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ACID SULPHATE SOILS

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ACID SULPHATE SOILS

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A typical example of a cat clay profile

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1. INTRODUCTION

During recent decades there has been increasing reclamation of coastal districts. Although there have been many very successful reclamation projects, there were others which gave some very unpleasant surprises. A good harvest was usually obtained during the first year of reclamation or drainage, but very soon afterwards the reclaimed area had to be wholly or partly abandoned because few if any crops could be produced from it. Further investigation showed that the soil which had first shown a basic to weakly acid reaction had very soon become extremely acid ($pH < 4$).

This rapid and extreme acidification of certain soils was first noted and studied in Holland (VAN BEMMELEN, 1886). The very low pH was due to very acidic iron- and aluminium sulphates and free sulphuric acid formed by the oxidation of iron sulphides. In Holland soils of this type are known as "cat clay", a name-associated with cat's excrement which has become a general term of abuse for all kinds of poor soils.

It was subsequently found that the occurrence of such special soils is not connected with a temperate climate and that they may be expected to occur in similar areas in any part of the world. So far as it has been possible to discover, these acid sulphate soils have hitherto been encountered in the following countries in addition to Holland:

AFRICA

Central Africa	(Chenery, 1954)
Nigeria	(Doyne, 1937)
Sierra Leone	(Doyne and Glanville, 1933; Dent, 1947; Tomlinson, 1957)

AMERICA

U.S.A.	(Edelman and van Staveren, 1958)
Surinam	(Muller, 1950)

ASIA

India	(Subramonyan, 1952)
Indochina	(Coyaud, 1950)
Indonesia	(van Wijk, 1938, 1951)
Malaya	(Dennet, 1933; Wilshaw, 1940; Coulter, 1952)
Pakistan (East-)	(Quadir, 1952; van Beers, 1955)
Thailand	(Pendleton, 1947)

EUROPE

Finland	(Aarnio, 1930, 1932; Kivinen, 1949, 1950)
Sweden	(Wiklander, et al, 1950)

The principal processes which may lead to the formation of acid sulphate soils may be summarised as follows:-

1. *In marine and brackish water sediments sulphur may be bound and accumulate under certain conditions in the form of iron sulphides and elementary sulphur as a result of the reduction of sulphates. The latter are chiefly derived from sea water;*
2. *When an area of this kind is drained the soil is aerated. The iron sulphides are consequently oxidised, sulphuric acid being formed together with Fe and Al sulphates which have a highly acid reaction;*
3. a. *When sufficient calcium carbonate is available the sulphuric acid released is neutralised to gypsum ($CaSO_4 . 2\ H_2O$); the adsorbed Na and Mg are moreover largely replaced by Ca and the soil remains in good condition. Hence in this case the presence of Fe sulphides has a favourable effect.*

b. If the amount of bases present is small compared to the sulphate present, there is extensive acidification of the soil, often resulting in a pH lower than 4, *usually in the region of* 3, *and in extreme cases even lower than* 2. *The ultimate pH depends on the sulphur/base ratio.*

In view of the fact that the reclamation or drainage and aeration of non-calcareous soils containing iron sulphides may involve great difficulties and considerable loss of time and money, the following questions arise:-

- Where are these hazardous soils to be expected?
- How can they be identified in the field and/or laboratory:
  a. in an unoxidised state, viz. when the potential acid sulphate soils are concerned?
  b. after reclamation or drainage, viz. in an oxidised state?
- If a clay has been identified as a potential sulphate soil, how can the extreme acidification of the soil be prevented?
- If the existence of an acid sulphate soil is found after reclamation, how can this soil then be restored to a good state?

It is not possible to provide a complete or entirely satisfactory reply to these questions. The research work undertaken to date is still too inadequate for this purpose, especially as regards the theoretical foundations. This is chiefly owing to the fact that it is not only chemical processes that occur but also very many microbiological ones, and moreover these processes are both extremely diversified and complex. What is more, the iron and sulphur may occur in a large number of different forms, with corresponding differences in the chemical and microbiological reactions.

It is the aim of the present article to give a survey of the chief theoretical and practical data which may be deemed important for providing the best possible reply to the abovementioned four questions and also for any further research.

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2. REGIONAL DISTRIBUTION OF ACID SULPHATE SOILS

2.1. RELATIVE AMOUNTS OF CALCIUM AND SULPHUR IN SEDIMENTS

An understanding of the regional distribution is intimately bound up with a knowledge of the conditions in which iron sulphides and calcium carbonate can be fixed and accumulate.

Both the amounts of fixed iron sulphides and accumulated calcium carbonate may vary considerably from one sediment to another. Consequently the presence or absence of cat clay entirely depends on the amount of each of these two materials in the sediment.

With respect to the occurrence of calcium and sulphur, four main combinations of much (+) and little or no (–) lime or sulphur may be distinguished:-

Ca +	Ca +	Ca –	Ca –
S –	S +	S +	S –
(1)	(2)	(3)	(4)

In cases (1) and (4) we are mainly concerned with river sediments respectively rich and deficient in lime since these invariably have a low sulphur content.

Cases (2) and (3) are more typical of marine or brackish water sediments. In case (2), which is characteristic of many Dutch soils, there is a possibility of cat clay occurring locally since local factors may still cause lime-deficient patches in a sediment which is mainly rich in lime.

On the other hand, in case (3), which frequently occurs in rainy and warm regions there is a very good chance that cat clay will form over a fairly large area after drainage.

We will now discuss in turn the conditions under which the iron sulphides and calcium carbonate are respectively fixed in the sediments.

2.2. CONDITIONS GOVERNING THE FORMATION AND ACCUMULATION OF IRON SULPHIDES

The formation of iron sulphides is a very frequent phenomenon in nature. All marine deposits contain iron sulphides, and in non-marine deposits also decaying organic material may cause H_2S formation, resulting in black coloration by FeS.

Generally speaking, however, the amount of sulphides formed in *non-marine* sediments is comparatively slight (0.1–0.3 grams of sulphur per 100 grams of dry soil). Hence the free or adsorbed bases present are usually sufficient to prevent extreme acidification of the soil after any oxidation of the sulphides.

In *marine* sediments, and particularly in brackish water sediments, the amounts of fixed sulphur are usually much greater, viz. of the order of several per cent.

The main factors that determine the possibility of the formation and accumulation of iron sulphides are as follows:-

a. *a supply of sulphur*;
b. *a highly reducing environment or the presence of* what, compared to the amount of oxygen present, is *a large quantity of readily oxidisable organic material;*
c. *the presence of mobile iron compounds;*
d. *consolidation of the iron sulphides* once they have been formed, *or accumulation caused by a "permanent" reducing environment.*

Re a: *The supply of sulphur*

Although H_2S is formed by the decay of organic material and this results in the formation of dark-coloured iron sulphide (FeS), this source of sulphur is insufficient to give the soil a high content of iron sulphides. Organic material usually contains 0,5–1 % of sulphur, much of which occurs in a difficultly oxidisable compound. The various fluviatile, marine and brackish water sediments generally only have a relatively low content of organic material (2 to 10 %), so that the decomposition of organic material cannot be expected to give more than a few tenths per cent of iron sulphide content in the soil.

Hence high contents of sulphur compounds can only occur when sulphur is supplied from another source, the chief being sea water. This contains about 2 grams of sulphate per litre, based on SO_4. It may be inferred from this that in a marine sediment saturated with sea water and containing, say, 80 g of water per 100 g of dry soil, there is approximately 0.16 % of sulphate (SO_4). When it is found that, as stated earlier, marine sediments often contain a few per cent of sulphur compounds, this can only be explained by assuming a process in which there is continuous accumulation of these sulphides. It is an equilibrium reaction in which the insoluble product is repeatedly fixed and the supply of basic material (SO_4) is constantly renewed. A sediment containing a few per cent of sulphur compounds is therefore certain to be repeatedly impregnated with sea water. In most cases this is only possible when there is slow sedimentation or re-transport of the sediment, although diffusion and/or infiltration may also play a part.

Re b: *A reducing environment and organic material*

The reduction of sulphate is a biochemical process. The micro-organisms derive their energy from the oxidation of organic material. The amount of oxygen required for this purpose is taken from the sulphates in an anærobic environment by the reduction of sulphates to sulphides. There are no indications that the bacteria required for the sulphate reduction might constitute a limiting factor.

The reduction process is extremely complex and passes through numerous intermediate stages. If we ignore these intermediate stages and represent the organic material in the soil (the source of energy for the reduction) by CH_2O, then the conversion formula is as follows (ZUUR, 1954):-

$$4Fe(OH)_3 + 4\,CaSO_4 + 9\,CH_2O \rightarrow 4\,FeS + Ca(HCO_3)_2 + CO_2 + 11\,H_2O;$$

Since FeS, unlike $CaSO_4$, is an insoluble material, sulphur is fixed in the soil by this reduction.

If a new sulphate is supplied, the reduction and fixation can continue. Simultaneously with the supply of new sulphate there is also a removal of the soluble Ca-, Mg-, or Na-bicarbonate formed in the reaction.

In an ærobic environment there can be no sulphate reduction, but in a sediment saturated with water an ærobic environment can change fairly rapidly into an anærobic one. This happens, for instance, when there is a large amount, compared to the oxygen present, of readily oxidisable, fresh organic material. The oxygen present is then rapidly consumed by the ærobic micro-organisms for the decomposition of the organic material. A distinctly ærobic environment will require more organic material than a moderately ærobic one in order to make the environment anærobic. From the analysis of a large number of soil samples PONS (1959) concluded that within a given sedimentation area there is a fixed ratio between the amount of sulphur compounds and the amount of organic material in the sediment. Moreover the amount of this S/organic matter ratio chiefly depended on the oxygen supply of the environment. The extent to which this environment was anærobic or ærobic could be deduced by examining the sediment for foraminifera and diatoma.

The variations found are summarised in the following table:-

Sediment	m.e. SO_4 per 100 g dry soil	Environment during sedimentation	Percentage of organic material
North Sea	7	highly ærobic	2–6
Zuiderzee	14	ærobic	1–3
Beemster	20	moderately ærobic	4–7
Wieringermeer	25	moderately anærobic	4–8
Watergraafsmeer	40	anærobic	3–6

The above sediments are all under-water sediments in a saline or fairly saline environment and without plant growth.

In the presence of plant growth, especially reed (Phragmites) and mangrove (Rhizophora racemosa, TOMLINSON, 1957), the amount of organic material present is no longer a limiting factor. In such a case the extent to which the sedimentation environment is ærobic or anærobic has little effect on the amount of sulphur that may be fixed, since the vegetation provides large amounts of fresh organic material, resulting fairly rapidly in the formation of an anærobic environment. In this case, however, the supply of sulphates may be inadequate.

This is because reeds and Rhizophora racemosa can only tolerate a low salt content. Very large amounts of sulphur can only be fixed when there is very slow sedimentation, enabling repeated supplies of sulphur to be brought up. Consequently the highest sulphur contents are found in clayey phragmitic peat. When the supply of sulphur is a limiting factor, as may be the case when there is fairly rapid sedimentation and a low SO_4 content of the water, there will be no further relationship between the amount of sulphur and organic material present.

Re c: *The availability of sufficient iron*

When there is only a small amount of organic material available, an oxidising environment prevails, and moreover the environment has a high pH, the amount of soluble iron required for the formation of iron sulphides may be a limiting factor. This is the case, for instance, in the more oceanic parts of marine deposits (EMERY 1952).

There can only be a considerable enrichment of sulphides in a highly anærobic environment. Iron is readily soluble in this environment as it occurs here in the ferro form. Since Fe usually occurs in large amounts in the soil the amount of iron required can only rarely be a limiting factor in the formation of potential acid sulphate soils.

Re d: *The accumulation of iron sulphides*

In addition to the possibilities of formation of iron sulphides consideration should be given to the accumulation of these sulphides.

The FeS originally formed undergoes a fairly rapid change into the more stable polysulphides, chiefly pyrite (FeS_2). So long as the sediment is not aerated, i.e. it remains below water or is insufficiently drained, no sulphur is removed.

On high tidal flats grown over with vegetation which are regularly flooded only by spring tides there is in fact some constant fixation of sulphur, but there is also simultaneous oxidation or removal. Consequently these elevated marine formations always have a fairly low sulphur content. The same is true of the parts of tropical coastal regions in which the Avicennia nitida is the chief type of vegetation (TOMLINSON, 1957; SAVORY, 1953), or in general of marine or brackish water sediments which are only flooded for a part of the year and are dry during the remainder. These marine sediments are only inundated during spring floods, whereas the area becomes dry during the normal tides. The brackish water sediments are flooded during the rainy season, but during the dry season a considerable portion of the area is dried out again.

Since, as was stated above, there is no substantial accumulation of sulphur in these sediments we need not fear that acid sulphate soils will be formed in these cases.

This does not mean, however, that no unfavourable soil conditions need be anticipated in this instance. When these sediments are entirely without free calcium, as often happens in brackish water sediments with a vegetation cover, the so-called Mg-clays are formed which, compared to the exchangeable calcium, have a high content of exchangeable magnesium. These Mg-clays have the same poor characteristics as the Na-clays, viz. a very unstable and unfavourable structure.

These unfavourable brackish water sediments occur more often than is generally recognised, for the simple reason that in the investigation of the soils of coastal areas it often happens that too little attention is paid to the exchangeable magnesium.

When finally we compare marine and fluviatile sediments for the above-mentioned conditions which favourably or unfavourably influence the fixation and accumulation of iron sulphides, we can distinguish three main cases.

It can be seen that in *marine* and *estuarine* sediments near the coast there is usually a strong tidal effect. This gives the water a high oxygen content and also results in fairly rapid sedimentation. Owing to the ærobic sedimentation the amount of organic material will have to be relatively large to allow substantial sulphur fixation as well. But the rapid sedimentation prevents such considerable accumulation of organic material, and consequently conditions that might result in the fixation of a large amount of sulphur are absent in this case.

On the other hand, the reduction possibilities are usually very good in *fluviatile* sediments, especially in the lower parts of the terrain (ZONNEVELDT, 1957). In this case, however, the sulphate supply is a very limiting factor, the sulphate content of river water generally being low.

The transition area between these two types of sediments, the *brackish water area*, generally provides the most favourable conditions for the accumulation of iron sulphides, particularly the parts of the brackish water area that are still regularly inundated by the saline flood water mixed with river water but where the drainage is naturally poor and permanently waterlogged conditions prevail.

2.3. THE PRESENCE OF CALCIUM CARBONATE IN SEDIMENTS

There are wide fluctuations in the primary $CaCO_3$ content of marine and other sediments; calcium-free sediments occur in addition to sediments very rich in calcium. In Holland most marine sediments contain from 10 to 20% of $CaCO_3$, while the river sediments (Maas, Rhine) contain from 6 to 15%.

In marine sediments a part of the $CaCO_3$ may be derived from organisms. On the other hand, in the coastal sediments the chemically precipitated calcium carbonate usually predominates.

For marine sediments in particular it is in many cases difficult to explain why one should be rich in calcium and another not, but the fact is very easy to establish. For this reason we shall content ourselves with a few general observations on the occurrence of calcium carbonate in sediments.

Of the processes that play a part in the formation of $CaCO_3$ carbon dioxide tension is the most important. In an environment of high carbon dioxide tension the precipitated $CaCO_3$ goes into solution with formation of $Ca(HCO_3)_2$. This environment produces calcium-free sediments, despite the high calcium content of the water (PONS, 1957). The carbon dioxide tension is influenced by decay of organic material (CO_2 production), stagnant water (any CO_2 formed is unable to escape), temperature (CO_2 has a greater solubility at lower temperatures), the respiration of and assimilation by animals and plants (respectively producing and consuming the CO_2 present).

In water containing many organic acids (fen water) or river water from a pedologically old area (numerous tropical rivers) the calcium is not chemically precipitated but occurs in the dissolved form, or they contain only a very small proportion of calcium, so that calcium-free sediments are formed in this case as well. Acidic river water may so affect the sea water along coasts that the calcium carbonate in the sediments goes into solution and only calcium-free sediments are formed.

In a sedimentation area the conditions may be such that calcium is deposited at one place but not at another. PONS and WIGGERS (1959) found that in a given period in each sedimentation area the calcium content per % of clay is characteristic and is subject to wide fluctuations, depending on the sedimentation conditions.

2.4. CONCLUSION

To sum up, it can be stated that both the sulphur and calcium content of a sediment may vary considerably, depending on conditions. High sulphur contents principally occur in saline and brackish sedimentation areas in which the water movements are gentle. Low calcium carbonate contents are due to the fact that the sedimentation environment is acidic or free of calcium, or because the originally calcium-rich sediment is decalcified during sedimentation. The same conditions that promote sulphur accumulation usually prevent the accumulation of calcium, so that there is a heightened possibility of acid sulphate soils occurring.

From the geomorphological standpoint it can be stated that this unfavourable combination of soil conditions can be expected in all parts of the world in marine and brackish water sediments containing no calcium, and especially in the lower reaches of the brackish water zone (at the end of the creeks) of a coastal area, which is sinking relative to the sea and when there is slow sedimentation. In areas other than coastal ones account should be taken of the possibility of sulphate being supplied by lateral infiltration of sea water or of water from old marine sediments.

The soil of such areas chiefly consists of heavy clays or peaty clays. The subsoil is usually very soft (soapy clays). On the borders of such areas there is often peat formation.

When the conditions in the coastal areas are such that there is no accumulation of sulphides there may still be unfavourable soil conditions. This is the case, for instance, when the sediments are entirely free of calciumcarbonate. The so-called magnesium clays are then formed which have the same poor characteristics as the sodium clays. These entirely calcium-free sediments are chiefly to be expected in old freshwater sediments that are occasionally flooded with sea water, or in brackish water sediments which have a hinterland mainly consisting of acid soil formations.

3. SULPHUR COMPOUNDS IN THE SOIL

Depending on the form in which the sulphur occurs in the soil, and the course, stage and final result of the oxidation process, the sulphur-rich soils may differ in appearance in the field, or behave very differently when examined at the laboratory.

3.1. Chemical and mineralogical characteristics

In the reduction of sulphates by micro-organisms in the absence of oxygen and the presence of readily oxidisable organic material the first product is usually hydrogen sulphide (H_2S). This reacts with the iron present, resulting in hydrotroilite ($FeS.nH_2O$). This may pass over into melnikovit ($FeS_2.nH_2O$; DORS, 1912), a cryptocrystalline form of pyrite (FeS_2). Both sulphides are black and give the soil a black tinge even when only present in minor amounts. When the sulphides are exposed to the air the black colour rapidly changes to grey owing to oxidation of FeS. In this connection it should be noted that certain soils may exhibit the opposite phenomenon, viz. a light-coloured soil rapidly turns black as a result of aeration; in this case there is oxidation of the humic acids present.

Hydrotroilite dissolves in dilute hydrochloric acid with formation of H_2S. Melnikovit dissolves in dilute hydrochloric acid when heat is applied, and in concentrated hydrochloric acid at normal temperature. When it is boiled in water there is a hydrolytic splitting into FeS and S, the FeS formed being converted into iron hydroxide.

Occasionally the H_2S is present in the free form in wet soils. The free H_2S can be removed by boiling or by oxidation with iodine. This oxidation produces a precipitate of free sulphur. H_2S may also be formed by oxidation of hydrotroilite. In soils containing many sulphur compounds a distinct odour of H_2S is often noticed when making a borehole or soil pit. All dark-coloured soils which in the field, or after treatment of a sample with hydrochloric acid, give off an H_2S odour and have a low calcium content are to be regarded as suspect and should therefore be further examined in the laboratory.

However not all sulphur-containing soils are dark-coloured. Grey colours predominate when the sulphides are chiefly present in the form of pyrite (cubical FeS_2) or marcasite (rhombic FeS_2). Other polysulphides (Fe_nS_{n+1}) may also be present. Pyrite is formed in a neutral or alkaline environment, whereas marcasite is chiefly formed in an acidic environment (ALLEN, 1912, EMERY, 1952). Both are yellow, metallic opal minerals, practically insoluble in concentrated hydrochloric acid, but soluble in nitric acid in which pyrite dissolves entirely, whereas marcasite deposits sulphur (ALLEN 1952; DOELTER, 1912). It is a reasonable assumption that in brackish water formations the polysulphides chiefly occur in the form of pyrite. The presence of other polysulphides was also assumed as a result of chemical analyses (VAN DER SPEK, 1934, HARMSEN, 1954). Eventually the $FeS.nH_2O$ originally formed in most cases wholly or partly passes over into pyrite, presumably in accordance with the reaction $FeS + S \rightarrow FeS_2$. Anyhow, pyrite can be produced in this way in the laboratory (FELD, 1911, VERHOOP, 1940, ALLEN, 1912). The period required for complete transformation under natural conditions can be estimated as varying from 50 years to several centuries (VAN STRAATEN, 1954). On the other hand pyrite has also been found in fresh mud (VAN STRAATEN, 1954, HARMSEN, 1954, ZUUR, 1936), although it is difficult to decide to what extent it is autochthonous or derived from older sediments.

As regards the form in which pyrite occurs, it can be stated that it is usually found as globular crystal aggregates having a diameter of from 5 to 40 microns and occurring alongside the remains of plant roots, in plant fibres, and in the valves of diatoms, etc. (VAN BEMMELEN, 1886, VAN DER SPEK, 1954, VAN STRAATEN, 1954). The presence of pyrite was established both by chemical means and X-rays. Distinctly cubic crystals were also found. Presumably pyrite is very difficult to oxidise in the latter form. This may be concluded from the fact that a considerable percentage of pyrite was still found in certain soils which have long been brought into cultivation (ZUUR, 1936), and from the fact that numerous pyrite crystals may occur in severely weathered tropical soils derived from marine sediments (VAN BEERS, 1949). Hence for fundamental research microscopic and mineralogical examination will be desirable in addition to chemical analyses.

It follows from what has been stated above that soils which have no dark coloration and give no reaction with HCl as evidence of the presence of FeS may still be potentially highly acidic when iron sulphides occur in the form of pyrite.

3.2. OXIDATION PROCESSES

Oxidation of sulphur compounds begins when the anærobic conditions are terminated; this may occur either under natural conditions or as the result of artificial drainage.

Regarding the course of the oxidation there is a paucity of reliable information. This is not surprising when we consider the variety of starting products, oxidation conditions, and especially the fact that both many microbiological and chemical processes occur. Very probably there are substantial differences both as regards the oxidation products and the rate of the oxidation process, depending on whether the iron sulphides chiefly occur in the form of FeS_2 or FeS (VAN DER SPEK, 1950, MAC INTYRE, 1930). When FeS_2 predominates, a distinctly yellowish basic ferrisulphate is usually formed in soils without $CaCO_3$ and oxidation proceeds fairly rapidly and is accompanied by a rapid decrease in the pH. On the other hand, should FeS predominate no basic ferrisulphate is formed but sulphuric acid and sulphate instead.

The *oxidation of pyrite* has been investigated in detail and described by VAN DER SPEK (1934, 1950). $FeSO_4$ and H_2SO_4 are formed in the first instance. Should an excess of H_2SO_4 be left which is not rapidly drawn off or neutralised by the bases present, further oxidation results in ferrisulphate $Fe_2 (SO_4)_3$, (also written $Fe_2O_3.3\ SO_3$). The ferrisulphate hydrolises very readily and this leads to the formation of basic ferrisulphate and sulphuric acid, e.g.:-

$$Fe_2(SO_4)_3 + 2\ H_2O \rightarrow H_2SO_4 + Fe_2(SO_4)_2\ (OH)_2 \text{ or: } (Fe_2O_3.\ 2\ SO_3.\ H_2O).$$

The name basic "ferrisulphate" given to this salt with a highly acid reaction is connected with the presence of OH groups.

The further hydrolysis of the ferrisulphate or splitting- off H_2SO_4 will depend on whether the sulphuric acid formed in the hydrolysis disappears as the result of eluviation, its reaction with the bases, or its reaction with the iron and aluminium of the clay material. In their investigation of the Fe_2O_3 – SO_3 – H_2O system POSNAK and MERWIN (1922) found that the compound 3 Fe_2O_3.4 SO_3. H_2O may be regarded as the end-product of the hydrolysis of ferrisulphate (1Fe_2O_3.3 SO_3). This gives a neutral reaction and is occasionally encountered in this form (PENDERS, 1937, VAN DER SPEK, 1950).

In most cases, however, the hydrolysis or splitting-off of sulphuric acid is incomplete, so that the basic ferrisulphate usually gives a very acid reaction. It has a distinct lemon-yellow colour. Occasionally it is blended with aluminium sulphate, and in this case the colour is more whitish-yellow. It is insoluble in water but soluble in 10% hydrochloric acid.

When the conditions for oxidation and removal of sulphuric acid are very favourable, e.g. in old root passages, no ferrisulphate is formed, but instead iron hydroxide in the form of iron tubes (VAN DER SPEK, 1950). When there is poor drainage, e.g. in the lowest parts of a terrain, a high water table and a pronounced dry season, the sulphuric acid reacts with the clay materials in particular and a concentration of salt is formed on the surface, much of which consists of aluminium sulphate (Borneo, Pakistan, India, Thailand, Indo-China, Finland). Occasionally the aluminium sulphate is encountered in a form in which it is well crystallised out (EDELMAN, 1938). In other cases only a concentration of coarse granular iron and manganese deposits are found in the topsoil in such places These parts of the terrain are characterised by very little or no vegetation and can readily be identified as such on aerial photographs.

As regards the mobility of the aluminium and iron compounds, it can be stated as a general rule that ferri-compounds precipitate at a pH higher than 3, aluminium compounds at a pH higher than 4, and ferrosulphate at a pH higher than 6.

When an area consisting of soils with a high content of iron sulphides is drained a large amount of brown or white precipitate of iron or aluminium hydroxide is sometimes encountered in the ditches and many calcium and/or magnesium sulphates occur in solution in the ditchwater. The yellow precipitate of basic iron sulphate soon appears on the walls of the ditches. In many cases this can also be observed on the excavated soil, e.g. either that removed from the ditches or used for making dykes. Moreover drainpipes often become clogged up in a very short while with a soft mass of iron and aluminium hydroxides (VAN DER SPEK, 1950, ZUUR, 1936) and have to be repeatedly cleaned out.

The *oxidation of* $FeS.nH_2O$ proceeds in two main steps, viz. $FeS \rightarrow S$ and $S \rightarrow H_2SO_4$. The former is a purely chemical process and very rapid; the black colour of the soil very soon turns to grey, yellowish ferrihydroxide and elementary sulphur being formed.

The oxidation of sulphur to sulphuric acid is a much more gradual process, i.e. under natural conditions it is a matter of weeks or months. It may be a purely chemical process, although it may be accelerated by bacterial processes. In a soil containing no calcium carbonate the sulphuric acid formed will in the first instance remove bases from the soil, thereby rendering it more acidic. If there are not many bases present the sulphuric acid will react with the iron and aluminium in the soil and convert it into acid-reacting ferrosulphate and aluminium sulphate (VAN DER SPEK, 1950). Hence oxidation of FeS will not produce any yellow, basic ferrisulphate, as is often the case in the oxidation of FeS_2. Presumably the research carried out in Sweden and Finland chiefly relates to soils containing FeS, since the presence of yellow ferrisulphate is not reported from these areas. The oxidation of sulphur to sulphate is accelerated by the addition of minor amounts of calcium; one to two tons per acre was found to be sufficient (MAC INTYRE, 1930, WIKLANDER, 1950). On the other hand the oxidation of pyrite proceeds most rapidly in an acid environment (HARMSEN, 1954, HART, 1959). The fact that pyrite is only soluble in nitric acid (HNO_3) points in the same direction since the HNO_3 has both an acidic and oxidising effect.

4. THE IDENTIFICATION OF ACTUAL AND POTENTIAL ACID SULPHATE SOILS

The identification in the field of actual acid sulphate soils, viz. in their oxidised state, does not give any difficulties. It may be done by showing the presence of the distinctly yellowish basic ferrisulphate or white aluminium compounds in the soil profile or in excavated soil, or by a field determination of the pH and by a field examination of the ditchwater and/or soil water for the presence of a high sulphate content.

The identification of a potential acid sulphate soil, viz. one in which the soil is in a non-aerated (non-drained) state, is chiefly a matter of laboratory examination and takes more time and trouble.

The possibility of extreme acidification of the soil can only be accurately determined by ascertaining the total content of sulphur and calcium. The advantage of this method of determination is that it also provides information on the degree of acidification of the soil to be expected after oxidation. If the amount of sulphur present is expressed as m.e.¹) of SO_4 and the free carbonates and exchangeable Ca + Mg present as m.e. of Ca, it can usually be expected that the pH will not fall below 5.0 when compared to the m.e. of SO_4 there is an excess of 50 m.e. of Ca per 100 grams of soil (BENNEMA, 1953, PONS, 1956). For the method of determining the total content of sulphur compounds reference may be made to LITTLE (1951) and SMITTENBERG et al. (1951).

The simpler qualitative determinations amount to the initiation and promotion of the oxidation process with subsequent pH or sulphate determinations. This oxidation may

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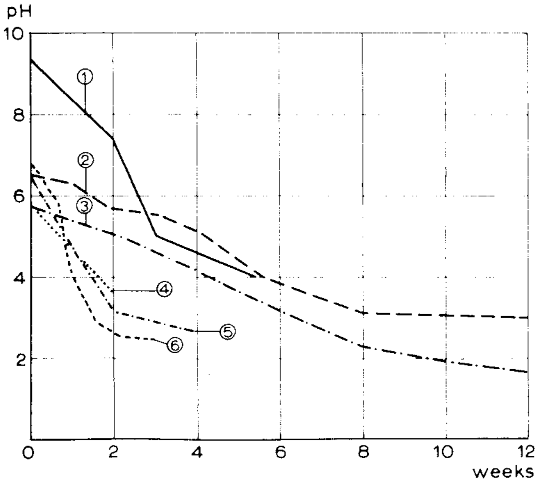
¹) milli-equivalent

be carried out in various ways, viz. exposing soil samples to the weather for a given period, which amounts to alternate drying and wetting (Dent, 1947, Kivinen, 1949, Pons, 1949), storing in a moist state for some weeks (Doyne, 1937), drying in air (Subramanyan, 1952, Muller, 1950), or drying on a glass plate or in a drying stove at 70° to 80 °C (van der Spek, 1934). Determinations of the pH, sulphate content (soluble in water or total sulphate), or electrical conductivity can then be made at regular intervals. As a rule no yellow ferrisulphate is formed on drying. It is only when a large clod of soil is dried intact that a distinct yellow colour may be observed in places.

The following figure shows the trend of the pH as found by various workers under varying oxidation conditions.



1. Dent (1947): exposed to the weather
2. Pons (1955): storage in moist stage
3. Doyne (1937): storage in moist stage
4. Doyne (1937): air dried after 2 weeks storage in moist stage
5. Subramanyan (1952): air drying
6. Tomlinson (1955): air drying

Compared to storage in a moist state, the pH falls far more rapidly with drying. But the data provided by Kivinen, 1949, show that under certain conditions the pH may fall very gradually; sampling in spring gave a pH of 8, after two months of exposure to air and rain it was 7.5, and at the end of the summer 4. In the following year it was 2.5. Here there was no question of a heavy clay and presumably the iron sulphides chiefly occurred in the form of FeS. Meanwhile the electrolyte content had risen to ten times the original figure.

Temperature is also an important factor both as regards biological and chemical processes. The optimum for the microbiological processes is probably in the region of 30 °C;

for the chemical processes the optimum temperature is much higher. When soil is dried in a drying stove at 70° to 80°C oxidation is a purely chemical process. This method of drying gives both a more rapid and greater decrease in the pH than slow drying on a glass plate (VAN DER SPEK, 1934). HARMSEN (1946) also found a rapid and considerable decrease in the pH when pyrite powder was oxidised by purely chemical means at a temperature of 60°.

From what has been said above it would appear that the identification of a potential sulphate soil by these methods is a difficult and time-consuming procedure. Hence these methods are unsatisfactory insofar as it is desirable to be able to identify these dangerous soil types even during a soil survey, enabling the soil cartographer to define the limits of this soil type. *What is therefore required is a rapid and simple field method.* In this connection the author has carried out supplementary research which has led to the development of a field method which appears to answer the purpose. Systematic supplementary research is still required, however, so that this field method is only mentioned here to enable it to be tested further and compared with other more time-consuming methods such as the periodical pH determinations and the determination of total S and Ca.

In *principle* this method is as follows: a small amount of soil is treated with concentrated hydrogen peroxide with careful heating over a water bath. When all the H_2O_2 is spent the pH is determined. In the case of the dangerous sulphate soil the pH will fall to 3 or 2 after this treatment.

As regards the *procedure* to be adopted in this method the following remarks may be made. By "small" amount of soil is meant an amount of about 5 c.c. If more soil than this is taken proportionately more H_2O_2 is required. About 20 c.c. of 30% H_2O_2 is added to this soil in a small porcelain dish (diameter 8–10 cm). Soil and H_2O_2 are mixed as well as possible by stirring. The dish is then placed on a beaker of boiling water. The oxidation is often very explosive so that it is advisable to have a siphon at hand or to stir continually and if necessary to remove the dish from the water bath in good time. When all the H_2O_2 is spent the pH is determined. The pH thus calculated may be regarded as the approximate end-value of the acidity of the soil if it were to be subjected to extensive oxidation (drainage).

The pH-meter used should be suitable for determining a pH < 4. The Hellige Truog Soil Tester is not so very suitable for this purpose although it can be used. At a pH of 4 a deep yellow colour is obtained, at a pH of 3 a light yellow colour, and at a pH of 2 an orange-yellow colour, while with a still lower pH a red colour appears. With the dangerous sulphate soils the indicator changes to the orange colour, which often turns to red after the soil paste has dried. Even without pH determination it is possible to obtain an indication as to whether the soil has become extremely acidic, as in such a case the mud left in the dish after the H_2O_2 treatment has a pronounced and typical green colour.

As regards the heating during the oxidation it may also be remarked that heating is not always necessary since there are certain catalytic effects which may cause the oxidation to proceed very rapidly, accompanied by such considerable heat evolution that the dish containing the soil, etc., can no longer be held in the hand. When working in a particular area it is advisable to first apply the test to a number of samples of the same type both with and without heating. When it is found that the same final result is obtained the other samples from this area can be examined without heating, thereby considerably simplifying the method.

The normal H_2O_2 commercially obtainable is always slightly acidified in order to increase its storage stability. Pure H_2O_2 is difficult to keep and the bottle should always be sealed with paraffin wax. Although a drop of the normal H_2O_2 used in the laboratory causes the indicator to change to yellow immediately, this acidification of the H_2O_2 is no drawback, the small amount of acid present being readily buffered by the soil. Consequently experiments made with pure H_2O_2 and normal commercial H_2O_2 gave the same result. The question now arises: how much H_2O_2 should be used? It is difficult to answer this, especially as the soils differ in their contents of organic material and sulphur compounds. The pH is determined when all the H_2O_2 is spent; 20 c.c. will probably be sufficient in most cases.

Finally it should be noticed that the pH of soils containing many sulphides but which are also rich in calcium shows very little or no decrease when they are treated with H_2O_2. Hence such soils are not dangerous. Only gypsum is formed during their reclamation or drainage and oxidation, and this does not give any difficulty.

5. RECLAMATION OF ACID SULPHATE SOILS

To what extent will soils that have been identified as *potential* acid sulphate soils be suitable for use as agricultural land?

Generally speaking, the reply to this question must be that in such cases one should proceed very cautiously. Often it will not be economically justified to invest large sums of money in such land (for drainage, road construction, irrigation, etc.) as there is a risk that it will only have a limited yield capacity for a considerable time, and in extreme cases may even fail to produce any harvest at all.

In doubtful cases it is advisable to subject the most dangerous soils, as identified from the indications provided in chapter 4, to a small-scale practical test.

Although in theory the consequences of the acidification might be overcome by the use of liming, in most cases it will be found that the lime dressings have to be so very large that the cost would be prohibitive.

If for some reason or other such land has nevertheless to be reclaimed (e.g. when it forms part of a larger area) it would be wise only to drain it to a very shallow depth and the bad layers of soil should remain as far as possible under water (or the water table). This is the best possible way of preventing acidification of the soil by oxidation.

It will be obvious that this shallow drainage can only be carried out provided the bad layer is not close to the surface and the water table can be kept fairly high and constant. In the humid climate zone it is advisable to use such land as grassland. Under such limited conditions rice would be the best crop to grow in the tropics and sub-tropics. If the irrigation water employed has a high calcium content the irrigation will also have a preventive effect on the formation of acid sulphate soils.

Under the climatic conditions prevailing in Holland (an evenly distibuted rainfall with fairly low summer temperatures) a layer of acid clay at a depth of over 50 cm has little effect on the crop when the overlying soil has a good moisture retaining capacity and an adequate nutrient content. Hence the depth at which the unfavourable soil layer occurs is of prime importance.

Although in principle it is possible to reclaim soils after they have become acidic it is often difficult to do this and always expensive.

In the first place good drainage would have to be provided so that the soil can be aerated and the insoluble sulphides oxidised to soluble sulphates, enabling these sulphates to be drawn off. In such a case there will be a gradual recovery of the pH.

In areas with a pronounced dry season there should also be sufficiently deep drainage so as to prevent sulphates in the topsoil from being enriched by upward capillary movement. In some cases (presumably where the iron sulphides chiefly occur in the form of FeS) the sulphate formation can be promoted by adding small amounts of lime (5 tons per hectare) (WIKLANDER, 1950; KIVINEN, 1949). But in most cases in which pyrite predominates it will be advisable to defer using this lime until the soluble acid products have been largely leached out (HART, 1959). This lime does not then serve to neutralise the sulphuric acid formed but to correct the acidity of the soil. Large amounts of lime are often needed for this purpose, especially on soils with a large adsorption complex. For heavy clay soils a normal amount is 30 tons of $CaCO_3$ per hectare for a number of years (ZUUR, 1952).

When the oxidation is a slow process (as, for instance, in ricefields) the annual amounts required may be much smaller, e.g. 3 to 5 tons per hectare.

In some cases (e.g. in Holland) calcareous subsoil can be ploughed up; the ability to do this is determined by the cost factor.

The approximate amounts required can be determined in the laboratory. In this connection it should be noted that 20 m.e. of $CaCO_3$ are equivalent to about 40 tons kg. per hectare for a 30 cm layer.

In addition phosphatic fertiliser is nearly always required for bringing the soil into cultivation. Potash and nitrogen should also be considered in this connection.

It will be seen from the above remarks that this is another case in which it is true to say that prevention is better, and cheaper, than cure.

In brackish water formations, or on the border of fluviatile and marine formations, one should always take into account the possibility of potential acid sulphate soils being locally present. In such areas in particular, reclamation should be preceded by intensive soil studies.

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