

POZZOLANIC ACTIVITY AND MECHANISM OF REACTION OF RED TROPICAL SOIL-LIME SYSTEMS

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This paper presents a study on the pozzolanic properties and mechanism of reaction of nineteen red tropical soils from north east Brazil. The results of the study show that the mechanism of reaction between soil and lime is mainly a diffusion controlled process which can be expressed by the equation of Jander. Based on the constant rates of reaction obtained from this equation and on the quantity of reactive components of the soils an activity index A_j is proposed and its validity tested against the unconfined compressive strength of the lime-soil system. A valid statistical correlation is presented linking the A_j value with the empirical reactivity parameter of Thompson.

Approximately one third of the land surface of the earth is contained within the huge belt limited by the tropics of Cancer and Capricorn. Within this area "red tropical soils" are the dominant feature of the landscape, and whenever man decides to construct any facility, red tropical soil properties have to be determined and their behaviour predicted.

Geologists, soil scientists, agriculturalists and engineers are interested in different aspects of the behaviour of red tropical soils. This interest is reflected in the great volume of work devoted to classification and preparation of an acceptable nomenclature of red tropical soils. Unfortunately for a civil engineer these classifications are very confusing and of limited use. The terminology and classifications in actual use are so varied that an engineer is tempted to agree with F.C. Bawden who said "Classification is easy: it is something you just do" (1). Ideally, a classification should serve not only as a practical tool to transmit information but as a tool to predict behaviour when experience is not available.

Information regarding the engineering behaviour of red tropical soils is still scanty, this is reflected in the lack of classifications with engineering validity. An example of a classification for engineers is the one proposed by Lyons Associates in their study of African soils (2) and of South American soils (3). These classifications, the first related to d'Hooere's simplified classification and the other related to the FAO-UNESCO classification have had moderate success and can only be used as a

very rough guide to the properties of red tropical soils. Perhaps a great part of the useful data gathered on the behaviour of red tropical soils has never been fully used due to the confusion and controversy originated by the very nomenclature proposed by researchers in their desire to precisely define these materials. Terms like laterite, lateritic soil, latosol, ferricrete, ferruginous soil, ferrisol, oxisol, ferralsol, etc. have been used for the same type of material in some cases, while only one name has been applied to various types of materials in other cases. Therefore in this paper use is made of the term "red tropical soil" and this is broadly a soil which fits the definition of laterite given by Siverajasingham et al. (4), while the term laterite is restricted to the original definition given by Buchanan (5).

This paper deals specifically with the pozzolanic properties of red tropical soils, it attempts to explain the mechanism of the reaction between lime and the constituents of the soil and proposes an index of pozzolanic activity which is related to the strength of the soil by an empirical statistical relation. Finally, it suggests the activity index as a possible parameter for the classification of red tropical soils.

Lime-Soil Reaction Mechanisms

The different theories on the mechanism responsible for the alteration of some of the engineering properties of soils when mixed with lime in the presence of water have been well reviewed by Diamond and Kinter (6) and more recently by the Transport and Road Research Board U.S.A. (7). The changes that take place when lime is added to a soil are broadly divided into amelioration and/or strength development.

Among the mechanisms offered for the explanation of the amelioration effect of lime on soils are: flocculation due to cation exchange (pH dependent), physical adsorption, minor pozzolanic reactions between soil minerals and lime leading to diffuse cementation.

Flocculation due to cation exchange has been supported by the Iowa State University researchers. Hilt and Davidson (8) explained that amelioration occurred because lime increases the pH of the system thereby generating negatively charged sites at the

edges of the clay resulting in an increase in cation exchange capacity. The small amount of lime required for these changes to take place was termed "lime fixation point". Hilt and Davidson indicated that strength increases in the lime-soil system only occur when lime is added beyond this point. Cation exchange phenomena is real enough in many materials, it is a very useful property utilized in a great number of industrial processes, thus it cannot be discounted here. However, the predominant clay mineral in red tropical soils is of the 1:1 type, these clay minerals have low cation exchange capacities and low specific surface values. It is therefore questionable whether this mechanism alone can explain amelioration in red tropical soils.

The physical adsorption theory offered by Diamond and Kinter (6) postulates that lime molecules are adsorbed by clay surfaces and react with edges of other contiguous clay particles to form minute cementitious products.

The diffuse cementation theory proposed by Stocker (9) does not postulate adsorption of lime on the surfaces of the clay but that lime reacts directly with clay crystal edges, generating minute accumulations of cementitious products at or near the edges.

Strength increase is generally attributed to pozzolanic reactions, i.e. a process involving the production of cementitious compounds between lime and soil minerals. Fades and Grim (10) suggested that this reaction involves the dissolution of silica and/or alumina from the clay to react with lime to form the cementitious products while Stocker (9) attributes the mechanism to diffuse cementation - an in situ phenomenon.

All the above postulations have one common shortcoming. They are empirical in nature since they are not based on theoretical models which could be used for prediction purposes.

In the present study the experimental data is used to assess the validity of a theoretical reaction model.

Theoretical Reaction Models

Two fundamental reaction models are considered:- (a) dissolution of minerals out of clay to react with lime, and (b) diffusion controlled reactions between lime and soil minerals.

Dissolution model

This model is based on the assumption that the rate of dissolution of a given mass is proportional to its surface area (10), thus:

$$\frac{dw}{dt} = -cA \quad (1)$$

where $\frac{dw}{dt}$ = dissolution rate at time t

c = rate constant

A = total surface area

For uniform sized spherical particles equation 1 becomes

$$\frac{dw}{dt} = -c_1 w^{2/3} \quad (2)$$

where w = weight of undissolved material at time t

$$\text{i.e. } c_2 t = w_0^{1/3} - w^{1/3} \quad (3)$$

$$\text{i.e. } c_3 t = 1 - w_m^{1/3} \quad (4)$$

$$\text{where } w_m = \frac{w}{w_0}$$

and w_0 = weight of undissolved material at time zero

Equation 4 is valid for a monodisperse suspension. For materials of particle size normally distributed, equation 4 becomes

$$c_4 t = 1 - w_m^{1/4} \quad (5)$$

For materials of particle size log normally distributed, equation 4 becomes

$$c_5 t = 1 - w_m^{1/5} \quad (6)$$

Hence if the dissolution theory is valid, experimental results should obey equation 5 or 6.

Diffusion controlled model

Kinetic models for solid state reactions based on diffusion controlled processes have been considered by Jander (12), Ginstling and Brounshtein (13) and Carter (14). The model used by Jander differs from the model of Carter in that it does not consider the effect of the varying volume of reaction products on the rate of formation of new reaction products. In the systems investigated in this study, the volume of products formed with time was not measured, therefore the experimental data was only tested with the model of Jander.

The model of Jander is based on the reaction which occurs between two spherical particles and on the assumption that the rate of thickening of the reaction products is inversely proportional to its thickness, hence:

$$dy/dt = k/y \quad (7)$$

where dy/dt = rate of thickening of the reaction products

k = rate constant

y = thickness of reaction products

Integrating equation 7:

$$y^2 = 2kt \quad (8)$$

The volume of unreacted material V , at time t is given by:

$$V = 4/3 (r - y)^3 \quad (9)$$

$$\text{or } V = 4/3 r^3 (1 - x) \quad (10)$$

where r = radius of reacting sphere

x = fraction of sphere which has reacted

From equations 9 and 10:

$$y = r [1 - (1-x)^{1/3}] \quad (11)$$

Substituting y from equation 8 in 11

$$[1 - (1-x)^{1/3}]^2 = 2kt/r^2 = Kt \quad (12)$$

Equation 12 is the well-known Jander relation.

Hence a plot of time against the first term of equation 12 should give a straight line.

Pozzolan Activity

Pozzolan activity of a soil is defined as the ability of some of the components of the soil to react with lime to produce cementitious products.

The classical method of assessing pozzolan materials (15,16) involves the determination of the amount of lime left in solution after a mixture of the material and Portland cement is kept in water at 313°K for eight days. This method does not take into account the rate at which reaction products are formed during the reaction.

Raask and Bhaskar (11) have proposed a method which takes into consideration the rate at which lime is consumed; they used a diffusion model and proposed a pozzolan index to assess the pozzolan activity of pulverized fuel ashes.

Other empirical methods to quantify the pozzolan properties of materials involve the measurement of some engineering property in relation to time, for example the method of Thompson (17), which is very useful for engineering purposes, consists in measuring the unconfined compressive strength of soils mixed with the optimum percentage of lime (for maximum strength), compacted at maximum density and cured for 28 days. The difference in the value of the unconfined compressive strength of the lime-soil and the strength of the pure soil is termed the lime reactivity of the soil. The reactivity parameter has been used to assess the pozzolan properties of red tropical soils by Harty and Thompson (18) and it is used in this paper in order to ascertain its relation with the method proposed in this study which consists of the assessment of pozzolan activity in terms of the reaction rate and the proportion of reactive components present in the soils.

Materials and Experimental Methods

Nineteen red tropical soils from the states of Paraiba and Pernambuco in Brazil were used for this investigation. The soils were treated with different percentages of high calcium hydrated lime, cured for 28 days at 22°C and their unconfined compressive strengths measured. Their properties and strength behaviour are described in detail elsewhere (19).

Table 1 presents data on the textural composition, compaction and strength characteristics of the 19 soils studied. The value of reactivity as used by Thompson (17) is also included.

The reaction between lime and soil was studied by a method similar to that used by Barret et al. (20) which consists basically in measuring the amount of lime left in solution at different time intervals. The proportion of lime in solution was measured in this study by atomic adsorption spectrophotometry.

The detailed procedure followed during the investigation is described below:

1. Preparation of water-lime solutions by mixing 0.15 g or 0.10 g of lime with 100 ml of CO₂ free distilled water.

2. Mixing of 2.5 g of soil with the appropriate lime solution so as to obtain either 6 or 4.5 per cent lime concentration in the soil. These percentages correspond to the optimum percentages of lime for maximum strength (see Table 1).

3. Placing the mixtures of soil and lime-water in plastic containers tightly sealed to prevent carbonation of the lime.

4. Storing the sealed containers in an oven at a constant temperature of 25°C.

5. Shaking of containers periodically and measuring the lime concentrations at predetermined times. To measure lime concentration by atomic adsorption it is necessary to have very clear solutions, therefore before measuring the amount of lime in solution the lime-water and the soil were centrifuged at 2,500 rpm for about 10 minutes.

A preliminary study was conducted with two soils using three size fractions of each one, i.e. 2mm-0.074mm, 0.074mm-0.02mm and <0.002mm. This was done in order to assess the magnitude of contribution to lime consumption by the coarser fractions of the soil. It is a well-known fact that many of the coarser particles in red tropical soils are only aggregations of the clay size fractions and thus they may influence the overall pozzolan activity.

The results of this preliminary study are presented in Table 2. As expected, the clay size fraction is overwhelmingly the most reactive and therefore it seems apparent that by using the clay size fraction a representative result can be obtained for any of the soils. Consequently the study was only conducted with the clay size fraction of the 19 soils.

Presentation of Results and Discussion

Lime consumption

One of the problems when dealing with red tropical soils, which is still a matter of debate, is related to the nature of the components of the soil which are responsible for its pozzolan properties. Though it is not the object of this paper to deal with this aspect of the overall problem of pozzolan activity, it appears of interest to point out that in red tropical soils the components which appear to be of major influence on the rate of lime consumption are mainly the amorphous silica, alumina and possibly iron compounds while the clay mineral kaolinite is only of minor importance. The amorphous components in terms of percentages on the clay size fraction of the soils studied are presented in Table 3. It can be seen that in terms of, for example, SiO₂ the variations are considerable as well as in terms of the other oxides.

The relations between lime concentration and time obtained during this study are presented in Fig. 1; for clarity of presentation the soils have been divided into four groups as shown in the Figure. These relations show that lime is consumed at a "fast" rate in the initial stages of the reaction, that is, up to five to seven days. From there on the rate of consumption of lime slows down drastically. This is an indication that the uptake of lime after the initial rapid period is very slow and limited in quantity. It is interesting to point out that the total consumption of lime between seven and twenty-eight days corresponds only to eight per cent of the lime consumed during the initial seven

days. Therefore, it appears that the increases in strength of a soil-lime system beyond the seven day period cannot be explained in terms of generation of pozzolanic reaction products, since this should be manifested by greater amounts of lime consumption. From the data shown in Fig. 1 it is apparent that the major part of the reaction takes place within the initial seven days. Thus it is suggested here that the increase of strength shown by these soils beyond seven days is mainly due to the changes that take place in the structure of the cementitious products formed during the pozzolanic reaction, i.e. hydration and increases in crystallinity of the reaction products.

Table 1. Physical and Engineering Properties of the Soils used for the Investigation.

Soil Name	Soil Symbol	Textural Composition			Compaction and Strength Characteristics							Lime Reactivity
		Sand 2mm-0.06mm	Silt 0.06-0.002mm	Clay < 0.002mm	Natural Soil		Lime Treated Soil					
					max. density kg/m ³	moisture content for max. density %	unconfined compressive strength MN/m ²	max. density kg/m ³	moisture content for max. density %	unconfined compressive strength MN/m ²	Lime for max. U.C.S. %	
JOAO P. ACIMA	JPA	79	14	7	1911	10.35	0.29	1901	10.50	1.21	4.5	0.92
JOAO P. MEIO	JPM	60	12	28	2029	13.20	1.80	1944	16.20	5.39	6.0	3.59
JOAO P. ARAUJO	JPB	81	10	9	1861	7.70	0.10	1880	10.10	0.51	4.5	0.41
CUITE	CT	68	20	12	1983	13.85	0.41	1904	15.35	1.14	4.5	0.73
AREIA I	AI	50	20	30	1839	17.35	1.55	1812	18.55	2.05	4.5	0.50
AREIA II	AII	65	18	17	1982	14.20	0.51	1788	17.25	1.60	4.5	1.09
SOLANEA IA	SIA	66	18	16	1980	11.00	0.55	1918	14.70	0.59	4.5	0.04
SOLANEA IB	SIB	86	7	7	1873	4.80	0.08	1950	6.75	0.21	6.0	0.13
SOLANEA II	SII	56	20	24	1982	10.25	0.80	1936	11.35	2.96	4.5	2.16
NOVA FLORESTA	NF	68	19	13	2061	13.30	0.50	1966	15.25	0.99	4.5	0.49
JUNCO I	JI	58	32	10	1939	10.10	0.52	1888	13.80	1.14	4.5	0.62
JUNCO II	JII	76	15	9	2079	12.35	0.20	2046	12.10	0.78	6.0	0.58
TEIXEIRA I	TI	48	30	22	1834	13.95	1.26	1782	15.40	3.22	6.0	1.96
TEIXEIRA II	TII	41	28	31	1753	16.60	1.28	1719	16.85	1.75	6.0	0.47
RECIFE	RC	60	10	30	1918	12.80	0.89	1880	14.40	2.59	6.0	1.70
USINA S. MARIA	USM	46	20	34	1742	17.30	1.85	1710	18.40	2.10	4.5	0.35
S. BANANEIRAS	SB	49	13	38	1916	12.50	1.92	1851	13.45	3.91	4.5	1.99
PLANIA	PI	64	15	21	1828	14.30	1.11	1777	16.50	0.84	4.5	0.00
SAPE MARI	SM	58	19	23	1918	15.30	0.94	1865	17.10	2.49	6.0	1.55

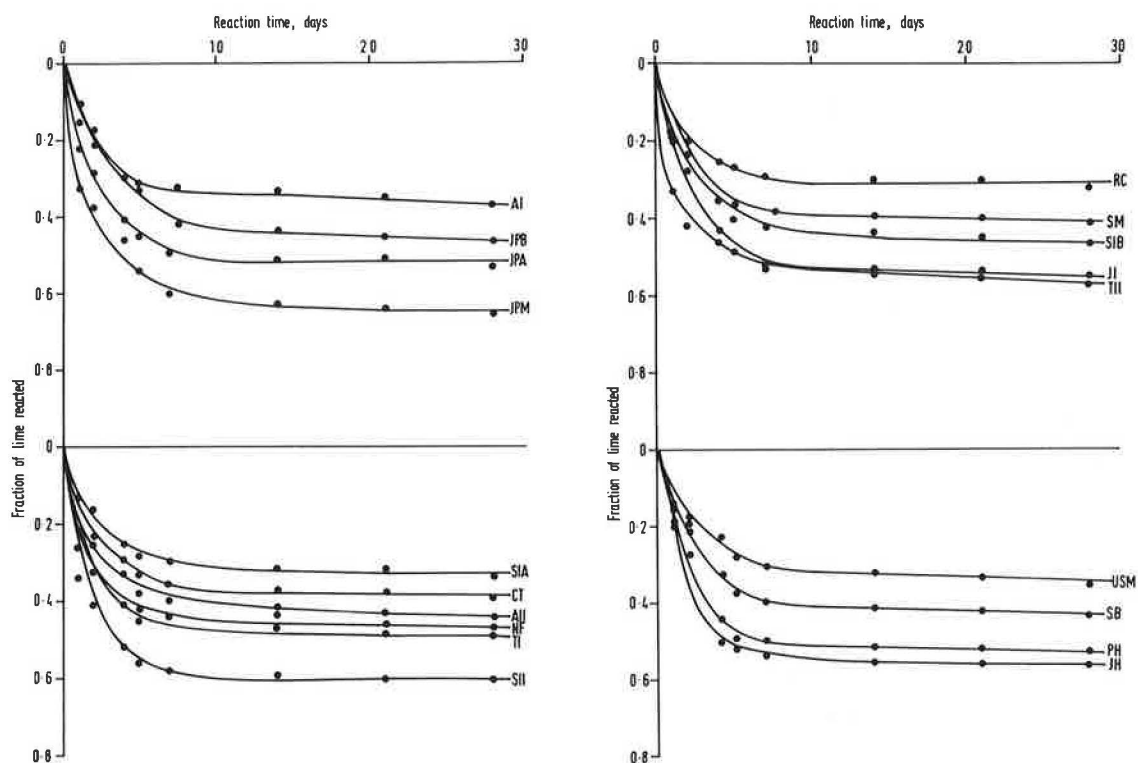
Table 2. Fraction of Lime Consumption at varying times for different fractions of two of the soils investigated.

Soil Name and Soil Symbol	Soil Fraction	Fraction of Lime reacted after				
		1 day	2 days	4 days	5 days	7 days
Joao P. Meio (JPM)	2mm-0.074mm	0.062	0.091	0.098	0.102	0.113
	0.074mm-0.002mm	0.124	0.140	0.162	0.168	0.198
	<0.002mm	0.298	0.367	0.461	0.517	0.589
Areia I (AI)	2mm-0.074mm	0.030	0.065	0.082	0.096	0.117
	0.074mm-0.002mm	0.102	0.115	0.146	0.172	0.196
	<0.002mm	0.092	0.147	0.289	0.314	0.342

Table 3. Amorphous Components of the Clay Size Fraction of the soils investigated.

Soil Name	Soil Symbol	Constituents extracted by 0.5N NaOH		
		%SiO ₂	%Al ₂ O ₃	%Fe ₂ O ₃
Joao P. Acima	JPA	2.81	3.10	0.46
Joao P. Meio	JPM	10.91	7.97	0.68
Joao P. Abaixo	JPB	6.71	6.11	2.51
Cuite	CT	7.84	4.12	0.80
Areia I	AI	8.11	6.95	1.07
Areia II	AII	9.69	5.89	0.62
Solanea IA	SIA	2.08	3.28	1.01
Solanea IB	SIB	4.13	3.97	0.79
Solanea II	SII	11.29	7.56	0.21
Nova Floresta	NF	8.31	7.02	0.43
Junco I	JI	5.02	6.11	0.78
Junco II	JII	6.69	4.07	2.05
Teixeira I	TI	8.12	6.98	0.30
Teixeira II	TII	4.68	4.02	0.87
Recife	RC	9.21	6.16	0.10
Usina S. Maria	USM	3.71	4.41	0.19
S. Banareiras	SB	8.19	6.84	0.28
Penha	PH	2.91	3.80	0.29
Sape Mari	SM	7.98	6.48	0.78

Figure 1. Relations between consumption of lime and time for the nineteen soils investigated.



Mechanism of reaction

The numerical values of the lime consumption vs. time relations were used to test the mathematical models discussed previously.

It was found that the experimental results do not follow the relations based on the equations which represent the dissolution theory, despite the fact that the experimental conditions were such as to give monodisperse suspensions for which the equations are intended to be valid. This finding coincides with the results of other investigators (10) which reported that the dissolution model was not applicable to pulverized fuel ashes.

The experimental results were found to approximate very closely to the values obtained with the equation of Jander based on the diffusion model. The deviation of the experimental results coincides with the time at which the rate of lime consumed changes, i.e. five days. The experimental points and the lines corresponding to the Jander equation are shown in Fig. 2. Fig. 3 shows the relation between reacted lime and time for the total period investigated for some of the soils. This figure highlights the fact that the experimental data follows the theoretical relation only for a limited time period, i.e. five days. The deviation beyond five days may be explained by the fact that one of the weaknesses of the Jander equation is its inability to incorporate the effect of the varying volume of reaction products being formed and therefore their influence on the reacting system. It is apparent then, that the increased volume of reaction products reduces the rate of diffusion as shown in Fig. 3.

Figure 2. Relations between the first term of Jander's equation $F(x)$ and the time of reaction for some of the soils.

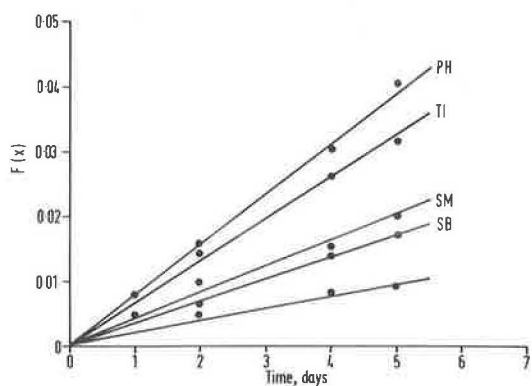
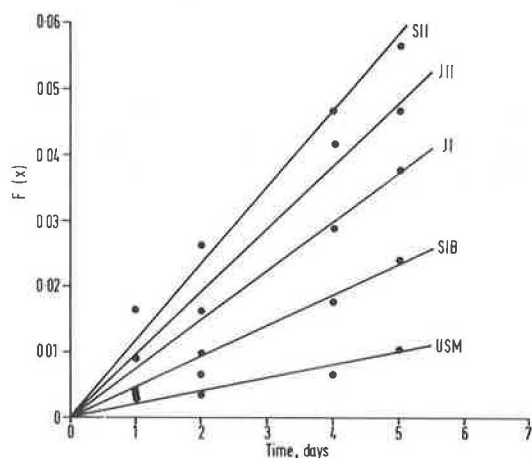
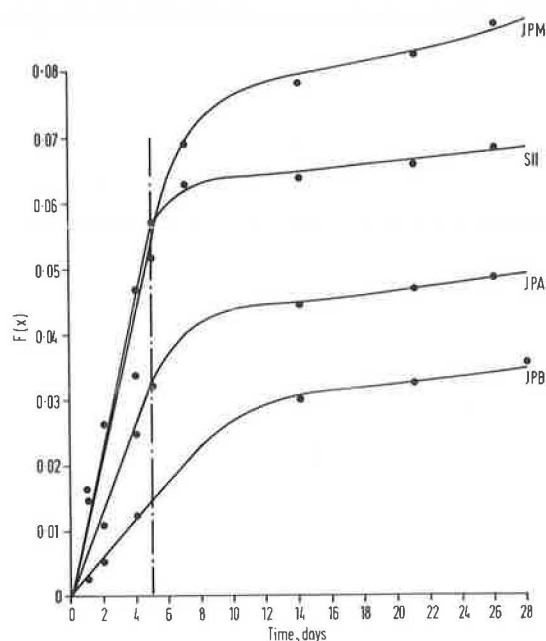


Figure 3. Relation between the first term of Jander's equation and the time of reaction up to 28 days for four of the soils studied.



It must be pointed out that the findings reported here may not represent other types of soils, since the major characteristic of the red tropical soils studied is their composition in terms of amorphous components, which react with the lime probably at a faster rate than crystalline clay minerals due to their high specific surface and their reactivity (21). Based on this fact, i.e. the high reactivity of amorphous components of the soil, the amelioration phenomena can be explained in terms of the rapid formation of cementitious products which cause changes in the engineering properties of the soils.

Figs. 4 and 5 are presented to show the micro-morphology of the reaction products, they were obtained with a GEOL Scanning Electron Microscope and show very clearly massive plates of hydrated calcium silicates, which are the main hydration products of red tropical soils and lime (22). Fig. 5 which is a magnification of Fig. 4 shows furthermore evenly distributed amorphous components which are presumably the non crystalline reaction products.

Pozzolanic activity

From the findings of other investigators and the results presented here it is apparent that pozzolanic activity is directly related to the reaction rate and the quantity of the reacting components of the soil.

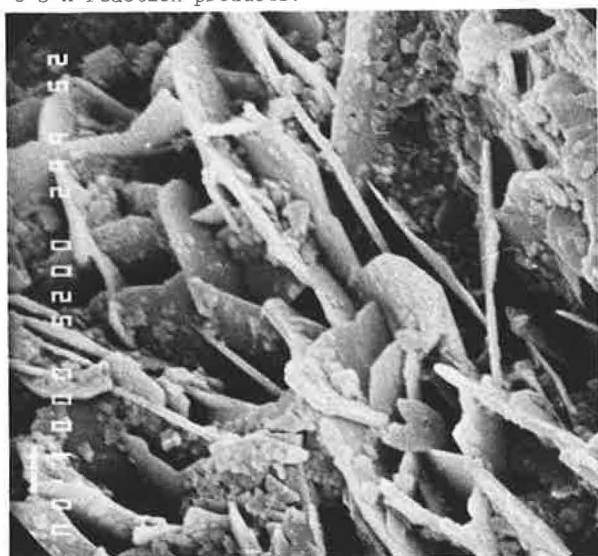
The slopes of the lines presented in Fig. 2 were used to determine the reaction rate constants for the nineteen soils and the quantity of reacting components was expressed by the amount of material smaller than 0.002 mm. It was decided that the clay size fraction should be the fraction more representative of the reacting components, though it is recognised that the most active components are really the amorphous materials in the clay size

fraction.

Figure 4. Scanning Electron Micrograph, Magnification X3000 showing the micromorphology of C-S-H and scattered amorphous reaction products.



Figure 5. Detailed view of Fig. 4, Magnification X6600 showing massive plates characteristic of C-S-H reaction products.



It is proposed here that the pozzolanic activity of a red tropical soil should be expressed by an activity index which is related to the rate constant and the percentage of reacting compounds by the following equation:

$$A_i = K n^x$$

where A_i = activity index

K = rate constant

n = percentage of clay-size fraction

x = a constant dependent on the type of material

Although the introduction of the constant x may seem arbitrary, it is based on the assumption that different materials may have different types of reacting components within their clay size fractions.

For the soils of this study it is proposed that x takes the value of 2.

The values of the rate constants and the activity indexes for the soils are presented in Table 4. It can be seen that the A_i values range from a value of 0.12×10^{-4} to a value of 4.08×10^{-4} giving really a wide range for classification purposes.

Table 4. Rate Constants and Activity Indexes for the Soils Investigated.

Soil Symbol	Rate Constant	Pozzolanic Activity Index $A_i \times 10^{-4}$
JPA	0.0035	0.172
JPM	0.0052	4.08
JPB	0.0015	0.12
CT	0.0016	0.291
AI	0.00124	1.116
AII	0.00234	0.791
SIA	0.00108	0.461
SIB	0.00245	0.120
SII	0.00543	3.13
NF	0.00279	0.472
JI	0.00384	0.384
JII	0.00471	0.567
TI	0.00340	1.646
TII	0.00384	3.689
RC	0.00118	1.062
USM	0.00108	1.248
SB	0.00174	2.512
PH	0.00411	1.812
SM	0.00202	1.07

The usefulness of the activity index should be proved against other accepted parameters, this was done in this paper by correlating the activity index with the unconfined compressive strength of the soil lime systems and also by investigating its relation with the well-known reactivity parameter of Thompson (17).

The relation of A_i vs. unconfined compressive strength is shown in Fig. 6. The equation obtained for this relation is:

$$UCS = 1.639 A_i + 0.0135 A_i^2 - 0.0276 A_i^3 + 0.33$$

where UCS = unconfined compressive strength of the soil-lime system in MN/m^2 .

A_i = activity index $\times 10^{-4}$ per day.

This equation was found to be statistically highly significant with a multiple R equal to 0.97327 at 95% confidence limit. The standard error of estimate is 0.3586.

The relation of the lime reactivity parameter of Thompson and the activity index proposed in this paper is shown in Fig. 7. This relation, unlike the one shown in Fig. 6, is a linear relation of the following form:

$$LR = 0.72 A_i + 0.278$$

where LR = lime reactivity parameter in MN/m^2

A_i = activity index $\times 10^{-4}$ per day

This equation is statistically highly significant, with R equal to 0.88 at 77% confidence limit. The

standard error of estimate is 0.4567.

Thus it can be seen that the activity index obtained by the use of a theoretical model is a valid representation of the pozzolanic activity of the red tropical soils investigated in this study.

Figure 6. Relation between activity index and the unconfined compressive strength of the red tropical soils.

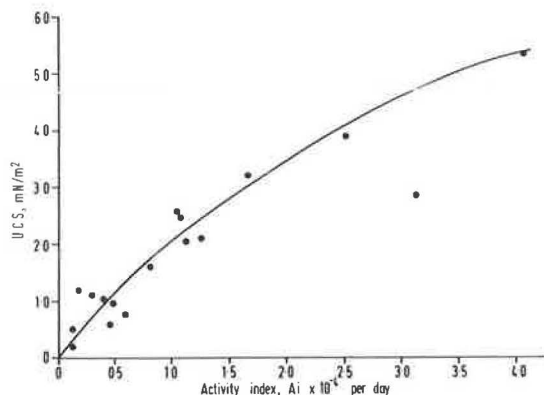
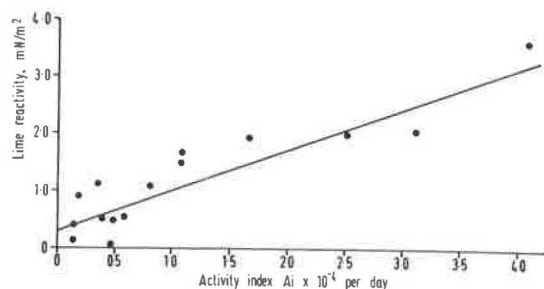


Figure 7. Relation between the activity index and the parameter of Thompson.



Conclusions

1. Formation of reaction products as represented by the consumption of lime takes place during the initial period of five to seven days, therefore the gains of strength beyond this period appear to be due mostly to the hydration and increased degree of crystallinity of the reaction products formed during the initial five to seven days period.

2. Under the experimental conditions of this study the reaction between the soil and the lime is mainly a diffusion controlled process obeying the equation of Jander for the initial period of five days.

3. Amelioration in red tropical soils appears to be a result of the initial rapid reaction between the amorphous soil minerals and lime which form products of initially very weak cementitious properties.

4. The activity index based on the diffusion model and the quantity of reacting compounds of the soil is statically related to the unconfined compressive strength of the soil-lime system.

5. A highly significant statistical linear

correlation links the empirical reactivity parameter of Thompson and the proposed activity index. This finding is of significance since it allows the use of either of the two values for assessing the pozzolanic properties of these materials. The advantages of the A_i value are that it is a parameter based on a theoretical model and that it can be obtained faster than the reactivity value which requires not only time but great input of labour.

6. Since the activity index gives a wide range of values it may be used to classify red tropical soils. However, a limiting value of the Activity Index A_i which will separate the pozzolanic from the non-pozzolanic soils is very difficult to propose at this stage of knowledge. Thompson (17) has indicated that a reactivity value of 0.35 MN/m² (50 psi) should be this limit, while other researchers (7) have emphasised a minimum value of strength linked to a particular requirement.

Acknowledgements

The authors would like to acknowledge the help given by the authorities of the University of Paraiba, Brazil for the collection of samples and some of the physical testing while the senior author was Visiting Professor of Civil Engineering at the "Centro de Ciencia e Tecnologia" of the University of Paraiba in Campina Grande. They would like also to thank the Central Electricity Generating Board, Scientific Division, Harrogate, U.K. for providing excellent facilities for the atomic absorption determinations and the scanning electron microscopy photographs.

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