EXPANSION OF CONCRETE CONTAINING NEW ZEALAND ARGILLITE AGGREGATE

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SUMMARY

The expansive behaviour of concrete containing (a) Matahina greywacke-argillite (0.04% in 12 years) and (b) a lightweight aggregate produced by heat treatment of Oxford argillite (0.13% in 5.5 years) is described. The reaction mechanism for the expansion of concrete containing Matahina greywacke-argillite is still not known. An alkali-silica reaction is unlikely to have been the cause of the expansion as both pozzolan and low alkali cement were used and no sign of alkali-silica reaction was found by petrographic examination. Calcium oxide was identified in the fired Oxford argillite, but the expansive reaction due to the rehydration of this calcium oxide is unusual because of its small grain size and low concentration in the fired aggregate.

INTRODUCTION

The expansion of concrete due to alkali-aggregate reaction has been found to be caused by a reaction between the alkalis present in cement and certain types of siliceous minerals giving rise to the formation of expansive alkali-silica gels. This reaction was extensively reviewed by Bredsdorff et al[1] and many other investigations have been reported since. More recently, another type of alkali-aggregate reaction has been described which appears to involve the interaction of the cement alkalis with some of the clays present in greywackes, argillites and phyllites, causing a slow but steady expansive reaction which in some cases has continued for over thirty years.[2,3]

The clays identified to date are interlayered vermiculite chlorite and possibly montmorillonite. It has been suggested that the cement alkalis remove interlayer cementing precipitates of alumina and silica from these clays allowing chemisorption of water on the newly formed surfaces with resultant swelling pressures. Generally the reaction has been characterised by the absence of alkali-silica gel, but where this has been present it has been attributed to being derived from the interlayer silica. Tests have indicated that the expansive reaction is suppressed by the addition of pozzolanic material to the concrete.[2]

Recently, in New Zealand, two cases of expansive reactions in concrete involving a mixed greywacke-argillite aggregate from Matahina and a fired lightweight argillite from Oxford became apparent. Preliminary investigations indicated that, in both cases, alkali-clay reactions might be involved. This paper reports the result of investigations on these two aggregates and attempts to relate the reactions to those already described.

AGGREGATES

(a) Matahina Greywacke-argillite

Prior to the construction of the Matahina hydro project, located on the Rangitaiki River, Bay of Plenty, the durability of the local greywacke was questioned. Breakdown of the aggregate in stock-piles was noted and ascribed to the presence of significant proportions of argillite. As a result of investigations it was recommended that Matahina greywacke should not be used in any concrete subject to exposure, especially face concrete, but its use in mass concrete should be satisfactory. Matahina greywacke was used for the main structure which is of earth core design.

As part of the above investigations, concrete test specimens were prepared using Matahina greywacke-argillite and Hutt Valley greywacke a material of long proven performance, to a mix design specified by the Ministry of Works and Development. Some of these specimens were retained on an outdoor exposure site to determine their long term durability. The details of the concrete mixes used are given in Table 1.

TABLE 1

Details of Concrete Mixes used with Matahina and Hutt Valley Greywackes and Results of Exposure Tests

Sample	15A	17A
Aggregate	Matahina	Hutt Valley
Cement + pozzolan (kg/m^3)	410	410
Pozzolan (% replacement of cement	12.5	12.5
W/C+P	0.50	0.50
Air content (%)	2.8	3.3
Slump (mm)	18	28
Date mixed	30/1/63	31/1/63
Length change (% in 12 years)	+0.04	-0.01

The cement used was an ordinary Portland type with 0.32% Na_2O equivalent alkali contant and the pozzolan Whirinaki diatamaceous pumicite. The beams, 75 x 75 x 290mm in dimension, were cast and cured in 100% r.h. for seven days at 21C and then transferred to the outdoor exposure site.(4)

The Hutt Valley greywacke is a good example of the New Zealand greywacke which is widely used as an aggregate and has been described by Kennerley and St. John.(4) The Matahina greywacke is similar but differs in some details. The aggregate contains about 10% of argillite (which is usually absent from good grade greywacke aggregates) and approximately 5% of rhyolite, ignimbrite and pumice are

present. The siltstone to sandstone grades of the greywacke show typical texture and are composed predominantly of quartz feldspars, scattered tiny mica flakes and tiny chips of fine grained volcanic rock and argillite. The greywacke contains rare irregular veinlets of white encrustations of laumontite. Some of the greywacke fragments are a yellow brown colour and possibly have been derived from weathered terrace gravels. The argillite fragments are smaller in size than the greywacke, less hard and often easily broken by hand. Mineralogically, the argillite is similar to the greywacke but contains a higher proportion of clay minerals. The above description of Matahina greywacke is a private communication from W.A. Watters, Chief Petrologist, N.Z. Geological Survey.

(b) Fired Oxford Argillite

The large deposit of argillite located in the Ashley Gorge, near Oxford in the Canterbury Province, has been investigated and found suitable as a raw material for the manufacture of an expanded lightweight aggregate.(5) Tests showed that another large deposit of argillite located behind the Wellington Patent Slip was also suitable and the results for this material are included for the purposes of comparison.(5) As these two deposits appeared to have good potential as raw materials for commercial production, quantities of expanded lightweight argillite were produced in a pilot rotary kiln at 1150C and a full range of concrete tests carried out. Details of mixes are shown in Table 2, and specimens were placed on the outdoor exposure site to determine their long term durability.

TABLE 2

Details of concrete mixes and results of exposure of expanded argillite specimens[a]

Bar No.	Aggr. C.A.	Source F.A.	Cement content (K/gm^3)	W/C+	Slump (mm)	Unit Weight (Kg/m^3)	Age (yrs)	Length change (%)	Carb.* area (%)	Carb.* Depth (mm)	Cracking*
F8/1/2	Ox.	Ox.	260	1.1	0	1120	5.5	-0.014	67	12	Nil
F8/1/1	"	"	290	1.1	50	-	"	+0.014	54	8	Mod.
F8/1/3	"	"	350	0.83	25	1215	"	+0.044	28	4	Slight
F8/1/4	"	"	435	0.67	68	1275	"	+0.088	12	2	Slight
F8/1/6	"	"	450	0.60	0	1340	"	+0.071	10	2	Mod.
F8/1/5	"	"	530	0.60	0	1445	"	+0.060	5	1	Mod.
F8/1/7	"	"	560	0.56	87	1415	"	+0.126	8	1	Severe
F8/1/12	H.V.	Ox.	330	0.93	25	1675	"	+0.037	55	7	Mod.
F8/1/13	Ox.	H.V.	360	0.63	43	1635	"	-0.028	5	1	Nil
F10/4	P.S.	P.S.	350	0.92	62	1345	6.3	-0.020	51	8	Nil
F10/9	"	"	540	0.56	87	1560	5.7	+0.009	5	1	Mod.

\+ W/C includes total water in mix.

Ox. = fired Oxford argillite

P.S.= fired Patent Slip argillite

* Based on measurements from thin section

H.V. = Hutt Valley greywacke (dense)

a. The 50 x 50 x 300 beams were cured 7 days in fog at 21C and then placed on the exposure site

Large deposits of argillite are not common in New Zealand as most argillite is interstratified with the greywacke. The Patent Slip argillite is a slightly metamorphosed, indurated, pale green-grey claystone with some chert pods. The Oxford argillite is slightly finer grained in texture and blue black in colour. Both argillites show incipient foliation.

EFFECTS OF LONG TERM EXPOSURE

(a) Matahina greywacke - argillite. For the first two years the specimens containing Matahina greywacke were relatively stable, but since then they have expanded at a slow rate, until at the end of twelve years the length change is +0.04% and still continuing undiminished. In contrast, the specimens containing the reference aggregate, Hutt Valley greywacke, contracted quickly within the first two years and the length change has now stabilised at -0.01%. Under low power stereomicroscopic examination both sets of specimens appear similar and show normal weathering and microcracking.

(b) Fired Oxford argillite. The dimensional changes of specimens containing fired Oxford argillite and some combinations with dense Hutt Valley greywacke, and also of fired Patent Slip argillite are shown in Table 2. Like the Matahina greywacke concrete these specimens were relatively stable for the first two years but once expansion started it continued undiminished. By contrast, the fired Patent Slip

argillite is stable. Under low power examination the concrete beams with low cement contents are showing signs of considerable weathering while some of the beams with high cement contents cracked in half when lifted from the site for measurement.

The details and results of the petrographic examination of the concrete beams containing the above aggregates are given in the appendix.

ANALYSIS OF CLAY FRACTIONS IN AGGREGATES

Samples of greywacke and argillite were handpicked from the Matahina greywacke-argillite used for the concrete tests, crushed to pass a 52 mesh sieve and shaken for sixteen hours in water at pH 9. The minus 2 um fraction was removed by centrifuging and the fractions were analysed by powder X-ray diffraction, thermal analysis and infra red spectroscopy. Similar procedures were applied to the Oxford and Patent Slip argillites both in the raw and fired states. In addition Whites microscope test for free lime was applied to some of the fired argillites.

RESULTS OF CLAY ANALYSES

(a) Results of the analyses showed the handpicked greywacke and argillite fractions of the Matahina aggregate to be almost identical apart from some minor details. Approximately 10% and 7% of the minus 2 um fraction were recovered from the greywacke and argillite respectively. The minus 2 um fractions were principally quartz, feldspars, iron chlorite and either illite or finely divided micaceous material. The chlorite peaks were destroyed by digestion in 1 N HCl at 85C for 4 hr and many of the chlorite peaks were removed or flattened by heating at 550C. Attempts to expand the clay structures, both with 10% glycerol in water and ethylene glycol in the vapour phase at 60C, with and without Mg as the cation were unsuccessful. A typical set of XRD results are shown in figure 1. The 2 to 300 um silt fractions consisted of quartz and feldspars with some residual clays present. The above results were confirmed by thermal analysis and infrared spectroscopy.

(b) Results obtained from analysis of the Oxford and Patent Slip argillites appeared similar to that reported above. Approximately 7% of the minus 2 um fraction was recovered from each of the raw argillites. In the minus 2 um fractions (0.3% recovered) extracted from the fired argillites no clays were detected. These minus 2 um fractions contained quartz, feldspars and calcite for the fired Oxford argillite and quartz and feldspars only for the fired Patent Slip material.

No clays were detected in the minus 2 um fractions. Whites test gave a positive result for crushed, freshly fired Oxford argillite but was negative for similarly treated Patent Slip material.

DISCUSSION

(a) Matahina greywacke argillite.

No observations of the Matahina greywacke-argillite show any reason why this aggregate should have expanded in concrete. The presence of chlorite and illite or micaceous materials as the main clay constituents is typical for these types of sedimentary mesozoic New Zealand rocks. (W.A. Watters. Personal communication). Further, the expansive reaction is unusual in that not only was a low alkali cement used, but a pozzolan was also present. Thus the possibility of an alkali-silica reaction seems unlikely and this is further supported by the petrographic examinations.

There remains two other possibilities. Firstly, the degree of expansion occurring is moderate, and thus the amount of reactive rock could be small and may have been undetected in the analyses. Secondly, mineral alteration could be occurring under the moist alkaline conditions in the concrete. Some alteration of the feldspars to sericite is common in these geosynclinal greywackes though it is not clear how much of this alteration is post-depositional. This existing

alteration made it impossible to determine whether any further feldspar alteration had occurred in the concrete.

Therefore at this stage it must be concluded that the expansive mechanism of Matahina greywacke-argillite is still unknown and cannot be related to the alkali-clay reactions reported by Gillott et al.[2] Neither has it been possible to associate the expansion with either the greywacke or the argillite individually. In the Matahina aggregate, greywacke predominates, but a small amount of reactive argillite could easily be responsible for the reaction. Generally, argillite is not a problem in New Zealand aggregates as quarries are chosen and worked to exclude this material because of its poor physical properties. It is only at the margins of the geosyncline that problems of argillite and marginally acceptable greywacke arises.

(b) Fired Oxford Argillite

The results of analyses of fired Oxford argillite indicate that the hydration of hard burnt lime is responsible for the expansion. Chemical analysis of the raw argillite[6] did not indicate widely disseminated calcite as being present, as the amount of calcia reported 0.50% was believed to be associated with the feldspars. Whites test on fired Oxford argillite which had been stored in open drums for five years was negative and it was not until the minus 2 um fraction from this five year old material was extracted that it was realised that calcite was probably present in the raw argillite. This

is the reason why it was initially believed that unburnt clays could still be present, as Blank[7] reports their presence is one of the causes for unsoundness in fired lightweight clay aggregates.

The small amount of hard burnt lime present, estimated at less than 0.5% by XRD, has been slow to carbonate and hydrate, the fired material was not mixed in concrete until approximately one year after manufacture and expansion is still occurring in the concrete beams six years later. The expansion is associated with the fine aggregate, as the results for beams F8/1/12 and 13 clearly show, which suggests that the hard burnt lime is protected by some type of sintered or glassy coating. If this is the case, the smaller particles of the fired argillite would be more reactive as there is more crushed material and thus more broken surfaces in this fraction. The fact that fired Oxford argillite is an overbloated, well sealed aggregate gives further support to this argument.

Data shown in Table 2 is confusing. If we consider the first series F8/1/1 to 7, carbonation, cracking and expansion can be related to cement content and increasing unit weight. However when we consider the other mixes reported the picture is not so clear. It is hypothesised that increasing alkali content due to increasing cement content is a factor in that sintered and glassy coatings on the hard burnt lime would be more efficiently attacked at higher cement contents. Further work is required to demonstrate this hypothesis.

CONCLUSION

In conclusion, the results suggest that the recent statement by Kennerley and St. John[4] "that no evidence has been found to suggest that the type of argillite commonly found in New Zealand would react deleteriously with cement alkalis" is no longer valid. In the case of Matahina greywacke-argillite, expansive alkali-aggregate reaction should not be possible as both low alkali cement and pozzolan were used and there should be insufficient alkalis available to initiate the reaction. Yet expansion has occurred in a manner similar to that reported for the Nova Scotia greywackes and argillites. While the underlying mechanism of the expansion in the Matahina greywacke-argillite is still unknown, the physical expansion of concrete containing this concrete is an indisputable fact. Previously, greywacke from the shallow margins of the geosyncline have not been used extensively for concrete aggregate, but their increasing use makes it imperitive that means be found for detecting potentially reactive greywackes.

In the case of the fired Oxford argillite, the cause of concrete expansion, at first suspected as an expansive clay reaction, is probably due to rehydration of lime mediated by alkaline attack on sintered or glassy coatings. It is necessary to be cautious in this conclusion. The fine grain size of the calcium oxide and its resistance to hydration is unusual. Both Blank[7] and Budnikov et al[8] state that particles of calcium oxide need to be at least approximately

5 mm in diameter to resist hydration by the water in the concrete mixer.

In both cases, discussed above, though the concrete expansions have been slow and moderate in size, they have been quite sufficient to cause damage to concrete as witnessed by breakage of specimens on the outdoor exposure site. Damage at the Matahina hydro project has not been reported to date, but investigation will be carried out to try and locate areas where greywacke may have been used inadvertently.

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APPENDIX

PETROGRAPHIC EXAMINATION OF CONCRETE SAMPLES

Matahina and Hutt Valley Aggregates

Concrete beams containing these two aggregates showed signs typical of exposure to the weather. Much fine aggregate was exposed and the surrounding cement paste was soft to a superficial depth. However most exposed particles were still firmly embedded. The original cast surfaces remained visible in a few isolated patches. Both beams were pattern-microcracked, with many of the cracks being filled with white crystalline material probably calcite. Examination of fracture surfaces in pieces broken from beams failed to reveal any reaction rims around pieces of aggregate or any other signs of deterioration apart from a few isolated microcracks.

For the purposes of microscopic examination thin sections were cut from beams 15A and 17A from the middle and near one end in each case. Drawings of the sections cut from 15A are shown in figure 2. Cracking is indicated by black lines and carbonation areas by cross hatching.

Beam 15A Containing Matahina greywacke-argillite

The cracking in figure 2 is extensive in some areas and almost absent in others. Main cracks are approximately 40 um

in width with smaller cracks ranging down to 10 um. Cracks generally skirt particles of aggregate but in a number of cases run through them. General carbonation of the cement paste extends to depth of 2-3 mm around the edges and this layer is not noticeably leached. In many of the larger cracks carbonation extends to a depth of 20 mm. Considering the cracking present the limited penetration of the carbonation suggests that drying of the specimens under the exposure conditions has not at any time been extensive.

The amount of calcium hydroxide in the cement paste is consistent with the high cement content used. Leaching of the cement paste is generally absent even at carbonated edges. However, around a number of the larger aggregate particles there is a ring of cement paste in which birefringent crystals are absent suggesting a deficiency in calcium hydroxide. While this effect is somewhat haphazard it is noticeable and suggests some movement of precipitated material into the aggregate particles concerned.

The larger air voids in the concrete are only occasionally rimmed with calcium hydroxide but many of the smaller pores are completely filled with this material. Ettringite was not present in any of the pores or cracks. Generally, the hardened cement fabric is dense and sound apart from cracks and no alkali-silica gel or other reaction products were noted either in the cement paste or particles of aggregates.

The coarse aggregate is a mixture of greywacke, sandwacke and argillite. The fine-grained matrix of the rock is variable and complex and appears to consist of a mixture of quartz, and feldspars in a highly birefringent fine-grained mass of mica flakes together with tiny chips of volcanic rocks and argillite. Laumontite and prehnite were present in some particles as vein material but not in significant quantities. The fine aggregate is similar to the coarse aggregate but individual pieces tend to be more homogeneous. There is a significant proportion of acid volcanic rocks present, such as rhyolite, ignimbrite, rhyolitic pumice and even a few pieces of andesite. The larger fragments of the aggregate are often rounded and appear water worn. Many of these pieces are cracked and weathered and physically these fragments cannot be considered suitable for high grade concrete. It was not possible to distinguish any signs of alteration in the fine-grained matrix of the greywacke or argillite due to possible reaction with the cement.

Beam 17A Containing Hutt Valley greywacke

The cracking and carbonation is outlined in figure 3. There is less cracking than in beam 15A and it is of a different character. Most of the cracks are shorter and do not extend far into the fabric of the concrete. Where cracks are present in the interior they are mainly discontinuous and much narrower than those present in beam 15A. Apart from this, the general state of the hardened cement paste is

similar to beam 15A except that the band of paste deficient in calcium hydroxide surrounding many aggregate particles is much less pronounced.

The aggregate is a typical example of Hutt Valley greywacke. It is more homogeneous than the Matahina greywacke in the sense that the fabric does not seem to be as complex or such a mixture of size ranges and the micaceous material is not as noticeable. Argillite is rare and the only volcanic rocks present are spillites. Prehnite is present as vein material but laumontite was not noted. In general, the aggregate is more compact, and less weathered than that from Matahina.

Fired Oxford and Patent Slip Argillites

The surfaces of the beams with low cement contents were soft with little aggregate exposed. Pattern microcracking was visible on the surface of the beams from F8/1/3 to 7 and displacement of these cracks was evident in beams F8/1/6 and 7, where some discolouration of crack edges occurred. Many of the cracks were filled with calcite. Examination of fracture surfaces in pieces broken from the beams did not reveal the presence of any rims around aggregate particles or any other signs of deleterious reactions. The same trends were found with beams F8/1/12 and 13 and F10/4 and 9.

Beams F8/1/1 to 7 Containing fired Oxford argillite.

The cracking and carbonation is shown in figure 4. There is a marked change in the texture of the hardened cement fabric of these beams as the cement content increases. At the lowest cement content, carbonation is intense and leaching is apparent but cracking is almost absent. Weathering and carbonation is sufficiently severe to have weakened the fabric and many of the larger particles of aggregate have been penetrated by carbonation products. In beam F8/1/3 carbonation is more normal and less leached. At the higher cement contents the hardened cement fabric is typical of dense concrete and penetration of aggregates by carbonation products is absent. However cracking in these samples is severe both in the cement fabric and also in many fragments of aggregate. The transition zone between carbonated and uncarbonated cement paste is abrupt even in the most intensely carbonated beams. Around many of the larger fragments of aggregates the zone of cement paste deficient in birefringent crystals was noted suggesting a movement of calcium hydroxide into the aggregate particles concerned.

The fired Oxford argillite is heavily overbloated and there are numerous very large pores separated by thin septums of highly frothed glass. The outer rims of the fired aggregates are brown and well sealed where the original surface is intact. Numerous minute birefringent crystals are present scattered through the glassy matrix of the aggregate fragments. These crystals were too small to be positively identified but generally appeared to be quartz and feldspars.

Beams F8/1/12 and 13 Containing mixtures of fired Oxford argillite and dense Hutt Valley greywacke.

The cracking and carbonation is shown in figure 5. The concrete in beam F8/1/12 appears similar to beam F8/1/3 or 4 in degree of carbonation and cracking. The replacement of the fired coarse aggregate by dense Hutt Valley greywacke has had little effect. In contrast, where the fired fine aggregate has been replaced in beam F8/1/13 the effect is dramatic. The concrete now has all the appearance of a normal dense concrete interspersed with lumps of fired argillite. This beam has weathered extremely well.

Beams F10/4 and 9 Containing fired Patent Slip argillite

Cracking and carbonation are shown in figure 6. Both these beams showed the same trends as the F8 series. The degree of cracking in beam F10/9 appears greater than warranted by the expansion of 0.009%, but cracks are much narrower than for beam F8/1/5.

The fired Patent Slip argillite is denser and not as heavily bloated as the fired Oxford argillite. Some foliation is present and more unbloated particles were noted. Penetration of carbonated products into rims of aggregate particles was absent.

CAPTIONS TO FIGURES

Fig. 1 Typical X-ray diffraction patterns of Matahina argillite.

a. K saturated

b. Mg saturated sprayed with 10% glycerol in water.

c. K saturated and heated 550 C/4hr.

d. No cation. Digested in 1N HCl for 4hr/85C

X-radiation Cu Ka Carbon monochrometer used.

Scan rate ½°/min sensitivity 200 c.p.s.

Time constant 4 sec. Scale factor 1

Fig. 2 Outline drawing of thin sections of Matahina greywacke-argillite showing cracking and carbonation. Cross hatching delineates carbonation. End and middle sections. 75 x 75mm sections.

Fig. 3 Ditto. Hutt Valley greywacke

Fig. 4 Ditto. Fired Oxford argillite. 50 x 50mm sections. See table 2 for details.

Fig. 5 Ditto. Fired Oxford argillite and dense Hutt Valley greywacke mixtures. 50 x 50mm sections.

Fig. 6 Ditto. Fired Patent Slip argillite. 50 x 50mm sections.

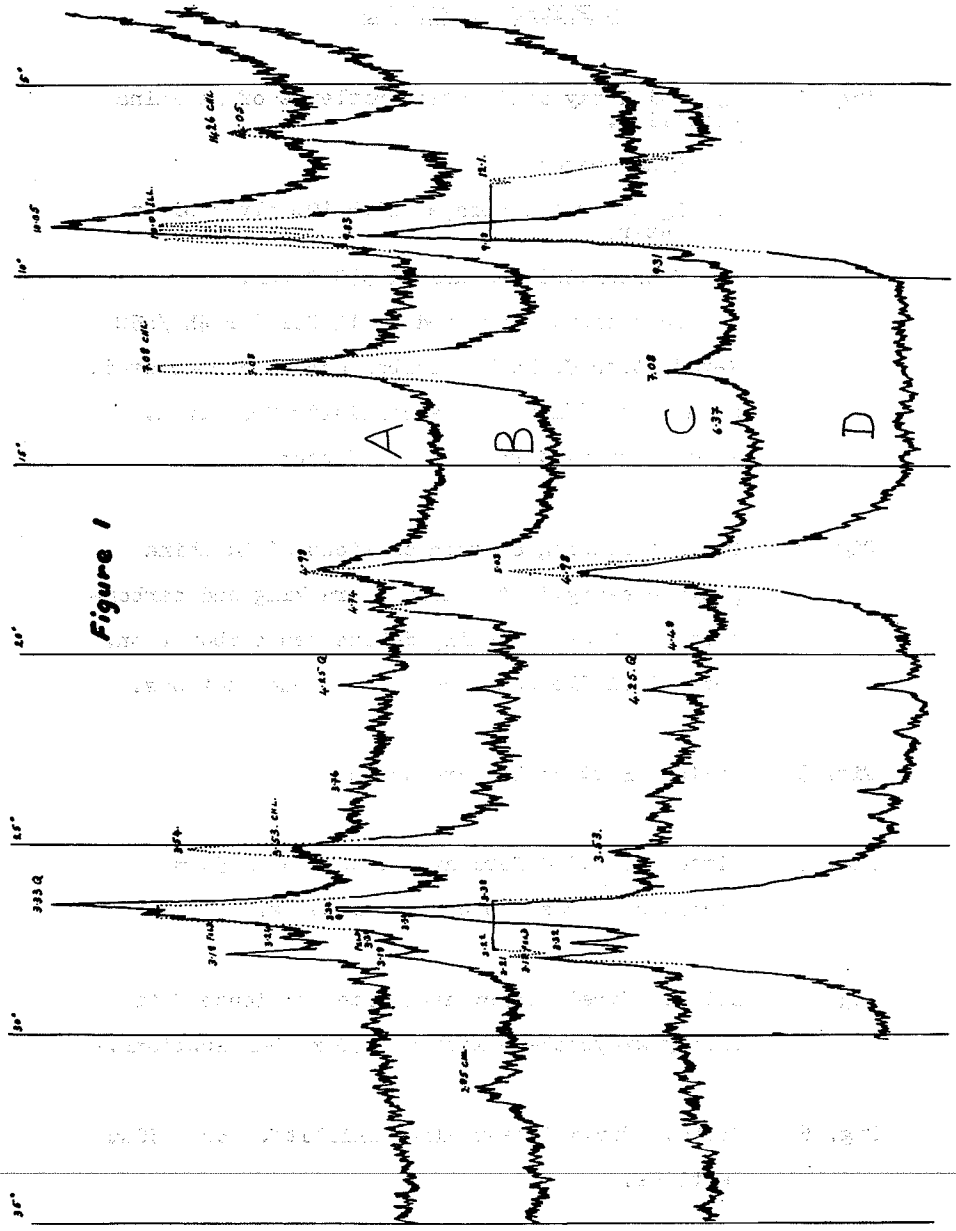
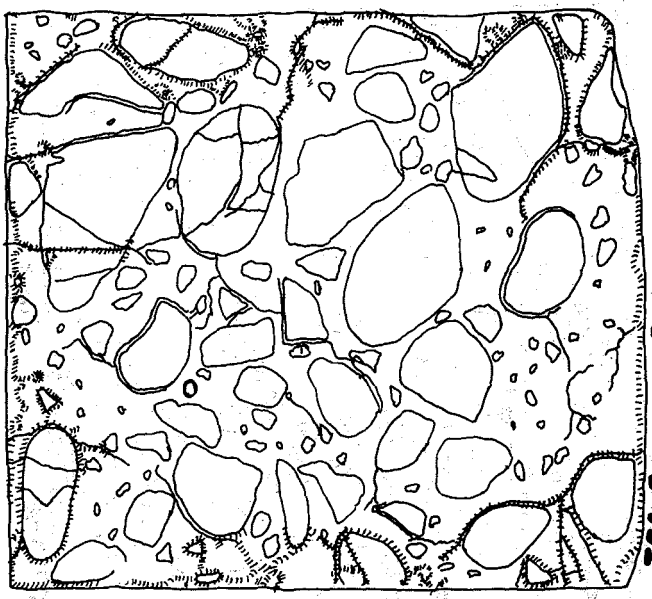
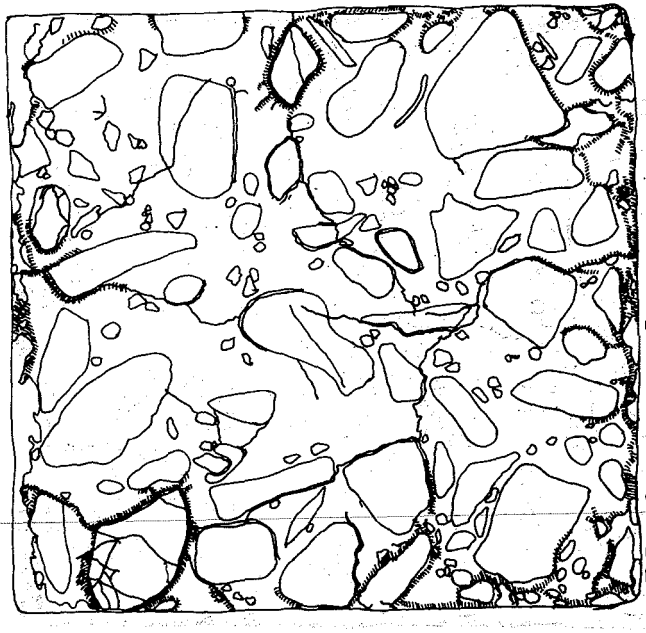


Figure 1



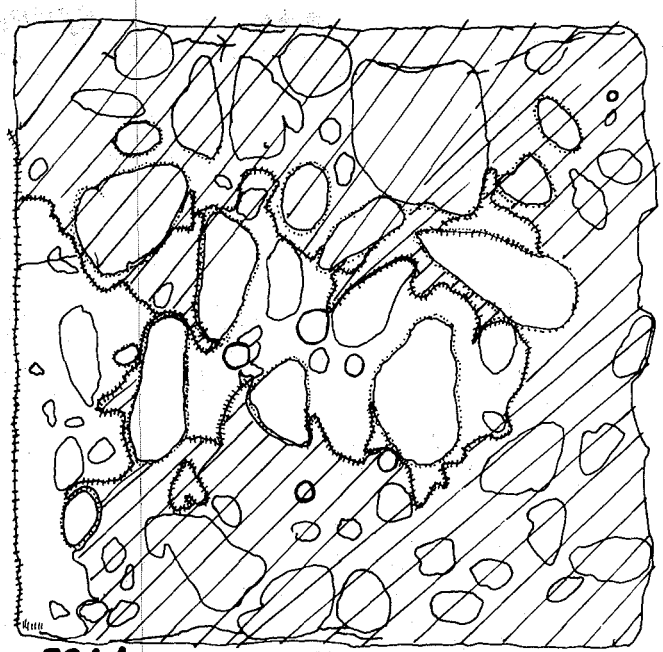
ISA-E Fig.2.



ISA-M Fig.2.

Fig.3
17A-E

Fig.3.
17A-M



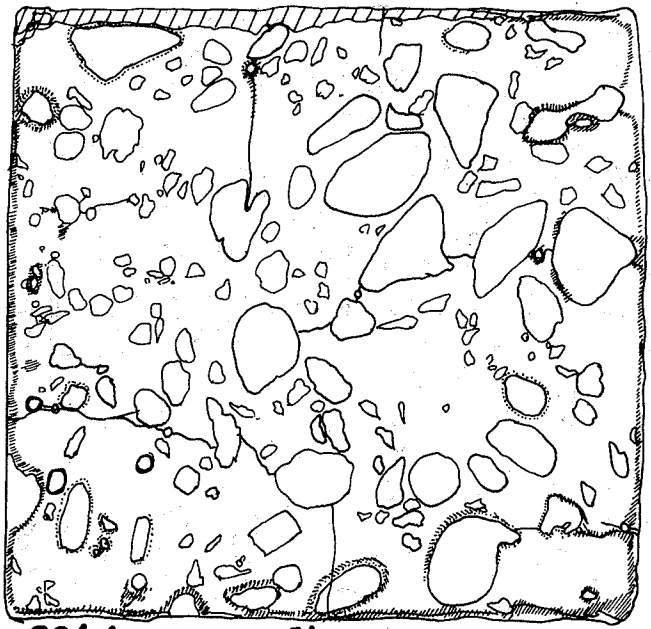
F8/1/2 Fig. 4

F8/1/1 Fig. 4

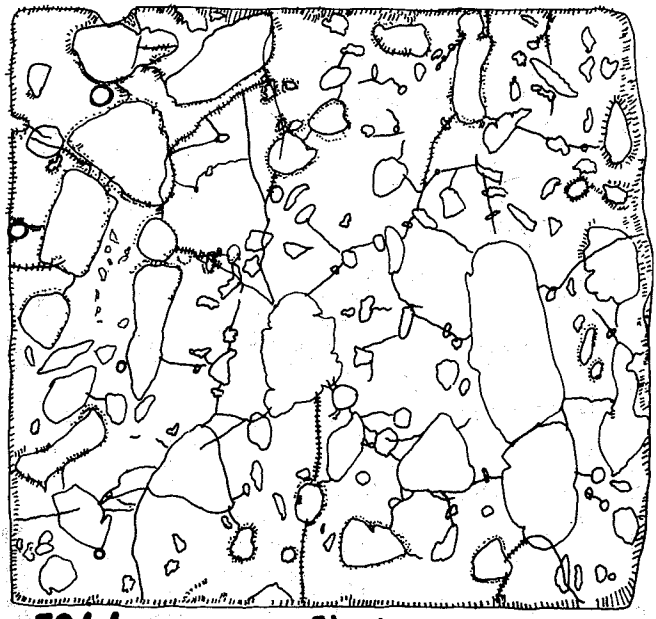
Fig.4
FB1/1/4

Fig.4
FB1/1/3

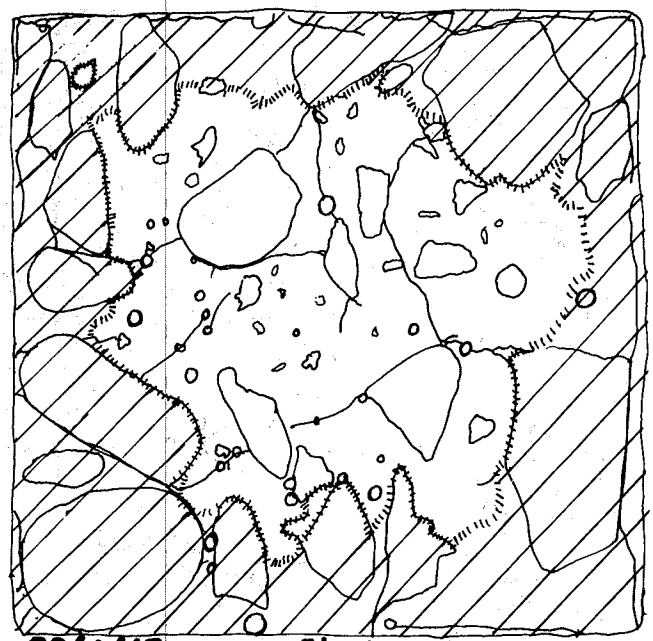
F81/1/6 Fig. 4



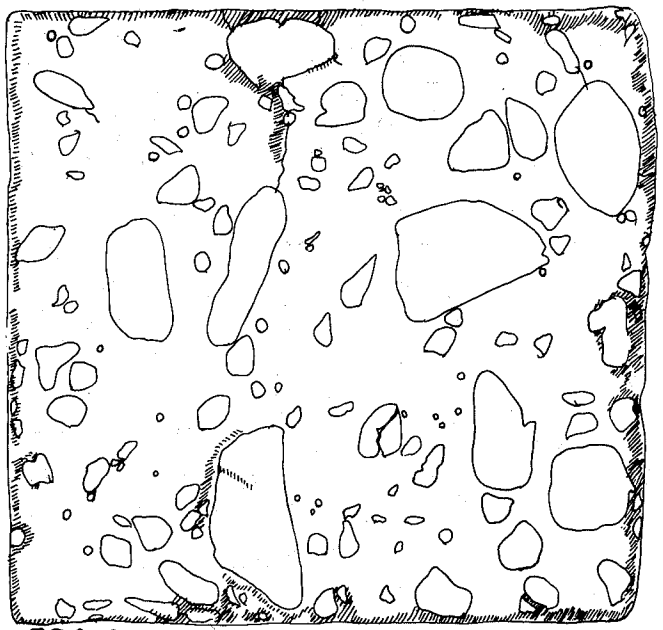
F81/1/5 Fig. 4



F81/17 Fig. 4



F8/1/12 Fig. 5



F8/1/13 Fig. 5

F10/9 Fig.6

F10/4 Fig.6