

Limestone Selection For
Permeable Plug Mine Seals

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INTRODUCTION

Research on new mine sealing techniques has indicated that mine openings discharging acid mine drainage can be sealed with crushed limestone¹, an inexpensive material widely used for neutralizing mine drainage. This type of seal is constructed by filling a length of the mine opening with limestone aggregate. Since the resulting seals are porous due to the granular nature of the stone fill, they have been called permeable plugs.

Mine drainage initially seeping through the aggregate is neutralized by the limestone particles as it passes through the plug. Ferric hydroxide and possibly calcium sulfate are precipitated within the plug and fill the voids between the stone particles, decreasing the permeability of the seal. This self-sealing property of the plug eventually eliminates or at least greatly reduces the discharge of mine water. Further, mine water which is discharged has been neutralized and filtered by the limestone aggregate. Mine water treatment by the seal is of secondary importance, however, as continued percolation and treatment of mine water would gradually result in chemical consumption of the plug, leading eventually to mechanical failure of the seal.

A variety of limestone aggregates, differing in both chemical and physical properties, are available for use in constructing permeable plugs. Previous research on the neutralization of acid mine drainage with limestone has shown that limestones from various sources can be classified into three groups according to their neutralization characteristics². These three groups were called Type A, Type B, and Type C limestones. Type A limestones neutralized mine drainage most effectively, while Type C limestones neutralized mine drainage least effectively.

Crushed limestone can also be categorized according to the sizes of the particles making up the aggregate. A number of grades of limestone having different particle size ranges are commercially available, and still others can be obtained by custom screening or blending. Although many of the commercial limestone grades specify the distribution of particle sizes (gradation) as well as the size range, different gradations within a single size range are sometimes available or can be obtained by custom blending.

To determine the behaviors of various limestone aggregate types and sizes when subjected to mine water percolation, numerous aggregate specimens were tested in a study sponsored by the U.S. Environmental Protection Agency under Contract No. 68-01-01353. The intent of the study was to identify which types of limestone aggregate would be most suitable for use in permeable plugs. All testing was performed on a pilot scale in the laboratory by continuously supplying synthetic mine water to aggregate specimens placed in six inch square by six foot

long horizontal vessels for periods of up to 100 days. During the test period, the flow rates of water percolating through the model plugs and the compositions of the effluents were monitored. This paper presents a summary of the findings and conclusions of the study.

EXPERIMENTAL METHOD

Testing equipment was developed which simultaneously produced three synthetic mine waters differing only in the relative concentrations of ferric and ferrous iron and supplied each type of test water to a battery of up to 24 test vessels containing the limestone specimens. Each vessel was supplied with test water at the rate of 1 gpm, and the water head at the inlet end of the vessel was limited to 72 inches by diverting any excess water back to the supply system. Water percolating through the specimens was allowed to discharge freely from the outlet end of the vessels. The system was sized to produce 25 gpm of each type of test water, permitting a total of 72 tests to be run at one time. An illustration of the test apparatus showing tests in progress is presented in Figure 1.

The three test waters were produced by an automatic blending system from tap water and concentrated solutions of seven technical-grade chemicals. Tap water and four of the concentrates (manganese sulfate, magnesium sulfate, aluminum sulfate, and lime) were first blended to form an iron-free base stock containing the desired concentrations of manganese, magnesium, aluminum, and calcium. Sulfuric acid and ferric sulfate concentrates were added to a portion of the base stock, producing ferrous test water. Equal amounts of these two test waters were blended to form ferric/ferrous test water. The average compositions of these waters are listed in Table 1.

TABLE 1

AVERAGE COMPOSITION OF SYNTHETIC ACID MINE WATERS

	<u>Ferric Water</u>	<u>Ferric/Ferrous Water</u>	<u>Ferrous Water</u>
pH	2.5	2.6	2.5
Sp. Conductance (μmho)	2700	2700	2850
Hot Pht. Acidity	743	894	874
Calcium	81	78	75
Magnesium	28	22	23
Manganese	5.3	5.8	5.0
Aluminum	16	16	18
Total Iron	205	209	198
Ferrous Iron	10	106	197
Ferric Iron	195	103	1
Sulfate	1055	1030	1122

NOTE: Constituent concentrations reported in mg/l

Each of the test waters was pumped from its supply tank to a manifold assembly serving a battery of test vessels. Overflow tees at each end of the supply manifold maintained a constant water pressure in the manifold, returning excess test water to the appropriate supply tank. Test water flowed from the supply manifold to a standpipe attached to the inlet end of each test vessel through a length of tubing calibrated to deliver a constant flow of 1 gpm. The test water head was limited to 72 inches by an overflow tee at the top of the standpipe which diverted any excess test water back to the supply tank. Thus a

maximum feed rate of 1 gpm and a maximum head of 72 inches were independently provided.

The test vessels containing the stone specimens were essentially square plexiglas tubes lined with PVC film, and were assembled from separate components. After first assembling the vessel bottom, sides, lines, and ends, limestone aggregate was placed in the open-topped, trough-like structure. The liner was then folded over the stone specimen and sealed and the top was secured, completing vessel assembly. Before test water flow was initiated through the stone, the outside of the PVC lines was pressurized with compressed air at about 5 psig to prevent water from channeling along the sides and top of the stone plug.

DISCUSSION

The study was conducted in two sequential laboratory cycles, Lab Cycle I and Lab Cycle II. In Lab Cycle I, three sets of 24 different limestone specimens were tested on ferric, ferrous, and ferric/ferrous synthetic mine waters for a total of 72 tests. The results of these tests were used to select the specimens tested in Lab Cycle II.

Limestones from three different sources were tested in Lab Cycle I. Selection of the limestones was based on a limestone neutralization study² which showed that limestones can be classified as Type A, Type B, or Type C according to their neutralization characteristics. One limestone from each group was tested, limestone No. 1809 (Type A), limestone No. 1355 (Type B), and limestone No. 1337 (Type C). The stones were obtained from Winfield Lime and Stone Company, Elkins Limestone Company, and Mineral Pigments and Metals Company, respectively.

The following six size fractions of each type of stone were tested:

- 1" to dust (called 1 x 0)
- 1" to 50 mesh (called 1 x 50m)
- 1/2" to dust (called 1/2 x 0)
- 1/2" to 50 mesh (called 1/2 x 50m)
- 1/4" to dust (called 1/4 x 0)
- 1/8" to dust (called 1/8 x 0)

All six size fractions were prepared by screening a blend of equal weights of three commercially available grades of crushed limestone. A blend was used rather than one standard grade because no single grade contained the entire range of one inch particles to dust.

In addition to these 18 different specimens (six sizes of each of three types of stone), three additives were investigated by testing limestone mixtures containing 5% bentonite, 10% flyash, and 10% air-cooled blast furnace slag by weight. The additives were blended with both the 1 x 0 and the 1/2 x 0 sizes of limestone No. 1809. Thus a total of six different specimens containing additives were also tested.

Three test vessels were loosely filled with each of the 24 different limestone aggregates, and the resulting three sets of specimens were tested on the ferric, ferric/ferrous, and ferrous test waters. Inlet heads on all the 1/8 x 0 and 1/4 x 0 size specimens quickly rose to the maximum 72 inches after start-up. Initial inlet heads on the coarser sized specimens were as low as six inches. Flow rates after one day of testing ranged from 15 ml/min to the maximum 1 gpm (3785 ml/min). After 20 days, the testing of specimens with flows in excess

of 0.5 gpm (1892 ml/min) at a 72 inch head was discontinued. Testing of the remaining specimens was continued for an additional 33 days for a total of 53 days of testing. Three of the specimens were tested for a total of 101 days.

Flow histories for the limestone specimens which had flow rates of 300 ml/min or less are presented for the three sets of tests in Figures 2, 3, and 4. Although all flow histories showed a considerable fluctuation, the fluctuation was least severe for specimens tested on ferric water and most severe for specimens tested on ferrous water. Specimens tested on ferric or on ferric/ferrous water generally had lower flow rates than specimens tested on ferrous water.

Initial flow rates were found to be lowest for stone No. 1337 (Type C) and highest for stone No. 1809 (Type A). Physical analysis of representative samples of the limestones before testing showed that stone No. 1337 consistently contained a greater fraction of fines (particles smaller than 200 mesh) than the other two types of stone and that stone No. 1809 generally contained the least amount of fines, particularly for the $1/8 \times 0$ and $1/4 \times 0$ sizes. Correlation of initial flow rates and fines contents indicated that the initial flow rates were dependent on the fines contents of the aggregates. Thus the flow behaviors observed during the initial portion of the test run were due to physical rather than chemical differences in the stone types.

Throughout the test run of 53 days, stone No. 1809 exhibited the greatest reductions of flow, while stone No. 1337 exhibited the smallest reductions. As a result, flow rates after 50 days of testing for stone No. 1809 specimens subjected to ferric or ferric/ferrous water were generally lower than for corresponding stone No. 1337 specimens. Flow rates for stone No. 1355 specimens tested on ferric or ferric/ferrous water were generally greater than flows for stone No. 1809 specimens and lower than flows for stone No. 1337 specimens after 50 days of testing. Even after 50 days of testing on ferrous water, flow rates for stone No. 1809 and stone No. 1355 specimens did not decrease to less than the flow rates for corresponding stone No. 1337 specimens, probably due to a greater importance of fines content for tests performed on ferrous water.

The lowest flow rates were observed for the $1/8 \times 0$ and $1/4 \times 0$ sizes of stone, while the highest flow rates were observed for the $1/2 \times 50\text{m}$ and $1 \times 50\text{m}$ sizes. All the $1/8 \times 0$ sizes and all but one of the $1/4 \times 0$ sizes maintained flow rates of less than 300 ml/min throughout the 53 day test period. Flow histories for these two sizes were similar, although the $1/8 \times 0$ size generally exhibited lower initial flow rates. Flow rates for the $1/2 \times 0$ sizes tested on ferric or ferric/ferrous water were only slightly greater than those for the $1/8 \times 0$ and $1/4 \times 0$ sizes. Most of the $1/2 \times 50\text{m}$ and $1 \times 50\text{m}$ sizes were discontinued after 20 days of testing due to their high flow rates. The significantly poorer flow behaviors of aggregates having a 50 mesh lower size limit again demonstrated the importance of fines content.

Specimens which had lower flow rates neutralized mine water percolating through the stone more effectively than those with higher flow rates. Also, the pH of the effluents tended to increase slightly throughout the tests as the observed flow rates decreased. Effluents from specimens with flow rates less than 300 ml/min typically had pH values of 6 or 7, while little or no increase in pH was observed for specimens with flow rates approaching 1 gpm.

Chemical compositions of the neutralized effluents indicated that iron was precipitated and retained within the stone aggregate, but that calcium sulfate was not. Ferric iron concentrations were typically less than 20 mg/l (a 90% removal for ferric water tests) and were often less than 0.03 mg/l. Ferrous iron was also removed in many cases, but not as effectively as ferric iron. Calcium concentrations were significantly higher in the effluents than in the influents (probably due to chemical stone consumption and erosion), and sulfate concentrations were essentially unchanged. It should be pointed out that calcium sulfate precipitation would be expected for mine waters having sulfate concentrations substantially higher than the 1000 to 1100 mg/l in the synthetic test waters.

Flyash and bentonite additives were shown to improve the performances of 1/2 x 0 and 1 x 0 stone sizes. Use of these additives, particularly flyash, resulted in lower flow rates and more effective water treatment than the same size stone without additives. Performances of specimens containing these additives were comparable to the performances of the 1/8 x 0 and 1/4 x 0 sizes without additives in tests using ferric and ferric/ferrous waters. Both the 1 x 0 and 1/2 x 0 sizes containing flyash were more successful than smaller sizes without additives in ferrous water tests.

After completion of the test runs, the tops of the test vessels were removed and the stone specimens were examined. A decrease in the heights and widths of the stone plugs, illustrated in Figure 5, was observed for all specimens. Since the inlet ends of the vessels were most severely affected, it is believed that stone consumption by neutralization reactions was a major cause of the volume losses. Settling of the stone upon being wetted, hydraulic erosion, and compression by the pressurizing air outside the PVC film may also have contributed to the volume losses.

The overall volume loss was determined for each specimen and ranged from 3% to 39%. The following general trends were observed:

- Volume losses were smallest for specimens tested on ferric water and largest for specimens tested on ferrous water.
- Limestone No. 1809 specimens had the smallest volume losses and limestone No. 1337 specimens had the largest volume losses.
- The intermediate size specimens, 1/4 x 0 and 1/2 x 0, exhibited the least severe losses.
- Bentonite and slag additives effectively inhibited volume losses.

Physical properties were determined for eleven of the specimens selected to represent a cross-section of the stone types, aggregate sizes, and test waters. None of the specimens were rigidly cemented, although some small blocks of lightly cemented material were observed within six to twelve inches of the inlet ends of several specimens. Density, compressibility, and shear strength analyses were performed on vertical cylinders trimmed from the first twenty inches of the inlet ends of the specimens.

All specimens were found to have low densities characteristic of loose to very loose materials. The densities of six of the eleven specimens were even less than the minimum densities obtainable by very loose placement of dry material (negative relative densities), indicating particle structures considerably looser and more fragile

than those which could be produced by physical placement. Apparently the particle structures of at least these six specimens were weakened by exposure to the mine water.

Compressibilities and shear strength parameters of the specimens were found to be primarily a function of the specimen densities. Further, both stiffness and shear strength were shown to be considerably improved by compacting the specimens. It was concluded that none of the limestones tested had inherently superior physical characteristics due solely to stone type or size.

Lab Cycle II was then conducted to further investigate the effects of gradation and placement densities on plug performances and to investigate several additives which might aid in cementing the stone particles. A total of twelve specimens were prepared using the commercially available 3/8" to dust (called 3/8 x 0) grade of limestone No. 1809. Ten of these specimens were tested for 50 days on ferric/ferrous water, and the two remaining specimens were tested for 50 days on tap water.

Four specimens containing increased quantities of fines were tested on ferric/ferrous water. Minus 50 mesh fines obtained by screening a portion of the raw 3/8 inch to dust stone were blended with 3/8 x 0 stone in sufficient quantities to increase the fraction of material passing a No. 200 sieve by factors of two and three. Each of the resulting mixtures was placed at both 30% and 60% relative densities for a total of four specimens.

Four specimens of "natural" 3/8 x 0 stone as received from the quarry were tested. Two of the specimens were placed at about 30% and 60% relative densities and tested on ferric/ferrous water. The other two specimens were placed at about 0% and 30% relative densities and tested on South Pittsburgh City Water.

Four specimens containing additives were tested on ferric/ferrous water. Portland cement, calcium sulfate hemihydrate (plaster of Paris), and sodium silicate were blended with 3/8 x 0 stone in 5% concentrations to prepare three of the specimens. A "zoned" plug containing 5% ferric sulfate and 15% sodium silicate in the first foot of the plug only (intended to be a water pretreatment zone) was also prepared. All four specimens were placed at a relative density of about 30%.

Flow and effluent composition data collected during the 50 day test run showed that all ten specimens subjected to ferric/ferrous water effectively obstructed water percolation and treated that water which did pass through the plugs. Both flow and neutralization behaviors were generally more satisfactory than those observed during Lab Cycle I.

Flow histories for specimens tested on ferric/ferrous water are presented in Figure 6. Flow rates for one specimen (3X fines, 60% relative density) were not plotted, as they were always less than 0.5 ml/min. The flow histories show that both increasing the fines content and increasing the placement density of the stone resulted in significantly lower flow rates. Specimens containing additives also successfully prevented mine water flow, although less effectively. Flow rates for the two specimens tested on tap water decreased over the test period, but were consistently greater than flow rates for specimens tested on synthetic mine water. These results indicate that physical effects of percolation contributed to the observed plugging of the specimens.

Overall volume losses were determined for the ten specimens tested on ferric/ferrous water at the completion of the test run. The losses ranged from 2% to 20% and showed the following trends:

- Volume losses were greater for specimens having larger quantities of fines.
- Higher placement densities resulted in lower volume losses.
- Volume losses for specimens containing additives were comparable to losses for corresponding natural stone specimens.

Densities of the specimens were then measured and showed that both increasing the initial placement density and increasing the fines content of the stone resulted in improved densities after testing. The densities of the two natural stone specimens, however, were found to be significantly less than initial placement densities. The specimens containing sodium silicate exhibited an appreciably negative relative density, suggesting the possibility of severe erosion and particle structure weakening.

Analysis of the specimen compressibilities and shear strength parameters again showed the strength of the plugs to be primarily dependent on density. The additives tested did not result in improved cementation and neither increased the stiffness nor increased the shear strength of the stone. Consequently, the additives were shown to be ineffective.

CONCLUSIONS

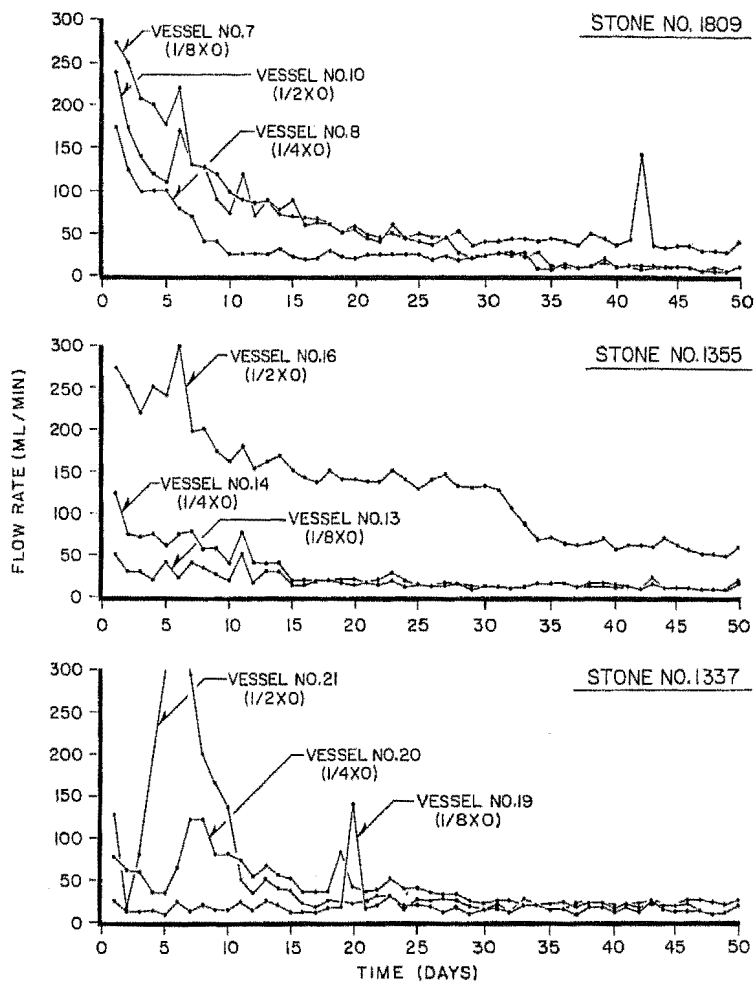
1. The results of the study indicate that limestone aggregate plugs are a feasible means of sealing underground mines which discharge water containing ferric iron.
2. The 3/8 inch to dust size of limestone No. 1809 placed at 60% relative density was the most satisfactory natural material tested.
3. The Type A limestone (found in previous tests to neutralize acid mine waters better than Types B and C limestones) had the best overall performance, while the Type C limestone had the poorest performance.
4. The 3/8 inch to dust grade of stone was the most satisfactory size tested.
5. Increasing the fines content of commercially available stone to twice the original amount (as determined by the fraction of material which passes a No. 200 sieve) results in improved performance.
6. High placement densities are essential for satisfactory plug performance.

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3. Penrose, R. G., Jr., and Holubec, I., "Laboratory Study of Self-Sealing Limestone Plugs for Mine Openings", Environmental Protection Agency Report No. EPA-670/2-73-081.

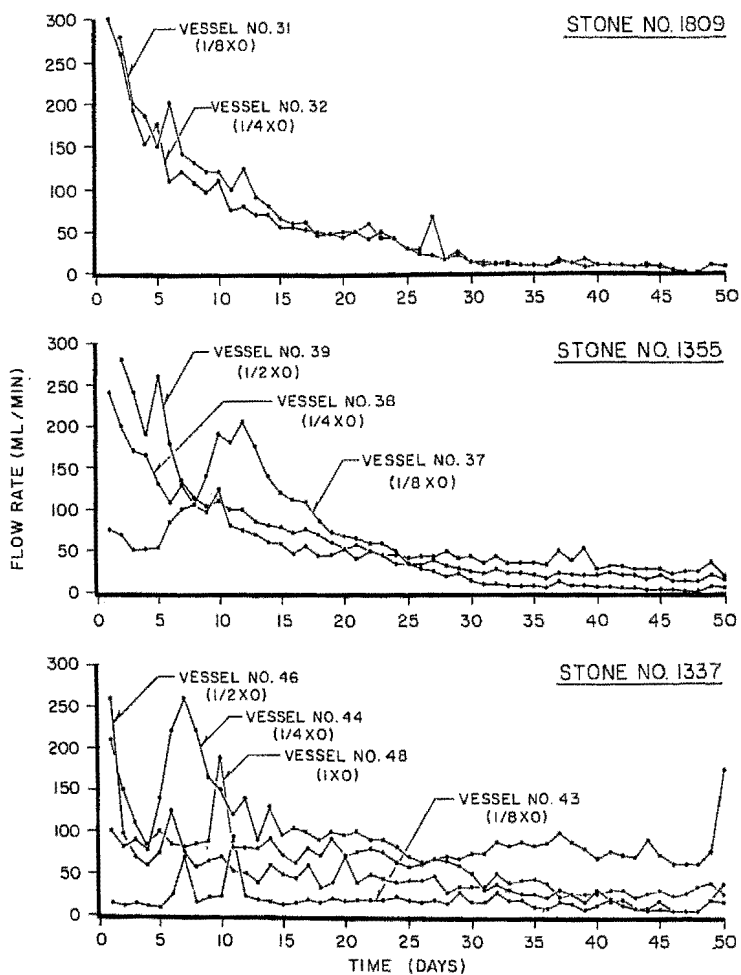


LAB CYCLE I TESTING
FIGURE 1



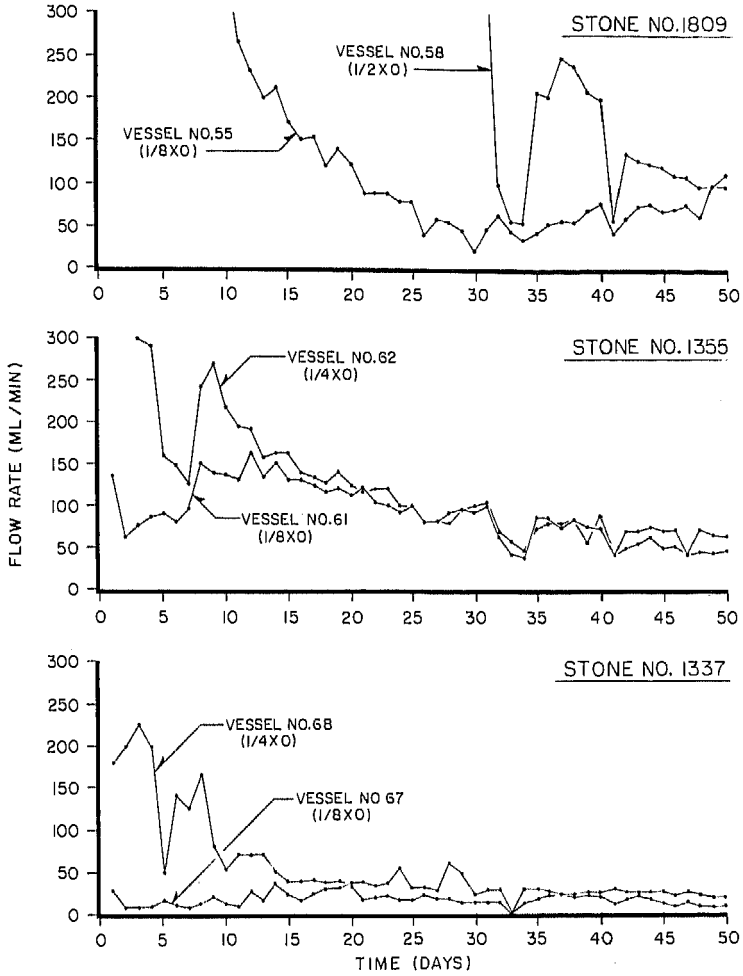
FERRIC WATER-SPECIMEN FLOW HISTORIES

FIGURE 2



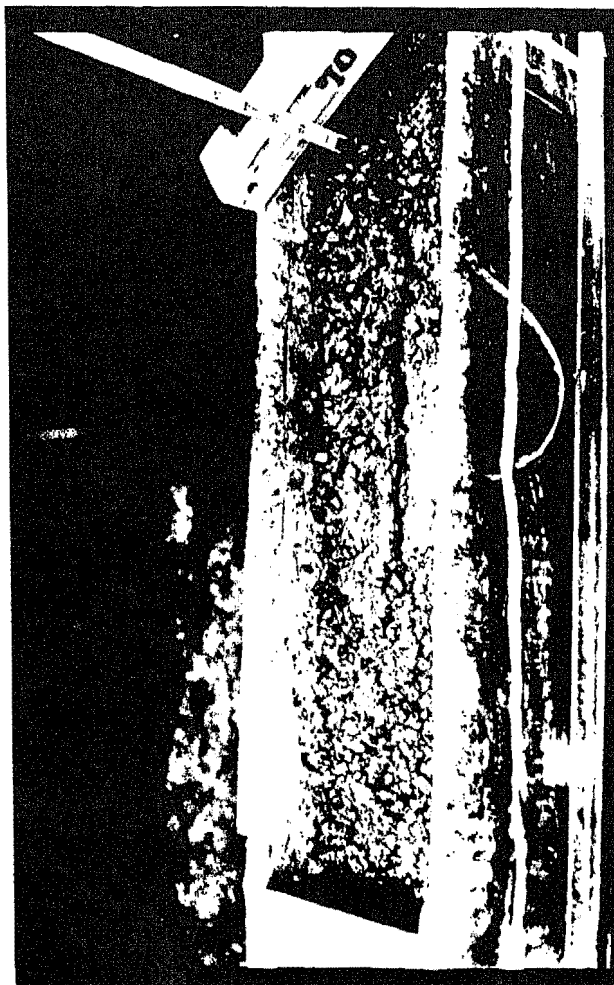
FERRIC/FERROUS WATER-SPECIMEN FLOW HISTORIES

FIGURE 3

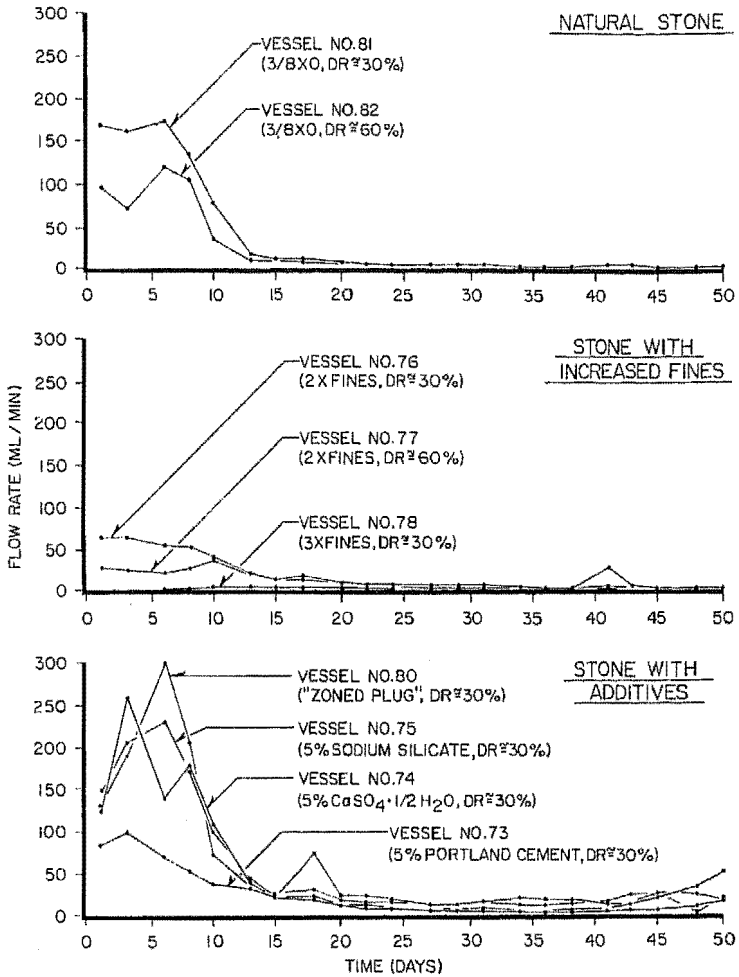


FERROUS WATER-SPECIMEN FLOW HISTORIES

FIGURE 4



TYPICAL SPECIMEN AFTER TESTING
FIGURE 5



LAB CYCLE II-SPECIMEN FLOW HISTORIES

FIGURE 6