon, are visible. Age: more than 100 years.
Profile 7, *Spolic Technosol* Wieściszowice, surface of a fine earth dump	**Sparse birch and pine forest** the main plants in the groundcover: grasses, mosses, young pines and birch, Common Heather	Weakly developed soil profile characterized by alternating sandy and silty-loam technogenic layers. White and grey spots as well as rusty trails resulting from oxydo-reduction (gleyic) processes and migration of iron occur in the whole profile. Age: more than 100 years.

(1) the abandoned "Siersza" hard coal mine in Trzebinia town (Silesian Uplands), closed in 1999
(2) the abandoned "Staszic" iron sulfide mine in Rudki village (Holy Cross Mts.), closed in 1971
(3) the abandoned pyrite mines in Wieściszowice village (Rudawy Janowickie Mts. Western Sudetes), closed in 1925.

The location and description of the studied soils were presented in Table 1.

Large amounts of sulfur, occurring among others in the form of iron sulfide, are typical for hard coal exploited in the "Siersza" mine [24]. Mine wastes containing iron sulfides were deposited in the neighboring dump. These dumps had not been reclaimed before the investigation shown herein was completed.

The mining exploitation of iron sulfides (mainly marcasite) in the "Staszic" mine in Rudki occurred between 1925 and 1971 [25]. Uranium ores were also exploited there for a short period immediately after World War II [26]. The remnants of the mine are flotation tanks and the former dump *Serwis*, reclaimed in the 1970s [9]. The reclamation succeeded only in part. Nowadays, places with high acidity still remain on the surface of the ground [10].

The mining exploitation in Wieściszowice occurred between 1785 and 1925 in three open pits. The object of the exploitation was pyrite present in chlorite-mica metamorphic schists [27]. Nowadays, the remnants of the mine are open pits filled with acid mine drainage, and mine spoils that were not reclaimed.

Analytical Methods

Soil properties (Table 2) were analyzed using common pedological methods [28]. The soil color (dry and wet) was determined using Munsell Soil Color Charts. The texture was determined using the Casagrande method (for the <1 mm fraction) and by dry-sieving (for the 2 and 1 mm fraction). The content of rock fragments was determined by dry-sieving. The pH (in distilled water and 1 M potassium chloride) was analyzed based on air-dry samples of fine earth using a soil/solution ratio of 1:2.5 (for organic samples the ratio was 1:10). The concentration of organic carbon was determined using dichromate oxidation techniques (modified Tyurin method; digestion reagent: $K_2Cr_2O_7$ and H_2SO_4; titrant: $FeSO_4 \cdot 7H_2O$). The content of carbonates was determined using the Scheibler volumetric method (reagent: 10% w/w HCl). Soil units (Table 1) were determined according to the World Reference Base for Soil Resources [5].

Forms of sulfur (Table 3) for the investigated soils were determined using Polish Norms [29-31]. Total sulfur (S_t) was determined with the use of a LECO SC 132 apparatus by sample combustion at 1350ºC, according to the Polish Norm PN-90-G-04514/16 [29]. Sulfate sulfur (S_{sulf}) was determined by hot extraction with the use of hydrochloric acid, according to the Polish Norm PN-77/G-04514.09 [30]. Sulfide sulfur (S_{py}) was determined by oxidation of sulfides to sulfates with the use of nitric acid, according to the Polish Norm PN-77/G-04514.11 [31]. The $S_t - (S_{sulf} + S_{py})$ value was called residual sulfur (S_r). The analyses were performed in the chemical laboratory of the Department of Economic and Mining Geology at the AGH University of Science and Technology, Kraków, Poland.

The major elements occurring in soils (Table 4) were determined using inductively coupled plasma-atomic emission spectrometry (ICP-AES). Most trace elements were determined with the use of inductively coupled plasma-mass spectrometry (ICP-MS) (Table 5). Contents of Cd, Cu, and Zn were analyzed using ICP-AES preceded by dissolution of the sample in a set of acids (hydrofluoric acid, nitric acid, perchloric acid, and hydrochloric acid). The analyses were performed in the Activation Laboratories Ltd. ACTLABS, Canada.

Results and Discussion

Properties of Soils

The soils investigated represent weakly developed technogenic soils classified according to the World Reference Base for Soil Resources [5] as diverse variants of *Spolic Technosols* (Table 1). The soil materials consist mainly of post-mining wastes occurring in soil profiles as noticeable layers (C1, C2 etc.) (Table 2). The older and the more overgrown soil profiles are, the better developed O and A horizons occur in the topsoil. The O and A horizons do not occur in the soils from Trzebinia (Profiles 1 and 2) because of the lack of plant cover (Table 1). The black layers in these soils contain crumbled hard coal deposited on the dump. In the topsoils of the Rudki (Profiles 3, 4, and 5) and Wieściszowice (Profiles 6 and 7) soils, not very thick O and A horizons occurred, because these soils were covered by meadow and forest plants (Table 1).

The influence of sulfide weathering on the soils studied was manifested by strong acidity caused by the presence of sulfuric acid. Low pH value is typical for soils containing sulfides [2, 32, 33]. The pH_{H_2O} in most soils investigated rarely exceeded 4.5 (Table 2). In technogenic soils from Trzebinia and Rudki (Profiles 1, 2, and 4), the pH_{H_2O} value drops below 3 in certain soil layers (Table 2). The highest acidification occurred in the layers in which large amounts of sulfides were present (as indicated by the observation made during field work, and by laboratory research, Table 3). Profile 3 from Rudki showed near neutral reaction (Table 2), even though vast amounts of sulfides were present (Table 3). The high pH value in that case is caused by the high content of carbonates (mainly dolomite) occurring as primary minerals in the post-flotation sludge (C2 horizon) or added to the soil during reclamation (A and C1 horizons). The upper part (A horizon) of Profile 5 had near neutral reaction, whereas the lower part of the profile was acid (Table 2). Such a feature is the result of neutralization performed during reclamation in the superficial part of the profile.

Table 2. Selected properties of investigated soils.

Profile	Depth (cm)	Horizon	Soil color		Percentage of fraction (mm)			Content of rock fragments (wt %)	pH		Content of organic carbon (wt %)	Content of car-bonates (wt %)
			moist	dry	2.0-0.05	0.05-0.002	<0.002		H_2O	KCl		
Profile 1	0-10	C1	N 1.5/0	N 3/0	80	15	5	18	2.5	2.3	-	n
	10-35	C2	N 2/0	N 4/0	44	37	19	44	2.5	2.4	10.4	n
Profile 2	0-40	C1	N 1.5/0	N 3/0	-	-	-	-	2.4	2.4	-	n
	40-76	C2	10YR 3/1	10YR 5/1	50	35	15	33	3.4	3.3	7.4	n
	76-98	C3	10 YR 2/1	10YR 4/1	44	36	20	23	3.1	3.0	14.8	n
	98-105	C4	10YR 4/4	2.5Y 6/4	76	14	10	30	2.4	2.3	1.6	n
Profile 3	0-1	Oi	-	-	-	-	-	-	-	-	-	-
	1-3	A	7.5YR 3/3	7.5YR 5/3	45	33	22	20	7.3	6.9	5.2	3.0
	3-30	C1	7.5YR 5/4	10YR 6/6	40	20	40	28	7.2	6.7	0.9	2.4
	30-95	C2	7.5YR 3/3	7.5YR 5/4	59	37	4	0	7.4	7.4	-	60.3
Profile 4	0-2	Oi	-	-	-	-	-	-	-	-	-	n
	2-7	A	10YR 4/2	10YR 5/4	69	17	14	2	4.4	4.0	2.5	n
	7-14	C1	10YR 4/4	10YR 7/4	47	29	24	12	4.3	4.1	0.6	n
	14-30	C2	10YR 3/4	10YR 6/4	51	29	20	22	3.0	2.8	0.3	n
	30-60	C3	10YR 4/4	10YR 6/3	52	26	22	24	2.7	2.4	0.3	n
	60-75	C4	10YR 4/3	10YR 6/3	54	28	18	20	3.3	3.1	-	n
Profile 5	0-1	OiOe	-	-	-	-	-	-	-	-	-	-
	1-13	A	10YR 3/1	10YR 5/2	59	30	11	9	7.4	7.1	2.0	3.3
	13-50	C1	7.5YR 4/3	7.5YR 6/3	77	10	13	15	5.0	4.2	0.2	n
	50-70	C2	7.5YR 3/2	7.5YR 5/2	56	25	19	9	4.4	3.6	0.1	n
	70-85	C3	7.5YR 4/2	7.5YR 6/2	88	5	7	31	4.7	4.0	0.1	n
Profile 6	0-1	Oi	-	-	-	-	-	-	4.8	4.3	-	n
	1-4	Oe	-	-	-	-	-	-	4.0	3.4	-	n
	4-9	A1	10YR 2/2	10YR 4/2	57	74	15	11	3.9	3.3	9.9	n
	9-10	A2	10YR 2/3	10YR 5/3	51	71	19	10	4.0	3.4	4.9	n
	10-45	C	10YR 6/6	10YR 7/6	70	75	17	8	3.9	3.5	0.7	n
Profile 7	0-1	Oi	-	-	-	-	-	-	-	-	-	n
	1-4	A	10YR 4/2	10YR 6/3	75	18	7	3	4.2	3.5	2.6	n
	4-15	AC	10YR 5/6	10YR 7/6	65	28	7	0	4.2	3.6	1.0	n
	15-30	C1	2.5Y 6/8	2.5Y 8/6	11	68	21	0	4.3	3.6	0.5	n
	30-35	C2	10YR 5/6	10YR 7/8	86	9	5	1	4.4	3.9	0.9	n
	35-86 rusty	C3	2.5Y 6/6	2.5Y 8/6	11	70	19	0	4.3	3.6	0.5	n
	35-86 white	C3	2.5Y 7/2	2.5Y 8/1	11	70	19	0	4.3	3.6	0.5	n
	86-112	C4	2.5Y 6/6	2.5Y 7/6	87	8	5	0	4.3	4.1	0.9	n

- not determined
n – Lack of carbonates

Table 3. Content and forms of sulfur in investigated soils.

Profile	Depth (cm)	Horizon	Content of sulfur (wt %)			
			Sulfate sulfur (S_{sulf})	Sulfide sulfur (S_{py})	Residual sulfur (S_r)	Total sulfur (S_t)
Profile 1	0-10	C1	0.64	0.12	0.68	1.44
	10-35	C2	0.93	0.76	0.46	2.15
Profile 2	0-40	C1	0.78	0.29	0.65	1.72
	40-76	C2	1.08	0.15	0.30	1.53
	76-98	C3	0.79	0.37	0.39	1.55
	98-105	C4	0.62	0.05	0.04	0.71
Profile 3	0-1	Oi	-	-	-	-
	1-3	A	0.13	0.61	0.12	0.86
	3-30	C1	0.13	0.32	0.16	0.61
	30-95	C2	1.30	2.76	4.16	8.22
Profile 4	0-2	Oi	-	-	-	-
	2-7	A	0	0	0.30	0.30
	7-14	C1	2.04	0	0.68	2.72
	14-30	C2	0.73	0	0.14	0.87
	30-60	C3	0.72	0	0.22	0.94
	60-75	C4	0.67	1.37	0.20	2.24
Profile 6	0-1	Oi	-	-	-	-
	1-4	Oe	-	-	-	-
	4-9	A1	0.07	0.18	0.17	0.42
	9-10	A2	0.08	0.12	0.09	0.29
	10-45	C	0.07	0.01	0	0.08
Profile 7	0-1	Oi	-	-	-	-
	1-4	A	0.01	0	0.08	0.09
	4-15	AC	0.01	0	0.03	0.04
	15-30	C1	0	0	0.04	0.04
	30-35	C2	0	0	0.05	0.05
	35-86	C3	0.03	0	0.05	0.08
	86-112	C4	0.28	0	0.16	0.44

- not determined

Forms of Sulfur in Soils

The occurrence of large amounts of sulfur in the soils investigated resulted from the presence of sulfides and products of their weathering. The highest total contents of sulfur were present in young soils from Trzebinia (between 0.71 and 2.15 wt %) and Rudki (between 0.30 and 8.22 wt %), and the lowest in old soils from Wieściszowice (between 0.04 and 0.44 wt %) (Table 3). The content of sulfur in the majority of natural Polish soils ranges from 0.007 to 0.107 wt % [34]. The differences in total sulfur contents between the soils investigated were most likely caused by different initial sulfur amounts. The percentage of the specified sulfur forms varied significantly even between adjacent soil horizons (Table 3). The results from these horizons consist of diverse industrial materials located in mine spoils by means of deposition techniques.

Young soils (Trzebinia and Rudki) contained higher amounts of sulfide sulfur than the old ones (Wieściszowice) (Table 3). Several-centimeter rock fragments composed exclusively of relatively poorly weathered iron sulfides were found during field work in the soils from Trzebinia and Rudki (Fig. 1, Table 1). Soils from Wieściszowice showed low concentrations of sulfide sulfur, and only a few sulfide crystals occurred, normally included in rock and mineral fragments. Sulfides in soils from Wieściszowice in most cases were partly or almost entirely weathered (Fig. 1).

Large amounts of sulfate sulfur occurred in technogenic soils from Trzebinia and Rudki (Table 3), caused by intense weathering of sulfides that were a relatively abundant constituent in these soils. In soils from Wieściszowice, sulfate sulfur predominated only in certain soil horizons (Table 3), e.g. in C4 horizon of Profile 7, where relatively large amounts of gypsum were determined during mineralogical research [11]. The occurrence of sulfur in the form of sulfates is typical for sulfide weathering zones [35], as well as for natural soils containing weathered sulfides [32], and similar technogenic soils developed on mine spoils [1, 3, 8-11].

In soils from Trzebinia (Profile 1 and 2), relatively large amounts of residual sulfur were found compared with other forms of sulfur (Table 3). Such a feature is most likely related to the presence of large amounts of organic sulfur occurring in crumbled hard coal (Table 1 and Table 2). The occurrence of organically-bound S is typical for hard coals [24], and soils developed on mine spoils from hard coal mines [3]. Large amounts of residual sulfur were also determined in C2 horizon of Profile 3 from Rudki (Table 3), constituting post-flotation sludge. In that case, the feature is most likely related to the presence of xanthates (or products of their decay) – organic-sulfur compounds used as flotation agents in mine processing [9].

Chemical Composition of Soils

The contents of major elements in the soils investigated differ both between and within individual soil profiles. This results from the location of diverse mine wastes during the mining activity due to deposition techniques. The chemical

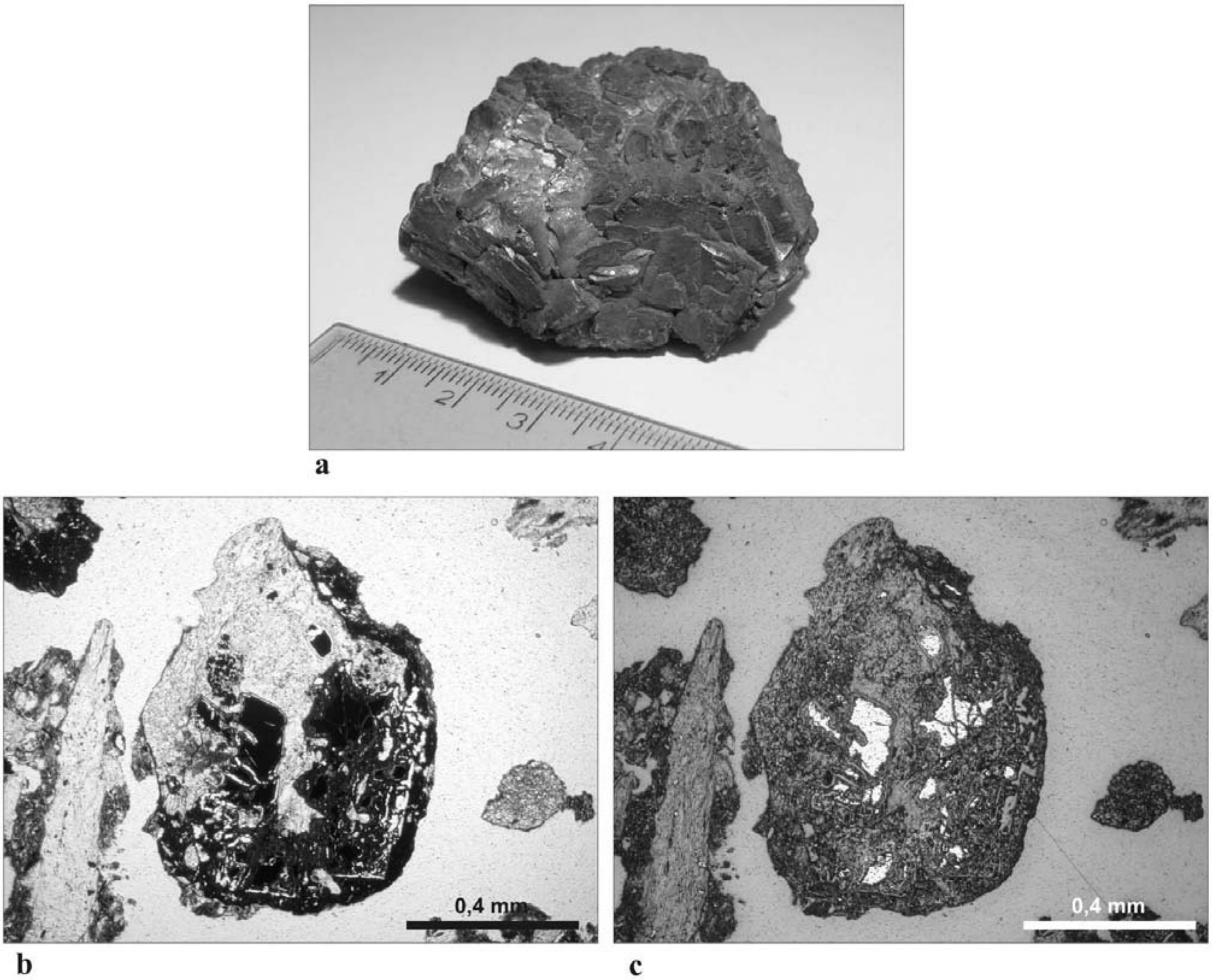

Fig. 1. a) – Rock fragment, consisting of iron sulfides, from Profile 2 (Trzebinia); b) and c) – plane polarized light and plane reflected light, respectively, optical microscope images of mineral grain, containing strongly weathered iron sulfide (light minerals on photo c), from Profile 6 (Wieściszowice).

Table 4. Chemical composition of soils – major elements.

Profile	Depth (cm)	Horizon	Chemical composition (wt %)										Loss on ignition (wt %)	Total (wt %)
			SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5		
Profile 1	0-10	C1	19.96	0.321	7.05	3.63	0.009	0.31	0.26	<0.01	0.71	0.05	67.67	99.99
	10-35	C2	44.12	0.709	16.72	4.83	0.018	0.76	1.03	0.27	2.06	0.07	28.10	98.68
Profile 2	0-40	C1	27.97	0.511	11.39	2.90	0.008	0.34	0.95	0.06	1.26	0.09	53.81	99.28
	40-76	C2	56.31	0.591	13.50	4.83	0.019	0.53	1.63	0.13	1.95	0.06	20.39	99.95
	76-98	C3	43.83	0.569	13.01	4.82	0.017	0.48	1.20	0.06	1.65	0.05	33.87	99.56
	98-105	C4	79.25	0.243	5.45	5.54	0.012	0.17	0.41	0.13	1.78	0.04	7.36	100.4
Profile 3	0-1	Oi	-	-	-	-	-	-	-	-	-	-	-	-
	1-3	A	35.65	0.625	10.10	26.48	0.341	1.47	1.77	0.28	1.60	0.15	21.55	100
	3-30	C1	38.43	0.887	14.17	27.99	0.249	1.27	0.89	0.13	2.21	0.09	13.30	99.61
	30-95	C2	5.16	0.039	1.20	17.82	0.309	12.90	21.19	< 0.01	0.14	<0.01	28.68	87.49
Profile 4	0-2	Oi	-	-	-	-	-	-	-	-	-	-	-	-
	2-7	A	73.33	0.322	5.91	6.39	0.365	0.35	0.19	0.35	1.03	0.23	10.24	98.71
	7-14	C1	63.40	0.440	8.44	5.79	0.259	0.50	4.27	0.37	1.42	0.16	13.16	98.21
	14-30	C2	74.17	0.474	7.45	6.66	0.028	0.48	1.12	0.40	1.50	0.06	7.51	99.85
	30-60	C3	72.70	0.489	8.21	6.62	0.036	0.60	1.04	0.51	1.40	0.07	7.91	99.59
	60-75	C4	74.55	0.466	6.99	6.49	0.036	0.51	0.83	0.43	1.38	0.05	7.73	99.47
Profile 6	0-1	Oi	-	-	-	-	-	-	-	-	-	-	-	-
	1-4	Oe	-	-	-	-	-	-	-	-	-	-	-	-
	4-9	A1	37.97	0.794	15.24	8.61	0.035	2.95	0.29	1.67	1.56	0.19	29.89	99.19
	9-10	A2	44.99	0.934	17.98	11.51	0.038	3.50	0.18	2.04	2.01	0.17	16.42	99.77
	10-45	C	48.15	1.894	25.45	6.98	0.028	3.38	0.07	2.19	3.80	0.16	8.08	100.2
Profile 7	0-1	Oi	-	-	-	-	-	-	-	-	-	-	-	-
	1-4	A	58.54	0.863	16.49	5.82	0.047	5.89	0.17	2.08	1.26	0.10	8.62	99.87
	4-15	AC	59.06	0.940	17.75	5.92	0.049	6.08	0.13	2.22	1.44	0.10	6.63	100.3
	15-30	C1	50.95	1.427	21.61	6.90	0.081	7.30	0.17	2.58	1.79	0.12	7.16	100.1
	30-35	C2	63.26	0.733	15.74	4.89	0.042	5.65	0.11	2.25	1.09	0.10	5.31	99.18
	35-86	C3	51.17	1.310	21.82	6.67	0.049	6.92	0.16	2.28	1.94	0.11	7.26	99.68
	86-112	C4	62.47	0.841	16.23	3.88	0.041	5.75	0.84	2.15	1.20	0.08	6.02	99.50

- not determined

composition of technogenic materials, especially the content of macronutrients (e.g. Ca, Mg, Na, K, N, P) and trace elements, is thought to be one of the most important properties that should be taken into account during their examination, as it may influence the success of their reclamation and subsequent plant growth [3, 6].

The chemical composition of investigated soils depends on their mineral composition [11]. Si and Al predominate in the studied soils from Trzebinia (Profiles 1 and 2, Table 4). Higher Si contents are indications of quartz abundance, whereas the occurrence of higher amounts of Al indicates that aluminosilicates (e.g. feldspars or layer silicates) are present in higher amounts. Fe is also present in the soils in the form of sulfide minerals and also as products of sulfide weathering (e.g. iron oxides) [11].

The investigated soils from Rudki (Profile 3 and 4) represent different mine wastes that cause differences in chemical composition between the soils. Profile 3 can be divided into two parts. The lower part of Profile 3 (C2 horizon), constituting post-flotation sludge, contains mainly Ca, Mg, and Fe (Table 4), as it contains dolomite, gypsum, and iron sulfides (marcasite and pyrite). In the chemical composition of the upper part of Profile 3 (A and C1 horizon), Si, Al, and Fe predominate (Table 4), as large amounts of quartz, layer silicates (mica and kaolinite), and iron oxides (goethite and hematite) are present. In the entire Profile 4 from Rudki, Si predominates because quartz is the main mineral occurring in the profile. Al and Fe are also present [11].

Si and Al are predominating elements in both investigated soils from Wieściszowice (Profile 6 and 7) (Table 4), as quartz, feldspars, and layer silicates (muscovite, paragonite, chlorite, and others) predominate in the mineral composition of soils. Larger amounts of Na in comparison with soils from Trzebinia and Rudki result from the presence of albite and paragonite, and larger amounts of Mg – from the occurrence of chlorite. Fe present in soils from Wieściszowice occurs mainly as iron oxides – the effect of pyrite weathering [11].

The soils investigated are potentially contaminated with trace metals, as they contain relatively high amounts of such elements as Pb, Ga, As, Tl, Cr, Co, Cu, and V (Table 5). The contents of the elements mentioned are higher than acceptable ones specified by Kabata-Pendias and Pendias [36]. The contents of Zn, Cd, Sb, Sn, Zr, Ni, and Cs are high, but they do not exceed values specified by Kabata-Pendias and Pendias [36] in any of the investigated soil profiles (Table 5). High contents of trace metals are typical for mine spoils of hard coal and metal ore mines [e.g. 33, 37], and are related mainly to the release of metals in the process of sulfide weathering. The investigation by Lu et al. [20] showed that some trace elements (e.g. As, Cu, and Zn) may be immobilized by adsorption on products of pyrite weathering (e.g. on iron oxides). The process most likely also concerns soils studied in this paper, and it may temporarily decrease the risk of mobilization of trace elements to the soil environment and ground waters during sulfide weathering.

Large amounts of radioactive elements – U and Th – are present in soils from Rudki and Trzebinia (Table 5). Comparison of the data obtained with the data specified by Kabata-Pendias and Pendias [36] reveals that the content of U in C1 horizon from Profile 3 from Rudki (23.2 mg/kg), containing the largest amounts of this element, is several times higher than the maximum specified by Kabata-Pendias and Pendias [36] (2.3 mg/kg). The occurrence of radioactive elements in soils from Rudki is related to the occurrence of polymetallic uranium mineralization in sulfide ores [26], and subsequent deposition of contaminated materials on the land surface. U and Th are common constituents of hard coals [22], and that is the reason for the occurrence of high contents of radioactive elements in technogenic soils developed on mine wastes from the hard coal mine in Trzebinia (Profile 1 and 2, Table 5). The presence of radioactive elements in the studied soils may cause a toxicological hazard, as those elements might be taken by plants in excessive amounts [e.g. 38], but it may also pose a problem caused by radiation released during the decay of radioactive elements, which may influence plants and animals living in the area of mine spoils [23, 36].

Application of Results in Reclamation and Management of Technogenic Soils Developed on Mine Spoils Containing Iron Sulfides

Long-lasting strong acidity caused by weathering of sulfides is the main reason the relation of sulfide-bearing mine wastes are reclaimed. The results obtained revealed that acid reaction is characteristic not only for young soils from Trzebinia and Rudki, but also for soils from Wieściszowice, which are older than 100 years, because iron sulfides are still present in these soils (Fig. 1). This indicates that complete decay of sulfides is not achieved during such a period (at least 100 years), and it probably needs much more time. The occurrence of sulfides and their weathering is responsible for the persistence of acid reaction in soils.

Despite strong acidity in soils (pH_{H_2O} ranging from 3.9 to 4.8) and the occurrence of partly weathered iron sulfides (Fig. 1), reclamation is unnecessary in the case of the soils developed on mine spoils in Wieściszowice. Ecological succession, which started after the cessation of mine spoils deposition (i.e. several tens or hundreds of years), contributed to the formation of forest communities, which resulted in the development of soil profiles with large amounts of soil organic matter in the topsoil (Table 2). Moreover, the soils developed on mine spoils are quite similar, with regard to their chemical properties (e.g. acidity), to soils occurring on natural outcrops of pyrite-bearing rocks in the vicinity of the abandoned mines in Wieściszowice [8, 11]. Furthermore, along with outflow of waters outside the weathering zone in Wieściszowice, a decrease of acidity of waters and a decrease of trace elements content in waters occur, which indicates that there is a low risk of the migration of pollutants from mines to the surrounding surface waters [35].

Unlike the Wieściszowice region, reclamation is necessary on mine spoils in Trzebinia and Rudki, e.g. because of strong acidity of soil materials (stronger than in Wieściszowice, Table 2), and high contents of trace elements. Moreover, mine spoils in Trzebinia and some of the areas in Rudki are soil-less terrains that need to be reclaimed (e.g. by covering with plants) to redress the ecological balance and restrain the erosion.

On the dump surface of hard coal mine in Trzebinia, reclamation has not been executed, because the mine was closed unexpectedly. Therefore, the dump requires the execution of full reclamation process. Manners of management of wastes from hard coal mines were widely discussed in Poland [1-3, 6, 15, 39-41]. Agricultural and forest management of such wastes was proposed [2, 3, 6, 42], and the latter is more frequently applied.

The results obtained revealed that one of the most serious problems concerning reclamation of wastes from hard coal mines is high content of rock fragments and intense weathering of superficial layers of sulfide-bearing mine spoils, causing strong acidification. Such features commonly occur on wastes from hard coal mines, and were described previously [e.g. 2, 3, 6]. Another problem that needs to be considered during reclamation is the high content of trace metals (e.g. Pb, Ga, Cr, Zn, and Cu) and

Table 5. Chemical composition of soils – trace elements.

Profile	Depth (cm)	Horizon	Chemical composition (mg/kg)																	
			Zn	Cd	Cu	Pb	Ga	As	Tl	Sn	Sb	Bi	Cr	Co	Ni	Zr	V	Cs	Th	U
			*300**	*3**	*100**	*100**	*3-12***	*20**	*0.02-2.8***	*50**	*10**	*0.2-1.5***	*100**	*30**	*100**	*90-550***	*150**	*0.1-26***	*1.4-10***	*0.1-2.3***
Profile 1	0-10	C1	69	<0.5	72	38	8	<5	<0.1	<1	2.6	<0.4	40	5	38	86	71	3.9	7.5	**3.6**
	10-35	C2	77	<0.5	29	**104**	**24**	6	1.5	1	2.9	<0.4	**110**	8	37	126	116	16.7	**14.2**	**5.4**
Profile 2	0-40	C1	94	0.9	45	57	**15**	17	1.3	2	3	<0.4	70	4	23	106	101	10.1	**13.3**	**4.7**
	40-76	C2	145	<0.5	53	74	**18**	6	0.3	3	7.7	<0.4	80	6	29	138	91	12.2	**10.5**	**4.2**
	76-98	C3	280	1.6	41	65	**16**	<5	<0.1	2	3.8	<0.4	70	5	26	123	88	10.5	**11.2**	**4.6**
	98-105	C4	39	<0.5	23	27	7	<5	0.8	1	0.5	<0.4	40	3	10	148	30	3.3	6.3	1.5
Profile 3	0-1	Oi	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	1-3	A	192	1.4	64	**1170**	9	**26**	0.5	<1	0.5	<0.4	70	17	50	243	99	4.9	8.8	**16.6**
	3-30	C1	146	1.2	55	**1490**	**19**	**41**	**7.3**	2	2.9	<0.4	**120**	17	47	271	**160**	8.6	**15.9**	**23.2**
	30-95	C2	133	1.1	23	**229**	1	<5	1.6	<1	<0.5	<0.4	<20	4	26	18	21	<0.5	0.8	**6.4**
Profile 4	0-2	Oi	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	2-7	A	119	1.3	17	**122**	6	10	0.3	<1	1.9	<0.4	30	25	42	179	38	1.9	4.9	**19.3**
	7-14	C1	106	0.7	21	77	10	8	1.1	<1	0.9	<0.4	50	**40**	46	208	52	3.4	7	**14.7**
	14-30	C2	47	<0.5	14	78	10	8	1.3	<1	0.7	<0.4	40	7	21	222	60	3.2	6.3	**3.2**
	30-60	C3	58	<0.5	13	59	10	8	0.9	<1	6.2	<0.4	50	9	25	229	59	3.4	6.8	**3.2**
	60-75	C4	54	<0.5	15	**126**	10	6	0.7	<1	<0.5	<0.4	50	7	20	261	48	2.6	6.4	**9.4**
Profile 6	0-1	Oi	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	1-4	Oe	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	4-9	A1	129	<0.5	81	**109**	**17**	**38**	0.3	6	5.6	<0.4	**100**	7	20	64	**264**	3	2.1	1.4
	9-10	A2	109	<0.5	**101**	70	**21**	**98**	0.3	9	9.7	0.7	**110**	7	24	78	**320**	3.2	2.4	1.7
	10-45	C	73	<0.5	22	36	**21**	**32**	0.5	7	1.7	<0.4	**130**	2	10	113	**536**	4.9	1.7	2.1
Profile 7	0-1	Oi	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	1-4	A	89	<0.5	89	23	**22**	17	1	9	2.4	<0.4	**100**	3	15	54	**255**	9.8	1.3	0.8
	4-15	AC	86	<0.5	72	10	**22**	16	0.4	5	2.2	<0.4	**100**	2	14	59	**287**	2.3	1.2	0.8
	15-30	C1	115	<0.5	36	16	**26**	19	0.5	5	1.3	<0.4	**100**	2	12	95	**443**	3.7	0.9	1.0
	30-35	C2	95	<0.5	70	11	**18**	**26**	0.2	3	2.4	<0.4	90	1	10	42	**250**	1.2	0.5	0.6
	35-86	C3	84	<0.5	66	13	**31**	**27**	0.5	15	2	<0.4	**150**	2	13	91	**399**	4.3	1.1	1.2
	86-112	C4	62	<0.5	61	5	**20**	11	0.3	3	2.1	<0.4	70	1	8	46	**259**	1.4	0.6	0.6

- not determined

* Maximum permissible quantities of trace elements in soils according to Kabata-Pendias and Pendias [36].

** Lack of standards regulating permissible quantities of trace elements in soils. Ranges given concern quantities of trace elements most frequently occurring in uncontaminated soils according to Kabata-Pendias and Pendias [36]. Maximum values given concern quantities of trace elements determined in soils occurring in industrial sites.

Soil samples for which quantities of trace elements are equal to or higher than those given by Kabata-Pendias and Pendias [36] are in bold in the table.

radioactive elements (U and Th) (Table 5). The results concerning the occurrence of radioactive elements in mine wastes from hard coal mines, presented in this paper, were also confirmed by other authors [43]. One more unfavorable feature of technogenic soils developed on mine spoils in Trzebinia is the mineral composition of the clay fraction. The clay fraction is predominated by kaolinite and mica, whereas swelling minerals (smectite and vermiculite), which may increase cation exchange capacity, occur in minor amounts [11]. The mineral composition of the clay fraction of mine wastes is thought to be one of the most important factors influencing the success of reclamation of wastes from hard coal mines [6, 44, 45].

Reclamation in the area of the abandoned sulfide mine in Rudki, carried out in the 1970s [9], succeeded only in part, as confirmed by the results presented in this paper and described by other authors [10]. Reclamation in the area of the former flotation tanks (Profile 3), consisting in covering post-flotation sludge with a layer of loamy material from ground stocks deposited by the mine, did not bring the expected results. The high content of colloidal clay and iron oxides in the loamy material occurring in the topsoil (Table 2), as well as high compaction of the post-flotation material in the subsoil, cause high cohesion of technogenic soil material [11]. The unfavorable physical properties mentioned above hinder rooting of plants, which is why the plant cover on the former flotation tanks is still sparse after several dozen years.

Despite reclamation consisting of double neutralization of sulfide-bearing wastes in the region of the former dump *Serwis* near Rudki with the application of lime and phosphorite flour [9], extremely acid ground patches with no plant cover still occur there (Profile 4). There are places in the neighborhood of such bare patches where reclamation was successful, and technogenic soils with near neutral reaction in the topsoil occur in such places (Profile 5). Supplementary neutralization is necessary in the majority of acid grounds in the area of the former dump *Serwis*.

One of the most hazardous properties of technogenic soils in Rudki is the high content of trace metals. High contamination by Pb, nearly fifteen times higher than permissible quantities specified by Kabata-Pendias and Pendias [36] (Table 5), was typical for the most contaminated soil (Profile 3) located on the former flotation tanks. High contents of other trace metals (e.g. Ga, As, Tl) were also determined in that profile. A serious environmental problem concerning soils developed on mine spoils in Rudki, both flotation tanks, and *Serwis* dump, is the occurrence of radioactive elements, particularly U. The amount of radioactive elements in these soils was considerably higher than maximum values specified by Kabata-Pendias and Pendias [36] (Table 5). As mentioned above, technogenic soils developed on flotation tanks in Rudki may also be contaminated with xanthates or products of their decay (e.g. dixanthogenate). Chemical contamination has to be taken into account during the management of mine spoils in Rudki. A repeated reclamation is suggested in the case of technogenic materials in the area of the abandoned sulfide mine in Rudki [10, 46].

Conclusions

1. The investigated soils from Trzebinia, Rudki, and Wieściszowice, developed on mine spoils containing iron sulfides, are weakly developed technogenic soils (*Technosols*). The weathering of soil materials containing iron sulfides and the accumulation of soil organic matter, if plant cover occurred, are the most important processes taking place in the soils studied and forming their properties. The weathering of sulfides causes strong acidification, unless neutralizing agents occur.
2. A high content of total sulfur, resulting from the presence of sulfides and products of their weathering, is typical for the soils investigated. Crystals or aggregates of iron sulfides were relatively poorly weathered in young technogenic soils from Trzebinia and Rudki. In some horizons of these soils sulfide sulfur was predominating form of sulfur. In old soils from Wieściszowice sulfides were almost entirely weathered.
3. The soils studied contain high amounts of heavy metals (e.g. Pb, Ga, As, Cr, and V), which most likely are released to soils in the process of sulfide weathering. Moreover, the occurrence of radioactive elements – U and Th – is characteristic for soils from Rudki and Trzebinia.
4. The case of Wieściszowice soils shows that it is possible to form relatively well-developed soils on mine spoils containing iron sulfides without any human help. However, some conditions need to be fulfilled, e.g. a sufficient period of time (several tens and hundreds of years) or not very strong acidity (pH ~4 or higher). Despite such possibilities, however, reclamation of soils containing iron sulfides should be performed, because of their unfavorable properties caused by weathering of sulfides. The reclamation of soils investigated should consider, first of all, strong acidity and chemical contamination, e.g. related to the occurrence of trace elements.

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