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Volume Expansion of Acidproof Brick Exposed to 20 Wt Pct HCl at 90° C

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UNITED STATES DEPARTMENT OF THE INTERIOR

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UNIT OF MEASURE ABBREVIATIONS USED IN THIS REPORT

°C	degree Celsius	pct	percent
g/cm^3	gram per cubic centimeter	psi	pound per square inch
h	hour	psig	pound per square inch, gauge
in	inch	wt pct	weight percent
L	liter	yr	year
μm	micrometer		

VOLUME EXPANSION OF ACIDPROOF BRICK EXPOSED TO 20 WT PCT HCl AT 90° C

By James P. Bennett[1] and Timothy A. Clancy[2]

ABSTRACT

In this Bureau of Mines study, portions of four acidproof bricks (two red shale and two fireclay) were exposed to 20 wt pct HCl at 90° C for periods of 30 days. Three different techniques were selected to determine volume expansion of these materials after exposure. Linear expansions ranging from about 0.02 to 0.12 pct were noted from Ta pin and dilatometer measurements. The use of strain gages to measure expansions was not successful. The red shale and fireclay brick with the lowest apparent porosities had the lowest expansions.

Expansion data obtained by direct measurement of samples exposed for 30-day periods or by dilatometric measurement of autoclaved samples exposed for 2 h were comparable. This suggests the use of an accelerated autoclave exposure procedure for determining volume expansion effects of ceramic materials in acid environments.

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INTRODUCTION

New mineral processing techniques being investigated by the Bureau of Mines and others, such as processing at elevated temperatures and pressures, leaching with acids and bases, chloride leaching, and dissolution in fused-salt baths, require the use of construction materials that have good corrosion resistance. One example is the construction material needed to line leaching vessels used in the extraction of alumina from clay using HCl (hydrochloric acid), HNO_3 (nitric acid), or H_2SO_4 (sulfuric acid). In the HCl extraction processes studied by the Bureau, acid concentrations range from 1 to 35 pct (1)[3] with temperatures from 50° to 900° C (2).

Industrial equipment designers, fabricators, and material suppliers of acid processing systems were contacted for their recommendations on ceramic construction materials to be used in HCl environments. Although the suppliers recommended acidproof brick, carbon brick, or dense firebrick, they advised that materials be tested in an environment simulating actual conditions because of limited experience with HCl processing and the uniqueness of each industrial process. In general, the suppliers could say only that their brick would pass ASTM-C279 (3) specifications, which deal with H_2SO_4 rather than HCl.

Previous Bureau investigations into the effect of various acids on ceramic materials (4-6) showed that certain physical property changes occurred, one of which was volume expansion. Inconsistencies in data trends, however, indicated that a closer examination of this phenomenon was warranted. It was necessary to identify a testing technique that would result in accurate and reproducible data.

Volume expansion, also known as irreversible growth or swelling, causes a dimensional increase in acidproof brick and is generally thought to be similar to moisture expansion observed in structural clay products.

Floor, wall, and structural tiles, building brick, or glazed materials have exhibited volume expansion, which can occur after a few months or time periods of 50 yr or more. The chemical adsorption of water vapor on the internal amorphous silicate surfaces or the internal relaxation caused by adsorbed water molecules are two proposed causes for this expansion (7). In these bodies, sodium and potassium oxides are known to aggravate expansion while alkaline earth oxides are reported to reduce it through the formation of crystalline material or a more chemically resistant glass phase.

Linear expansion of up to 0.35 pct in acidproof brick has been documented (8), but no published technical information or standard test exists to measure expansion or the influence of such factors as temperature, pressure, or exposure environment. Methods have been proposed to measure the swelling potential of structural clay products, which include the application of steam, pressure, and elevated temperature (9-10) to materials. Actual expansion that occurs in structural clay under industrial use conditions, however, is normally less than that predicted from laboratory studies (11).

Expansion of structural units is a critical factor in vessel design, because improper design or construction may cause acid brick to separate from the impermeable membrane backing, resulting in possible leakage or rupture to the system.

This report presents expansion results obtained using three different methods: (1) physical measurement of fixed points on a sample, (2) strain gage sensors, and (3) a thermal expansion technique. Two red shale and two fireclay brick samples typical of acidproof brick used as chemical-resistant structural material were studied. The materials were exposed to various temperature and pressure conditions in deionized water and 20 wt pct HCl. The water test environment served as a reference condition.

[3]Underlined numbers in parentheses refer to items in the list of references at the end of this report.

TEST EQUIPMENT AND SAMPLE DESCRIPTION

Chemical and physical properties of the four different acidproof brick tested for linear expansion are listed in table 1. Results of earlier tests (5) on the effect of exposure to 20 wt pct HCl at 90° C for 110 days on strength and weight loss properties are also shown in table 1. Statistically significant changes in weight loss occurred in all samples. A statistically significant strength loss was observed only in fireclay brick A. The total ions leached from the brick into solution are the sum of the weight percents of Al, Ca, Fe, K, Mg, Na, Si, and Ti ions. In general, the Fe and Al ions had the highest removal rate, and Ca, Mg, Na, K, and Ti removal rates were minor. No Si ion was removed from any brick.

One method used to monitor length changes in this study was to measure the distance between tantalum (Ta) pins cemented to opposite ends of a 7-1/4- by 1-7/8- by 1-1/8-in sample, cut from full-size production brick. Two Ta pins, 1/4 in long and 3/16 in. in diam, with one end rounded to a 3/16-in-diam hemisphere, were attached opposite each other at the center of both ends of the 7-1/4-in dimension of the sample using a chemical-resistant epoxy, as shown in figure 1. Sample length was measured at 25° C at the start and end of the 30-day test period, with a micrometer accurate to 1.0×10^{-4} in.

Another method of measuring expansion consisted of attaching strain gages to the center of the 7-1/4- by 1-7/8-in face surface of the samples using a chemical-resistant epoxy (fig. 1). Lead wires coated with Teflon[4] fluorocarbon polymer were attached to the strain gages, and a protective barrier coat system was applied over them. Strain gage monitoring was done on an intermittent basis during the 30 days.

[4]Reference to specific products does not imply endorsement by the Bureau of Mines.

TABLE 1. - Chemical and physical properties of commercial acidproof brick

	Red shale brick		Fireclay brick	
	A	B	A	B
BEFORE EXPOSURE				
Chemical composition, wt pct:				
SiO_2	64.6	63.3	59.4	68.6
Al_2O_3	20.6	20.7	31.7	22.9
Fe_2O_3	6.4	5.9	2.0	1.9
K_2O	3.6	4.6	3.0	1.5
TiO_2	1.6	1.6	1.7	1.4
MgO	0.99	1.3	0.35	0.56
Na_2O	0.58	0.57	0.27	0.27
CaO	0.39	0.05	0.03	0.06
Apparent porosity........pct..	10.86	3.26	5.66	11.06
Bulk density...........g/cm^3..	2.39	2.56	2.38	2.26
Cold crushing strength...psi..	18,800	20,300	9,800	9,700
AFTER EXPOSURE[1]				
Weight change............pct..	-6.13±0.33*	-0.95±0.32*	-0.90±0.07*	-6.57±0.32*
Cold crushing strength...psi..	19,300±1,500	21,600±1,900	8,100±1,200*	8,800±920
Total ion leached.....wt pct..	3.57	1.31	0.48	3.14

[1]To 20 wt pct HCl at 90° C for 110 days (5).
*Statistically significant change at 95-pct confidence level, compared with data for untreated specimen (Student's t-test).

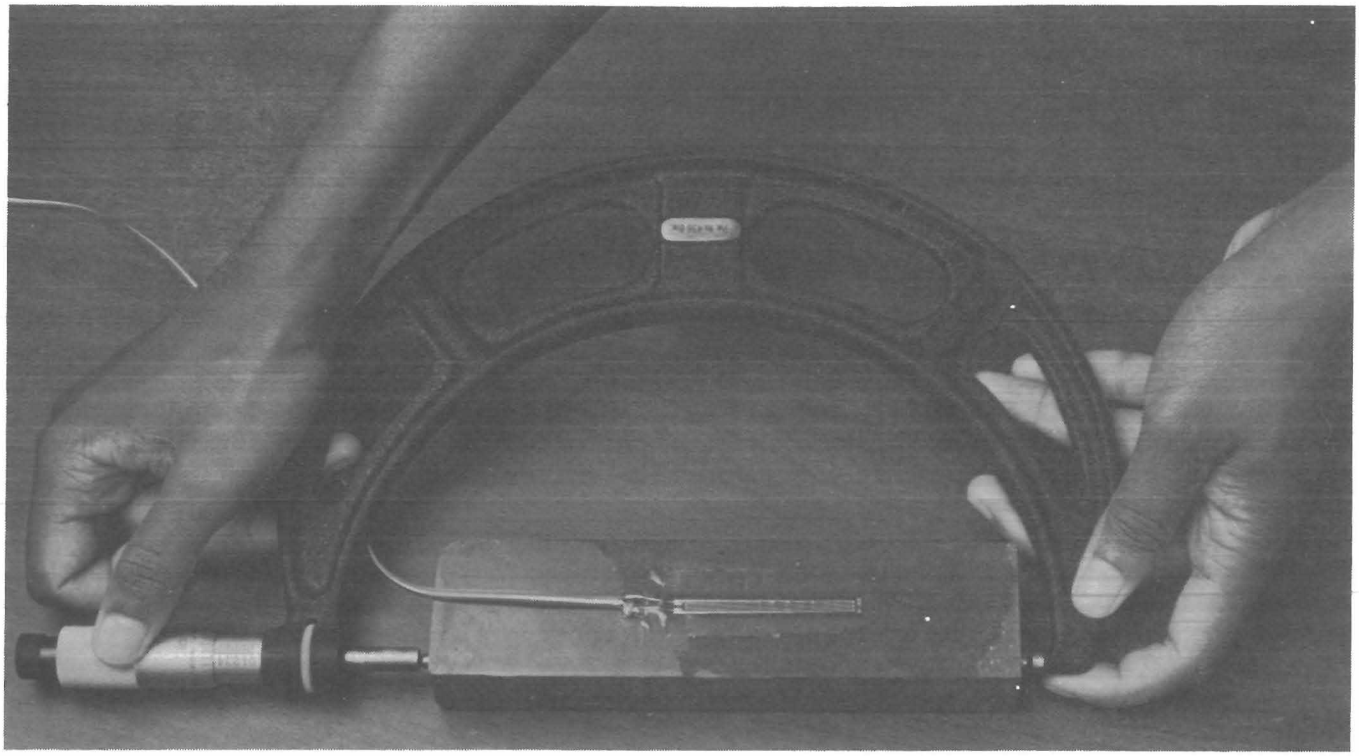

FIGURE 1. - Measurement of linear distance between Ta pins attached to brick sample. (Strain gage is attached to center of sample.)

For the third method, thermal expansion measurements, test samples 3/16 in. in diam were cored from brick specimens and cut to a final size of 1/2 in long. The samples were cored in the same direction as the 7-1/4-in sample length used for Ta pin and strain gage measurements. After exposure, cored samples were heated in a dilatometer to 700° C and sample length changes were monitored.

The 7-1/4- by 1-7/8- by 1-1/8-in brick samples with Ta pins and strain gages attached and the 3/16-in-diam, 1/2-in-long cored cylinders were exposed to 20 wt pct HCl or deionized water in 12-L spherical glass flasks enclosed in heating mantles. Temperature was controlled by a variable power source and monitored by type K thermocouples. The test environment of 20 wt pct HCl at 90° C had been found to be the most corrosive in an earlier study (5). Five bars and five cylinders of each brick type were exposed simultaneously. A condenser refluxed any vapor back into the system. Samples remained submerged in either 20 wt pct HCl or deionized water for 30 days.

Thermal expansion curves of autoclaved samples were used as an accelerated means of determining the amount of linear expansion of cored samples. The cored samples, 3/16 in. in diam by 1/2 in long, were exposed to 20 wt pct HCl and to deionized water in an autoclave for 2-h periods at temperatures of 150°, 200°, and 250° C, at pressures of 80, 220, and 580 psig, respectively. After exposure, cored samples were heated in the dilatometer to 700° C and cooled back to room temperature while the sample length was monitored.

Samples of each brick exposed to the 580-psig, HCl conditions were also examined for postexposure weight loss in a thermogravimetric analysis (TGA) apparatus. A small section of each exposed brick was heated to 960° C and the sample weight monitored during the heating.

RESULTS AND DISCUSSION

The use of strain gages to monitor linear changes was not successful. Apparently, chemical leakage through the epoxy base coat and/or through the lead wires resulted in erratic data after about 5 days exposure.

Linear changes based on Ta pin measurements as well as a dilatometer runs are listed in table 2. Statistically significant[5] changes occurred for all samples except red shale brick B in the water environment and for all samples exposed to the acid environment. Red shale brick A had the largest expansion of any sample, ranging from 0.035 pct in deionized water to a high of 0.115 pct in 20 wt pct HCl. Fireclay brick B had the larger expansion of the two fireclay brick in both water (0.033 pct) and acid (0.073 pct). Red shale brick A and fireclay brick B, the two samples with the largest linear expansion in water and acid, are also those bricks with the highest porosity, as shown in table 1. In all cases, expansion occurring in HCl was greater than that observed in a water environment under similar conditions of temperature and time.

TABLE 2. - Linear expansion of acidproof materials after 30-day exposures in 20 wt pct HCl or water at 90° C, percent

Brick	Ta pin	Dilatometer[1]
WATER		
Red shale:		
A.........	0.035±0.009*	0.035
B.........	-.003± .011	.000
Fireclay:		
A.........	.018± .009*	.026
B.........	.033± .014*	.039
20 WT PCT HCl		
Red shale:		
A.........	0.115±0.030*	0.060
B.........	.044± .011*	.019
Fireclay:		
A.........	.049± .021*	.031
B.........	.073± .036*	.056

[1]Single data point.
*Statistically significant change at 95-pct confidence level compared to untreated specimen (Student's t-test).

Dilatometric runs were made on autoclaved samples as well as samples exposed at ambient pressure.

The dimensional changes determined by dilatometric runs on autoclaved samples are shown in table 3. A typical dilatometric curve for a red shale sample exposed at ambient pressure is shown in figure 2. Heating the sample to 700° C removed the moisture expansion, and subsequent cooling then returned the sample to its original, or preexposed, length. Therefore, the difference shown as Δ in figure 2 associated with the dilatometer heating is equivalent to the expansion produced by autoclaving. The data indicate that the expansions associated with the HCl autoclave exposures are generally larger than expansions associated with water autoclaving exposures. The same relationship holds for the dilatometric data for samples exposed to acid and to

[5]Statistically significant changes (Student's t-test) at a 95-pct confidence level, compared with data for untreated specimens (12).

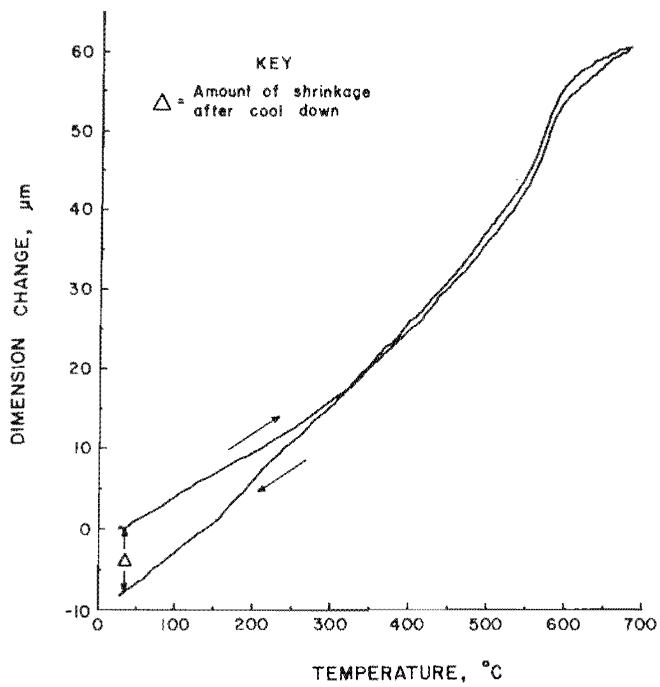

FIGURE 2. - Dilatometric curve of red shale A brick sample exposed to 20 wt pct HCl for 30 days at 90° C.

water for 30 days at atmospheric pressure as well as for the data obtained from the Ta pin measurements (table 2). Also, the exposure of samples of a particular brick to higher autoclaving pressures resulted in greater expansions. The expansions associated with autoclaving at the lower two pressures, 80 and 220 psig, coincide better with expansion data obtained from the Ta pin measurements than those at 580 psig. However, expansions associated with the 580-psig autoclaving were generally larger. These results indicate that the dilatometric measurement of samples after autoclaving in acid environments could be a valid accelerated test to determine the expansion effects of various acids on different ceramic materials.

Thermogravimetric analysis curves (a typical curve is shown in figure 3) for a sample of each of the four bricks exposed to 20 wt pct HCl at 580 psig indicated that these samples had gained some weight during the autoclave exposure. Measured as weight loss, the results were as follows: red shale A, 0.35 wt pct; red shale B, 0.20 wt pct; fireclay A, 0.50 wt pct; and fireclay B, 0.35 wt pct.

TABLE 3. - Linear expansion of acidproof materials exposed for 2 h to autoclave conditions, percent

Brick	80 psi	220 psi	580 psi
WATER			
Red shale:			
A............	0.045	0.047	0.077
B............	.000	.013	.000
Fireclay:			
A............	.024	.019	.027
B............	.031	.048	.103
20 WT PCT HCl			
Red shale:			
A............	0.048	0.053	0.161
B............	.044	.040	.054
Fireclay:			
A............	.025	.023	.134
B............	.058	.083	.167

NOTE.--Single data points for all values.

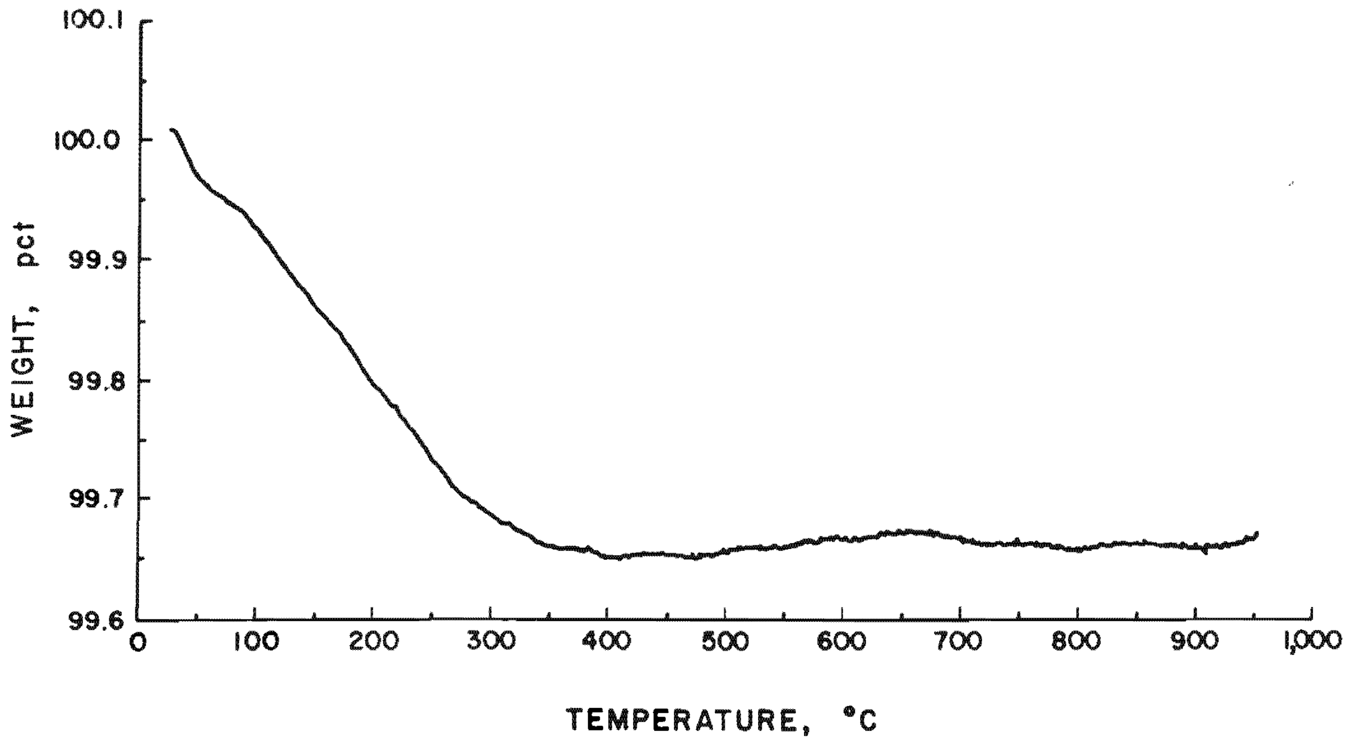

FIGURE 3. - Thermogravimetric curve of red shale A brick sample exposed to 20 wt pct HCl for 2 h at 580 psi and 250° C.

CONCLUSIONS

Based upon the study of the volume expansion of acidproof brick exposed to 20 wt pct HCl at 90° C, the following conclusions can be made:

1. Acidproof brick samples exposed to 20 wt pct HCl at 90° C for 30 days showed linear expansions ranging from about 0.02 to 0.12 pct. The red shale and fireclay brick that had the lowest apparent porosity had the lowest percent expansions.

2. Expansion data from direct measurement of samples exposed for 30-day periods or by dilatometric measurement of autoclaved samples exposed for 2 h were comparable. This suggests the use of an accelerated autoclave exposure procedure for determining volume expansion effects on ceramic materials in acid environments.

3. Samples that had undergone volume expansion returned to their preexposed length when heated to 700° C during dilatometric testing.

4. The use of strain gages to measure expansions was not successful.

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