MINESOIL WEATHERING ON KITTANNING ABANDONED MINE LAND (AML) SITES IN WEST VIRGINIA[1]

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Abstract. A study was conducted to assess minesoil weathering as it affects remining and reclamation of 4 different-aged, abandoned mine land (AML) sites that were mined for Lower Kittanning coal in northern West Virginia. The products of acid sulfate weathering and remaining pyritic sulfur (S) contents were used to evaluate the weathering process and estimate future weathering. On each site, 3 1-m deep pits were dug and minesoil samples were extracted from 3 discernable horizons. Samples were analyzed for pH, exchangeable acidity, and S forms and amounts (pyritic, sulfate, and non-extractable). Active and exchangeable acidity increased and relative pyritic S decreased with age in the top horizons due to pyrite oxidation. Active and exchangeable acidity were relatively constant throughout the profile on each site. The youngest and oldest sites had constant amounts of pyritic S (about 13% on the youngest and 0% on the oldest) throughout each of their individual profiles. The 2 middle-aged sites had 8%, 1 to 2%, and about 25% relative pyritic S in the top, middle, and bottom horizons, respectively. The 3 older sites had increased levels of sulfate S in the middle and bottom horizons which was attributed to leaching of sulfates from the top horizons into the lower horizons. The top horizon on the oldest site had the lowest pH, highest exchangeable acidity, next-to-lowest relative sulfate S content, and the lowest level of pyritic S; and no pyritic S in the middle and bottom horizons. Thus, this 35-year-old site may be near the end of the acid generating phase.

ADDITIONAL KEY WORDS: Acidity, pyrite, remining, reclamation, sulfur, sulfuricization.

Introduction

Throughout central Appalachia there is an abundance of abandoned mine land (AML). In West Virginia, approximately 34,000 hectares of mined land were designated as AML in 1977 (Soil Consevation Service 1979). By definition, these lands were mined and abandoned in an inadequate reclamation status before 1977.

The Surface Mining Control and Reclamation Act (SMCRA) of 1977 provides for an abandoned mine land reclamation fund to be used for reclamation of AML sites. The fund is generated by taxing coal mine operators and is projected to generate $3 billion during its 15 year tenure. The U.S. Congress (1977) realized that this fund would be inadequate to reclaim all AML sites and consequently established the following priority system for reclaiming such sites:

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1. Protection of public health and property from extremely dangerous AML sites;
2. Protection of public health from adverse AML sites;
3. Restoration of the environment on AML sites;
4. Research on reclamation and water-quality control;
5. Protection or enhancement of public facilities on AML sites;
6. Development of publicly-owned AML sites;

In West Virginia, the focus of AML reclamation has been directed toward priorities 1 and 2. This emphasis is very unlikely to change because the reclamation cost of identified priority 1 and 2 sites has been estimated to be $2.3 billion, and to require an estimated 100 years of work (West Virginia Mining and Reclamation Association 1989). Consequently, the Office of Surface Mining (OSM) foresees that only about 10% of the nation's AML problems will be corrected within the tenure of this program. Since 1979, the AML program in West Virginia has reclaimed approximately 4% of their total AML area (Neil Robinson, West Virginia Department of Energy, personal communication). Therefore, alternative solutions must be found to help reclaim remaining AML sites. One possible solution is remining.

Most coal states have had some degree of industry-based reclamation where coal companies reclaimed on-site or adjacent AML problems while mining with current permits (Blauch 1986). However, remining and reclamation of AML sites has declined especially in areas where operators may become responsible for existing AML problems such as acid mine drainage and refuse piles. Therefore, the mining industry, OSM, and researchers are attempting to find methods for promoting remining and reclamation of AML sites that would maximize environmental improvement and resource recovery.

It is known that in some cases, remining and/or reclamation may create a worse environmental problem than was initially present on AML sites (Lindsay and Nawrot 1981). For example, an unsuspecting operator may remine what appears to be a relatively environmentally harmless AML site (i.e., abundant vegetation cover, neutral soil pH, and little or no acid mine drainage (AMD)), but in the process expose buried pyritic sulfur (S) materials that would generate high amounts of AMD and dramatically increase reclamation costs. Therefore, understanding the properties of minesoils on AML sites (especially in terms of acidity) may be helpful to assess the site's potential for reclamation.

Three forms of acidity (active, exchangeable, and potential) should be taken into account when assessing AML sites for remining or reclamation. Active acidity, expressed as pH, is a measurement of H^+ ion activity in the soil. Exchangeable acidity is correlated to exchangeable hydrogen, aluminum, and iron and their hydroxy polymers (Thomas and Hargrove 1984). Extraction by KCl is one method to measure exchangeable acidity. Both of these acidities relate to acid that is already present in the soil. Potential acidity is that acidity that may be generated as a result of unoxidized pyritic S (Caruccio et al 1988). This oxidation occurs through a series of reactions that are limited by the supply of oxygen (Fanning and Fanning 1989). Oxidation of these unweathered minerals can release enormous amounts of hydrogen, iron, and sulfate with time, thus greatly increasing the lime needed to neutralize the acidity generated in the soil (Skousen 1987).

Sulfuricization is the process whereby 1) reduced S materials are oxidized, 2) the generated sulfuric acid degrades other minerals present in the soil, and 3) new mineral phases are formed from the dissolution products (Carson et al 1982). The dissolved products of sulfuric acid weathering (measured by active, exchangeable acidity, and sulfate S) can be leached into lower horizons and/or precipitate in response to Eh or concentration factors (Fanning and Fanning 1989).

The purpose of this paper is to assess the amounts and types of acidity present in different-aged, Kittanning AML minesoils. The products of acid sulfate weathering and remaining pyritic S contents were used to evaluate the weathering process and to estimate future weathering. Relative amounts and forms of acid in minesoils are important when considering remining and reclamation of AML sites because buried sulfide materials, upon exposure, may create a worse environmental problem than was present before redisturbance.

Materials and Methods

A total of 4 AML sites in West Virginia were sampled. The AML inventory list developed by the West Virginia Department of Energy was used as the pool of available sites for selection. This pool was then reduced to sites with south- to west-facing highwall aspects to limit micro-climate variation (Hicks and Frank 1984). These available sites were then examined to identify the coal seam that had been mined by using General and Economic Geology maps (Hennen and Reger 1913; Reger 1931; and Reger et al 1918). After determining the coal seam mined on each site, four AML sites from the Kittanning coal seam were randomly selected to reduce the variation among parent materials of the total pool of AML sites. Time since abandonment was not purposely selected; however, a conservative age of each site was obtained by aging tree cores. Table 1 lists the West Virginia county where each site was located, site age, and a short description of the vegetation cover.

The soil was sampled at each AML site from 3 randomly selected soil pits. These pits were located by placing a grid over a map of each site and randomly selecting 3 points using computer generated random coordinates. Pits were then dug to a depth of 1 m and described. Soil samples were collected from 3 discernable horizons (top, middle, and bottom). Soil samples were air dried and sieved to <2 mm (#10 mesh sieve). Samples were analyzed for pH using a 1:1 soil/water ratio (Sobek et al 1978). These soil samples were analyzed for exchangeable acidity using a 1 N KCl extraction and the extracts were then titrated with 0.02 N NaOH to determine exchangeable acidity (Thomas 1982).

Soil samples were then hand-ground using a mortar and pestle to <250 micrometer (#60 mesh sieve). The <250 micrometer samples were separated into 3 0.5 g subsamples and analyzed for total S and fractionated for S forms (sulfate, pyritic, and non-extractable) according to the method described by Sobek et al (1978). Non-extractable S in minesoils includes primarily organic S, but also contains some amount of pyritic S encased in soil particles. Although this would indicate that there is an unaccounted store of pyritic S, this pyrite generally would have a low tendency for oxidation under natural conditions, since it withstood the nitric acid leachings. Results for each analysis were averaged from the 3 pits, analyzed by ANOVA, and relationships between age and measured parameters in each horizon were determined by linear regression.

Although additional chemical and physical analyses have been conducted, this paper reports only on pH, exchangeable acidity, and S forms and amounts found in these minesoils. In addition, average morphological properties of the pits are reported.

Results and Discussion

In the following discussion, the top horizon will primarily be used for comparison between sites because the surface horizon was assumed to be the most oxidized layer in each profile, and had not received weathering products from leaching that could mask the effects of sulfuricization. Not surprisingly, pH decreased with age (Table 2) while exchangeable acidity increased with age (Table 3). Coefficient of determination (r^2) values between age and these acid parameters were very high.

The S forms and amounts in the minesoils of our 4 sites are shown in Table 4. There was no trend in any S form with age probably due to parent material differences in S content of each site. To better understand the proportions of S in each form, relative amounts were calculated and are displayed in Table 5. Relative pyritic S decreased with age in the top two horizons indicating increased pyrite oxidation with age. As the degree of pyrite oxidation increased with age, more sulfuric acid (hydrogen and sulfate) was generated, thereby lowering the pH and increasing exchangeable acidity in these horizons.

The fact that pH and exchangeable acidity were relatively constant with depth on each one of our West Virginia sites (see Tables 2 and 3) would indicate leaching and/or a constant amount of pyrite oxidation throughout the profile. This presumed constant amount of pyrite oxidation, however, was not evident when looking at pyritic S with depth. The youngest site (CAR) and the oldest site (BAK) each had a similar amount of relative pyritic S throughout their individual profiles (Table 5). However, the 2 middle-aged sites (CO and RB) had their lowest relative pyritic S content in their middle horizons and their highest relative pyritic S in the bottom horizons. This low level of pyrite oxidation in the bottom horizons could be due to a reduced zone because oxygen controls to a great extent the rate of sulfuric acid production (Fanning and Fanning 1989). Although the soil matrix chromas (Tables 6 and 7) did not consistantly indicate that the bottom horizons of these sites were reduced, this does not preclude that these horizons may have low oxygen concentrations. The colors could be artifacts of parent material mixing during mining and not reflect actual redox conditions or not enough time has elapsed for those colors to be observable. The presence/absence of roots may be an additional indicator of redox conditions on minesoils. On our AML sites there were no roots in the bottom horizons of the RB and CO sites, and roots were present in all the horizons of the old and young sites (Tables 6 and 7).

The middle horizons on the two middle-aged sites had low relative pyritic S. It is not clear why pyritic S was lower in this horizon. Perhaps, the balance of moisture and oxygen may have favored pyrite oxidation above the reduced bottom horizon.

The top horizon of the oldest site (BAK) exhibited the lowest pH, the highest exchangeable acidity, next to the lowest relative sulfate S content, and the lowest relative pyritic S than any other site. Increased levels of sulfate in the middle horizon on this site indicates leaching of this mobile anion to lower levels (Horbaczewski and Van Ryn 1988).

By comparison, a more extended chronosequence (3 months to 50 years) of minesoils was conducted in Texas (Skousen et al 1990). In that study, the pH of surface horizons decreased with age for the first 25 years, but then pH gradually increased until the 50-year-old site had pH values similar to the youngest sites. The authors felt that most of the acid-producing materials were completely oxidized on the 50-year-old site and that the acidic products had leached from the surface. They also hypothesized that the younger sites would progress through a similar pyrite oxidation-leaching process and eventually increase minesoil pH.

The relative non-extractable S (see Table 5) showed no distinct trend with age but generally decreased with depth. The relative amount of non-extractable S in the top horizons should be higher due to reaction of pyrite and the leaching of sulfate. With depth, less pyrite should have reacted and sulfate should have accumulated, thereby reducing the relative non-extractable S. Another possible reason for increased non-extractable S in the surface is the formation of new mineral phases from pyrite oxidation (Carson et al 1982).

Non-extractable S was the most dominant S form in every horizon (see Tables 4 and 5). To determine if the non-extractable S form included some pyrite and sulfate, a few minesoil samples were shattered in a shatterbox for 2 minutes followed by the HCl and HNO_3 leachings. This experiment was conducted on the top horizon samples of the BAK site and yielded 1.12 times more total S, an increase from 1 to 10% relative pyritic S, a 6 to 11% increase in relative sulfate S, and a decrease from 92 to 79% in relative non-extractable S. The additional relative pyritic S and sulfate S were assumed to have been made more available for leaching by the shattering procedure. However, it is doubtful that the additional pyrite found through this procedure would actually react to produce acidity under natural conditions. Renton et al (1988) determined that about 35% of the original pyrite remained unreactive after extensive simulated weathering tests because the pyrite was encapsulated in the rock matrix or chemical kinetics terminated the reaction.

One of the objectives of this study was to evaluate the future weathering of these minesoils and the length of time that may be required for the majority of pyrite to be oxidized followed by leaching. This process can be assessed by the changing proportions of S with depth and by checking pH and exchangeable acidity values. Due to the relative amounts of pyritic S and sulfate S in the top horizons of the 3 younger sites, it appears that these sites were in the early- to mid-stages of acid sulfate weathering. The oldest site, on the other hand, had very low amounts of pyritic S and the sulfate S appeared to have been leached to the middle horizon. This site, despite its low pH and high exchangeable acidity, may be near the end of the acid generating stage because the oxidizable pyrite has been exhausted in all three horizons. Therefore little acidity will be generated unless some type of redisturbance occurs. Redisturbance could either break-up the rocks exposing more pyrite or could bring up reduced materials from below 1 m deep. In cases where virtually all the pyrite has reacted in the minesoil, the addition of lime to neutralize exchangeable acidity may be all that is necessary to amend the site for plants to invade and proliferate. For a major redisturbance, such as remining or AML reclamation, a few rock samples should be extracted to the depth of disturbance to evaluate the acid-producing potential of those rocks. S fractionation and/or soxhlet extractions (Renton et al 1988) would be better indicators of acid potential than traditional soil tests.

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Table 1. West Virginia county, site age (yrs), and vegetation cover of 4 different-aged, unreclaimed Lower Kittanning coal seam minesoils.

	SITES			
	CAR	CO	RB	BAK
County	Barbour	Randolph	Monongalia	Barbour
Age	13	25	27	35
Vegetation cover	Nearly total herb cover and few trees	Mix of vegetation islands, grassy meadows, and forested areas	Mix of vegetation islands, grassy meadows, and forested areas	Mostly vegetation islands and some forested areas

Table 2. Average pH and coefficient of determiniation (r^2) between age (x) and pH (y) of top, middle, and bottom horizons of different-aged, unreclaimed Lower Kittanning coal minesoils.

	SITES AGE				r^2
	CAR 13	CO 25	RB 27	BAK 35	
HORIZON					
TOP	5.3*	4.3	4.1	3.8	0.97
MIDDLE	4.7	3.5	4.1	3.5	0.84
BOTTOM	5.2	4.1	4.2	3.6	0.99

*Average from 3 pits

Table 3. Average exchangeable acidity (cmol+/kg) and the coefficient of determination (r^2) between age (x) and acidity (y) of top, middle, and bottom horizons of 4 different-aged, unreclaimed Lower Kittanning coal minesoils.

	SITES AGE				r^2
	CAR 13	CO 25	RB 27	BAK 35	
HORIZON					
TOP	3.23*	7.82	9.86	13.06	0.99
MIDDLE	3.90	11.40	7.84	16.98	0.84
BOTTOM	0.23	7.06	8.74	11.41	0.99

*Average from 3 pits

Table 4. Average percent S (total, pyritic, sulfate, and non-extractable) and the coefficient of determination (r^2) between age (x) and sulfur percentages (y) of top, middle, and bottom horizons of 4 different-aged, unreclaimed Lower Kittanning coal minesoils.

	SITES AGE				r^2
	CAR 13	CO 25	RB 27	BAK 35	
HORIZON					
TOP					
Total S	0.098	0.170	0.100	0.170	0.41
Pyritic S	0.009	0.008	0.010	0.002	0.50
Sulfate S	0.008	0.033	0.011	0.007	0.00
Non-ext S**	0.081	0.130	0.079	0.160	0.54
MIDDLE					
Total S	0.087	0.223	0.119	0.228	0.57
Pyritic S	0.004	0.000	0.001	0.000	0.85
Sulfate S	0.010	0.052	0.018	0.034	0.25
Non-ext S**	0.072	0.171	0.101	0.194	0.66
BOTTOM					
Total S	0.101	0.262	0.162	0.222	0.48
Pyritic S	0.023	0.082	0.029	0.000	0.05
Sulfate S	0.010	0.020	0.028	0.013	0.08
Non-ext S**	0.069	0.161	0.105	0.209	0.77

*Average from 3 pits.

**Non-ext S = non-extractable S (mostly organic S, but also contains some amount of pyrite and sulfate encased in soil particles).

Table 5. Average relative percent S (pyritic, sulfate, and non-extractable) and the coefficient of determination (r^2) between age (x) and relative sulfur percentages (y) of top, middle and bottom horizons of 4 different-aged, unreclaimed Lower Kittanning coal minesoils.

	SITES AGES				r^2
HORIZON	CAR 13	CO 25	RB 27	BAK 35	
TOP					
Pyritic S	13	8	8	1	0.94
Sulfate S	5	16	8	6	0.02
Non-ext S**	82	76	83	92	0.38
MIDDLE					
Pyritic S	13	1	2	0	0.82
Sulfate S	9	17	7	18	0.30
Non-ext S**	78	82	91	81	0.15
BOTTOM					
Pyritic S	15	23	27	0	0.14
Sulfate S	14	5	9	7	0.45
Non-ext S**	71	72	63	92	0.32

*Average form 3 pits per site.

**Non-ext S = non-extractable S (mostly organic, but also contains some amount of pyrite and sulfate encased in soil particles).

Table 6. Average morphological properties* of 2 unreclaimed, Lower Kittanning coal minesoils (CAR & CO).

Hor	Depth	Matix colors	Mottles	Struc	Grav	Cob	Roots	Bndy	Tex
CAR									
Top	0-4 cm	10YR 5/3, brown 10YR 4/2, d grayish brown	cff 2.5Y 2/0, black	1 f cr	5	--	mf	a,s	SL
Mid	12-42	10YR 5/3, brown 10YR 5/8, yellowish brown	cfd 2.5Y 2/0, black mfp 7.5YR 5/8, s brown cfd 10YR 6/1, gray	1 vf sbk to m	32	--	cf	a,s	CL
Bot	48-93	10YR 5/3, brown 10YR 5/6, yellowish brown	cfd 2.5Y 2/0, black mfp 5YR 2.5/2, d reddish brown mfd 10YR 6/1, gray	m	15	15	cf	--	SCL
CO									
Top	0-5 cm	2.5Y 4/4, olive brown 10YR 3/2, v d grayish brown	mfp 2.5Y 2/0, black cmd 10YR 5/6, s brown	1 vf cr to m	32	--	cf	a,s	L
Mid	16-44	2.5Y 4/4, olive brown 7.5YR 5/6, s brown 10YR 6/6, brownish yellow	mfp 2.5Y 2/0, black mmp 2.5Y 5/2, grayish brown mmp 10YR 5/6, yellowish brown	m	45	--	ff	a,s	L
Bot	56-94	2.5Y 4/2, d grayish brown 10YR 6/4, l yellowish brown	mfd 2.5Y 2/0, black mmp 7.5YR 4/4, olive brown cfp 10YR 4/6, d yellowish brown	m	18	20	--	--	L

*Codes are as follows: Hor = Horizon; Struc = Structure; Grav = Gravels %(v/v); Cob = Cobbles %(v/v); Bndy = Boundary; and Tex = USDA Texture. Colors: d = dark; v = very; l = light; and s = strong. Mottles abundance: c = common; f = few; and m = many. Mottles sizes: f = fine; m = medium. Mottles distinctness: f = faint; d = distinct; p = prominant. Structure: 1 = weak, m = massive; f = fine, vf = very fine; and sbk = subangular blocky. Roots: c = common; f = few; and m = many. Boundary: a = abrupt; c = clear; w = wavy; and i = irregular.

Table 7. Average morphological properties* of 2 unreclaimed, Lower Kittanning coal minesoils (RB & BAK).

Hor	Depth	Matix colors	Mottles	Struc	Grav	Cob	Roots	Bndy	Tex
RB									
Top	0-12	10YR 4/2, d grayish brown 10YR 5/4,	cfd 2.5Y 2/0, black mmf 10YR 5/4, yellowish brown	m	37	--	cf	a,s	CL
Mid	19-53	2.5Y 6/2, l brownish gray 10YR 4/2, d grayish brown 10YR 6/4, l yellowish brown	cfd 2.5Y 2/0, black mmp 7.5YR 7/8, reddish yellow mmf 10YR 5/4, yellowish brown	m	50	--	ff	c,s	L
Bot	53-95	2.5Y 6/2, l brownish gray 10YR 4/2, d grayish brown 10YR 5/6, yellowish brown	cfd 2.5Y 2/0, black cfp 2.5Y 6/2, l brownish gray mmp 7.5YR 7/8, reddish yellow	m	45	--	--	--	L
BAK									
Top	0-8	10YR 3/1, v d gray 10YR 5/3, brown	mfd 2.5Y 2/0, black cmd 10YR 5/6, yellowish brown	m	40	--	ff	a,w	L
Mid	38-70	2.5Y 2/0, black 10YR 4/1, d gray	mmf 2.5Y 2/0, black cmd 10YR 6/2, l brownish gray	m	53	--	ff	c,w to i	L
Bot	70-100	2.5Y 2/0, black 10YR 5/2, grayish brown	cfd 2.5Y 2/0, black cmd 7.5YR 6/8, reddish yellow	m	25	--	ff	--	L

*Codes are as follows: Hor = Horizon; Struc = Structure; Grav = Gravels %(v/v); Cob = Cobbles %(v/v); Bndy = Boundary; and Tex = USDA Texture. Colors: d = dark; v = very; l = light; and s = strong. Mottles abundance: c = common; f = few; and m = many. Mottles sizes: f = fine; m = medium. Mottles distinctness: f = faint; d = distinct; and p = prominent. Structure: 1 = weak, m = massive; f = fine, vf = very fine; and sbk = subangular blocky. Roots: c = common; f = few; and m = many. Boundary: a = abrupt; c = clear; w = wavy; and i = irregular.