

Fatty acid separation of siliceous carbonate phosphates

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Abstract—The University of Alabama Mineral Resources Institute (MRI) has developed a unique process for selective fatty acid flotation of carbonate gangue from sedimentary apatites (francolite) in the pH range of 4-6 without collector conditioning of the pulp. The process, which does not require an apatite depressant, was modified to beneficiate a high-MgO siliceous phosphate matrix from south Florida. The modified process includes two flotation stages, one for carbonate removal and the other for phosphate recovery from the siliceous gangue. The selective flotation separation of the carbonate and phosphate from siliceous constituents of the matrix at various pH levels and collector dosages are discussed. In the carbonate flotation stage, about 70% of the MgO was rejected in the froth. The phosphate flotation stage produced concentrates analyzing about 31% P_2O_5 , 0.7% MgO, and 4% insol. The overall P_2O_5 recovery in the two stages was about 85%

Introduction

The MRI "No-Conditioning Process" for carbonate gangue removal from phosphate ores has been demonstrated to be effective on sedimentary phosphate ores from Asian sources (Anazia and Hanna, 1987). The Asian ores are liberated at 300-150 μm (48-100 mesh) and are essentially free of acid insoluble matter. For example, Jhamarkotra phosphate ore from Udiapur, India analyzes about 18% P_2O_5 , 10% MgO, and 3% SiO_2 . Single-step fatty acid flotation of the carbonate gangue via the MRI process yielded a phosphate concentrate analyzing about 38% P_2O_5 , 0.8% MgO, and 6% SiO_2 , with an accompanying P_2O_5 recovery of more than 80%. The high quality of the product and its low silica content obviated the need for de-oiling and cationic silica flotation. For ores containing significant amounts of siliceous material, how-

ever, the phosphate can be separated from the silica in a single flotation step after the carbonate flotation.

This paper presents results of work done in applying the MRI process to a siliceous-carbonate phosphate matrix from south Florida. The primary objective was to achieve maximum removal of dolomitic limestone and siliceous gangue and to recover high grade phosphate concentrates.

Experimental procedures

Materials

Ore tested: A high-MgO phosphate matrix from the W.R. Grace Co.'s Four Corners Mine in Bartow, FL was tested. The run-of-mine material analyzed 12.1% P_2O_5 , 24.1% CaO, 8.9% MgO, and 29.0% acid insoluble matter (insol).

Examination of the sample using polarized light and scanning electron microscopy revealed that it consisted of apatite, dolomite, feldspar, and quartz (Fig. 1).

The apatite was brown to black and occurred as smooth pebbles, as pebbles with inclusions of quartz and dolomite, as pebbles with partial rims or depressions filled with carbonate, as phosphate cementing dolomite and quartz, and as fossil

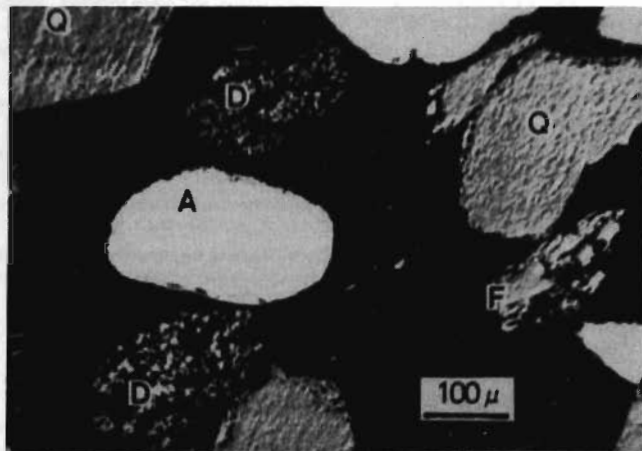


Fig. 1—Photomicrograph of matrix from the Four Corners mine, showing quartz (Q), dolomite (D), apatite (A), and feldspar (F).

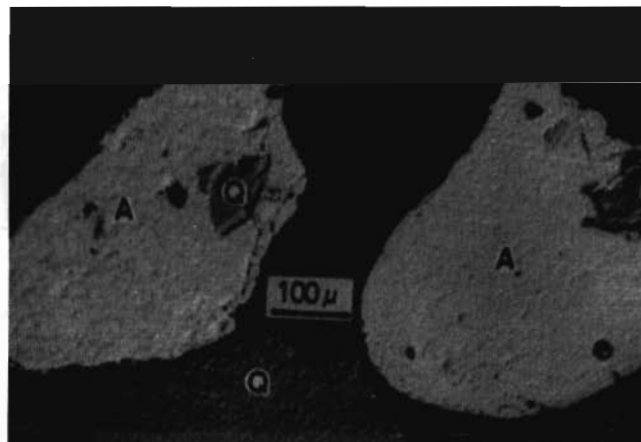


Fig. 2—Photomicrograph of apatite (A) particles showing quartz (Q) inclusions.

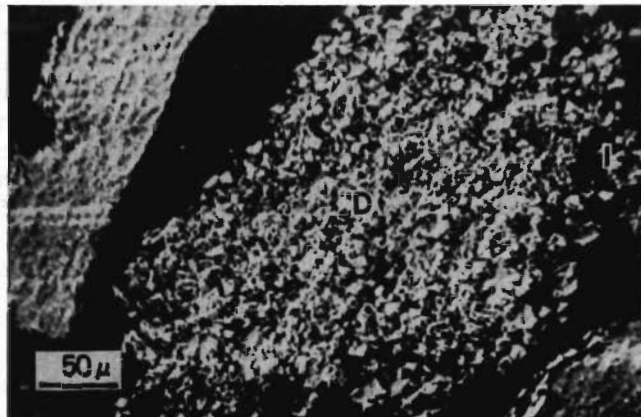


Fig. 3—Photomicrograph of a dolomite (D) aggregate with an inclusion (I) containing Mg, Al, Si, and Ti.

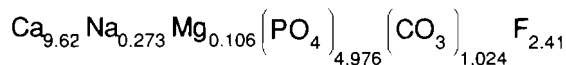
J. Hanna, member SME, and I. Anazia are with the Mineral Resources Institute, University of Alabama, Tuscaloosa, AL. SME preprint 89-144, SME Annual Meeting, Las Vegas, NV, February 1989. M&MP paper 89-625. Manuscript Dec. 6, 1988. Discussion of this paper must be submitted, in duplicate, prior to Aug. 30, 1990.

shark teeth. The photomicrograph in Fig. 2 shows apatite particles containing quartz inclusions.

The dolomite occurred as soft agglomerates of rhombohedral grains (typically <30 μm, 530 mesh) with quartz and apatite pebble inclusions, and as carbonate rims (described above). A dolomite aggregate with an inclusion of Mg, Al, Si, and Ti is seen in Fig. 3.

The quartz occurred as rounded grains and as inclusions in apatite or dolomite agglomerates.

X-ray diffraction analysis and cell parameters *a* and *c* indicated that the apatite mineral in the matrix was francolite having the following empirical formula:



Based on McClellan and Lehr's (1969) analytical method, the theoretical composition of the francolite mineral was 36.28% P₂O₅, 55.34% CaO, 4.7% F, 4.63% CO₂, 0.87% Na₂O and 0.44% MgO. The X-ray diffraction pattern of the carbonate mineral in the samples matched the pattern for dolomite. However, the refractive indices for the carbonate mineral were between the literature values for dolomite and calcite.

Flotation feed: The matrix was slurried in tap water and screened at 500 μm (35 mesh) and the minus fraction was deslimed at 104 μm (150 mesh). The plus 35-mesh material was roll crushed to minus 35-mesh and deslimed at 150-mesh. The resulting 35 x 150-mesh products of both steps were mixed and stored wet until used. Chemical analysis of this feed was approximately 10% P₂O₅, 1% MgO, 11% CaO, and 65% insol. Thus, most of the MgO in the matrix was removed in the minus-150-mesh fraction (slime).

Point counting and sink/float separation of the 35 x 150-mesh feed, indicated that the apatite particles were not completely liberated from the dolomite and quartz. About 56% of the phosphate occurred as free particles analyzing 31.4% P₂O₅, 0.6% MgO, and 3% insol. About 40% of the phosphate occurred as locked carbonate/phosphate particles analyzing 21.4% P₂O₅, 5.4% MgO, and 4.7% insol. About 92% of the siliceous impurities occurred as free particles. The characterization studies suggested that the maximum phosphate concentrate grade that can be produced by flotation would have an analysis of about 32% P₂O₅, 0.6% MgO, and 3% insol.

Collectors and frothers: Four collectors, two fatty acids and two fatty acid salts, were tested. The two fatty acids were tall oil and oleic acid. The fatty acid salts were OA-5 (a sodium salt of sulfonated oleic acid) and a commercial fatty acid soap (designated WRG) from W.R. Grace Co., Bartow, FL.

Pine oil was used as a frother in tests with the fatty acid collectors. No frother was used in tests with fatty acid soap collectors.

Flotation procedure

Flotation tests were carried out on 250-g (8.8-oz) batches of the feed (dry wt. basis) using a Model D-1 Denver flotation machine, with the impeller speed set at 1100 rpm. The feed was pulped at 8%-24% solids in the cell using tap water. The pH level was adjusted between 4-6 and maintained throughout the carbonate flotation step. Unless otherwise stated, sulfuric acid was used for pH adjustment.

The pine oil frother and fatty acid collectors were emulsified by several drops of dilute NaOH prior to addition to the pulp in three equal increments. Immediately after each collector addition, the pulp was aerated, and the carbonate froth was

collected for about 1.5 minutes. Thus, no collector conditioning time was allowed.

The flotation products were analyzed for MgO, P₂O₅, and insol.

Experiment design

Tests to study the effect of collectors and pH were randomly conducted on the phosphate matrix, using a 2^k full factorial design for collector screening and process optimization. Starpoints needed to estimate non-linear factor responses were included in the optimization test series and in some reagent screening tests. For the fatty acids, the two collector levels tested were 0.75 and 2.25 kg/t, with 1.5 kg/t as centerpoint (1.7 and 5.0 lb per st, with 3.3 lb per st as centerpoint). For the fatty acid salts, the two levels were 0.26 and 0.78 kg/t, with 0.52 kg/t as replicate centerpoint (0.57 and 1.72 lb per st, with 1.14 lb per st as replicate centerpoint). The pH levels tested were 4.5 and 6.5, with 5.5 as centerpoint for the fatty acid tests, and 3.5 and 5.5, with 4.5 as a centerpoint for the fatty acid salts.

The response variable used in the statistical evaluation of test data, which represented the MgO separation efficiency (MgO S.E.), was related to the grade and recovery of the carbonate froth product, as follows (Schulz, 1970):

$$\text{MgO S.E.} = \text{RC}_f - \text{RP}_f$$

where: RC_f = MgO recovery in the carbonate froth; and RP_f = P₂O₅ recovery in the carbonate froth.

Results and discussion

Carbonate flotation

Collector screening: The 2² test designs, for the collector screening tests, are shown in Table 1 together with the MgO S.E. responses. Statistically generated response surfaces from these data are presented in Figs. 4, 5, and 6 for tall oil, oleic acid, and OA-5, respectively. Preliminary testing indicated that collector selectivity for carbonate flotation was negligible in case of the WRG soap, and, therefore, this collector was not tested in a factorial.

The two fatty acid collectors gave similar responses,

Table 1—Experimental Design for Screening of Carbonate Flotation Collectors

Collector Type	Experiment Notation*	[A]	[B]	Response	
		Collector Dose, kg/t	pH	MgO S.E. %	
Tall Oil	(1)	0.75	4.5	3.1	
	A	2.25	4.5	20.0	
	B	0.75	6.5	11.2	
	AB	2.25	6.5	-0.1	
	M	1.50	5.5	39.9	
	M	1.50	5.5	40.8	
	M	1.50	5.5	60.6	
	MA-	0.75	5.5	40.1	
	MB-	1.50	4.5	14.7	
	Oleic Acid	(1)	0.75	4.5	10.1
A		2.25	4.5	16.4	
B		0.75	6.5	30.3	
AB		2.25	6.5	27.6	
M		1.50	5.5	58.2	
M		1.50	5.5	54.9	
MA-		0.75	5.5	40.1	
MB-		1.50	4.5	26.6	
OA-5		(1)	0.26	3.5	19.3
		A	0.78	3.5	45.0
	B	0.26	5.5	34.8	
	AB	0.78	5.5	17.0	
	M	0.52	4.5	24.3	
	M	0.52	4.5	26.7	

*Listed in Standard Yates Order (Davies, 1954)

however, oleic acid was less sensitive to pH variations and was more effective than tall oil (Figs. 4 and 5). Oleic acid gave the highest MgO S.E. of more than 54% at pH 5.5 and collector dosage of 1.5 kg/t (3.3 lb per st). The results with OA-5 (Fig. 6) were not as good, and they indicate that more data (at star-points) would be needed to delineate other regions of curvature.

The results of OA-5 flotation suggest that optimum carbonate flotation would be obtained at a pH range of 3-3.5. It would be difficult to maintain the pulp pH at this low level

without using excessive acid, therefore, further testing with OA-5 was discontinued.

Optimization of collector dose, pH, and pulp solids: Oleic acid was selected for further test work involving a three-factor, two-level (2^3) full factorial design to optimize the carbonate flotation step. The factors included in the design and the factor levels tested are given in Table 2. Also, included in Table 2 are the MgO-S.E. for the 19 experiments conducted.

Statistical analysis of the data indicated that pH and collector dose factors and cross factors had predominant effects on MgO S.E. compared to the effect of pulp solids.

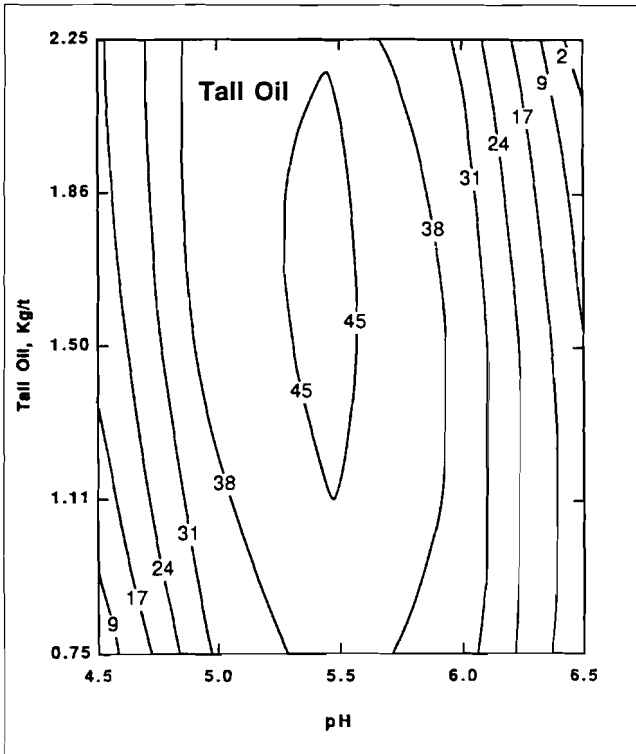


Fig. 4—MgO separation efficiency contours of carbonate flotation with tall oil.

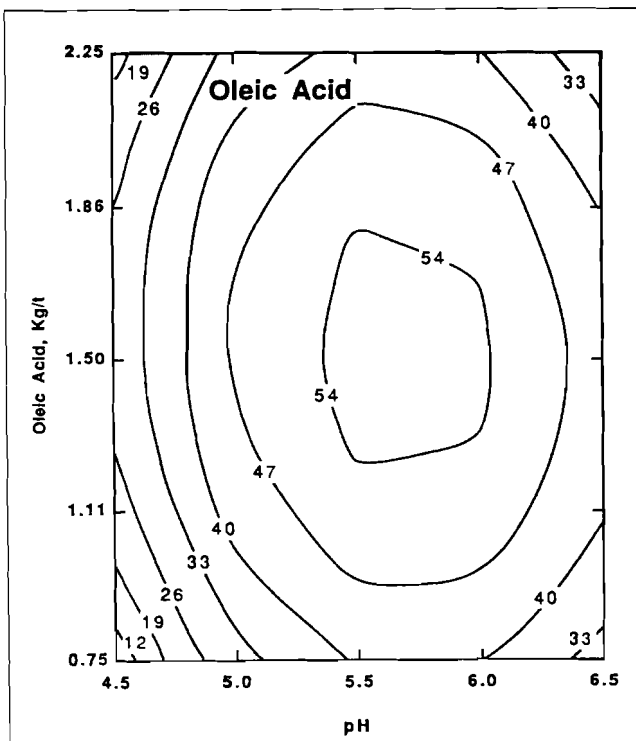


Fig. 5—MgO separation efficiency contours of carbonate flotation with oleic acid.

Experiment Notation*	[A]	[B]	[C]	Response
	Collector Dose, kg/t	pH	Solids, %	MgO S.E., %
(1)	0.75	4.5	8	21.4
A	2.25	4.5	8	31.9
B	0.75	6.5	8	38.6
AB	2.25	6.5	8	5.7
C	0.75	4.5	24	17.7
AC	2.25	4.5	24	48.5
BC	0.75	6.5	24	1.9
ABC	2.25	6.5	24	-2.0
M	1.50	5.5	16	58.2
M	1.50	5.5	16	65.3
M	1.50	5.5	16	60.6
M	1.50	5.5	16	58.5
M	1.50	5.5	16	58.3
MA+	2.25	5.5	16	27.0
MA-	0.75	5.5	16	40.9
MB+	1.50	6.5	16	14.1
MB-	1.50	4.5	16	15.6
MC+	1.50	5.5	24	55.5
MC-	1.50	5.5	8	58.6

* Listed in Standard Yates Order (Davies, 154)

Based on the MgO S.E. model developed, the response surfaces at 8%, 16%, and 24% solids were plotted separately and stacked as shown in Fig. 7.

The contours show that the region of best response shifts toward lower pH and higher collector dose as percent solids is increased. In addition to a higher collector dose, a lower pH

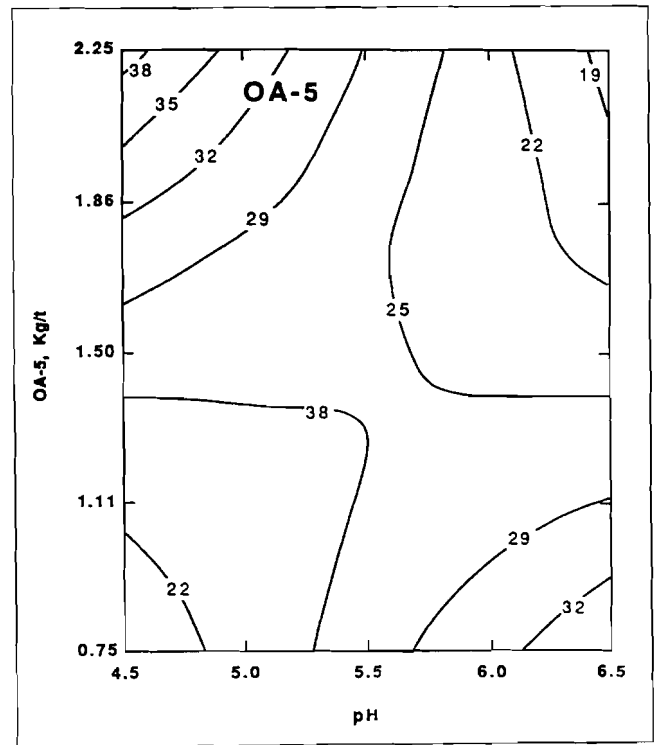


Fig. 6—MgO separation efficiency contours of carbonate flotation with OA-5.

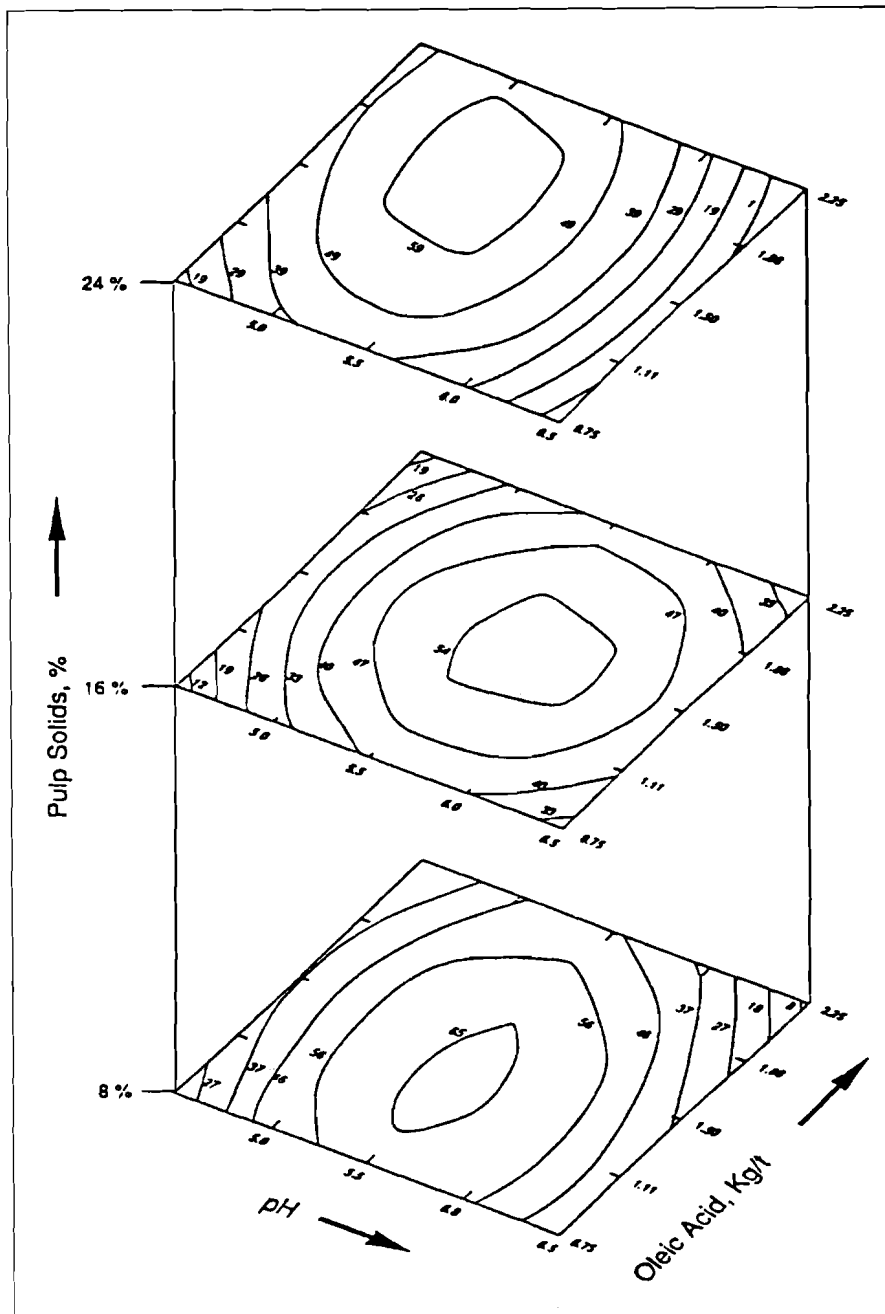


Fig. 7—MgO separation efficiency contours as a function of oleic acid dose, pH, and pulp solids.

implies higher pH regulator consumption. However, because flotation processing of dilute slurries is usually uneconomical, a pulp solids higher than 8% was preferred. Hence, 16% was considered to be the best of the three pulp densities tested.

From the response surface figure for 16% solids, a pH of 5.5 and a collector dose of 1.5 kg/t (3.3 lb per st) oleic acid were considered the best conditions to use in further optimization tests.

Effect of collector conditioning: The effect of conditioning the pulp with the oleic acid collector prior to aeration was studied under conditions stated above: pH 5.5, 1.5 kg/t (3.3 lb per st) oleic acid and 16% pulp solids. The results are shown in Fig. 8. The conditioning times shown are the totals for the 3-stage collector additions, i.e., three-minute conditioning time entailed a one-minute conditioning after each collector addition.

As shown in Fig. 8, collector conditioning had a deleterious effect on the selectivity of the carbonate/phosphate separation; while Fig. 9 shows that even at 'zero' conditioning time,

the dolomite content in the froth decreased and the apatite increased after each addition of the collector. Under this condition, the pulp was unavoidably agitated (conditioned) for about three minutes during the froth skimming.

The amounts of phosphate material reporting to the carbonate froth increased with conditioning time. This confirmed the merit of not conditioning the pulp with the collector prior to carbonate flotation.

Effect of frother dose: Tests were conducted to study the effect on carbonate flotation of increasing pine oil frother additions. Results of the tests, presented in Table 3, indicate that pine oil additions of 0.25 kg/t (0.55 lb per st) or more improved carbonate flotation and provided better frother stability. This may be attributed to frother/collector interaction.

However, flotation without a frother also was possible, indicating that the oleic acid collector had a considerable frothing property even at a pH as low as 5.5.

Effect of type of pH modifier: The results of carbonate flotation using oleic acid as collector and five inorganic acids, HNO₃, HCl, HF, H₂SO₄, and H₃PO₄, as pH modifiers are shown in Table 4.

The data indicate that the type of acid used played a significant role in determining the MgO S.E. in the carbonate froth. For monovalent acids, the carbonate separation efficiency decreased in the order HCl > HF > HNO₃. The use of these pH modifiers resulted in the removal of about 76% of the total MgO in the feed as carbonate froth. The di- and trivalent acids H₂SO₄ and H₃PO₄ gave the highest MgO S.E. but slightly lower MgO distributions of 73% and 68%, respectively, in the froth. However, use of the H₂SO₄ and H₃PO₄ produced relatively clean carbonate froths as shown in Table

3, respectively, in the froth. However, use of the H₂SO₄ and H₃PO₄ produced relatively clean carbonate froths as shown in Table

Pine oil Weight*, Dose, percent kg/t	Analysis, %			Distribution, %			MgO S.E., %	
	P ₂ O ₅	Insol	MgO	P ₂ O ₅	Insol	MgO		
0.000	3.90	13.02	7.29	11.57	5.5	0.5	50.4	44.9
0.250	6.40	12.34	9.92	11.28	7.8	1.0	65.9	58.1
0.375	3.90	3.93	7.46	16.55	1.5	0.4	61.4	59.9

* Flotation Feed: 10.16% P₂O₅; 64.03% insol; 1.01% MgO

pH Modifier Type	Weight* kg/t	percent	Analysis, %			Distribution, %			MgO S.E., %
			P ₂ O ₅	Insol	MgO	P ₂ O ₅	Insol	MgO	
HCl	0.263	10.4	14.78	9.71	8.63	16.0	1.5	74.9	58.9
HNO ₃	0.340	14.4	22.03	6.14	5.13	34.7	1.3	76.5	41.8
HF	0.113	11.1	18.37	8.23	7.56	21.2	7.8	75.9	54.7
H ₂ SO ₄	0.353	7.4	12.99	8.04	10.22	10.2	0.9	72.9	62.7
H ₃ PO ₄	0.486	6.0	8.36	9.38	11.05	6.2	0.8	68.6	62.4

Flotation Feed: 9.16% P₂O₅; 66.34% insol; 1.05% MgO

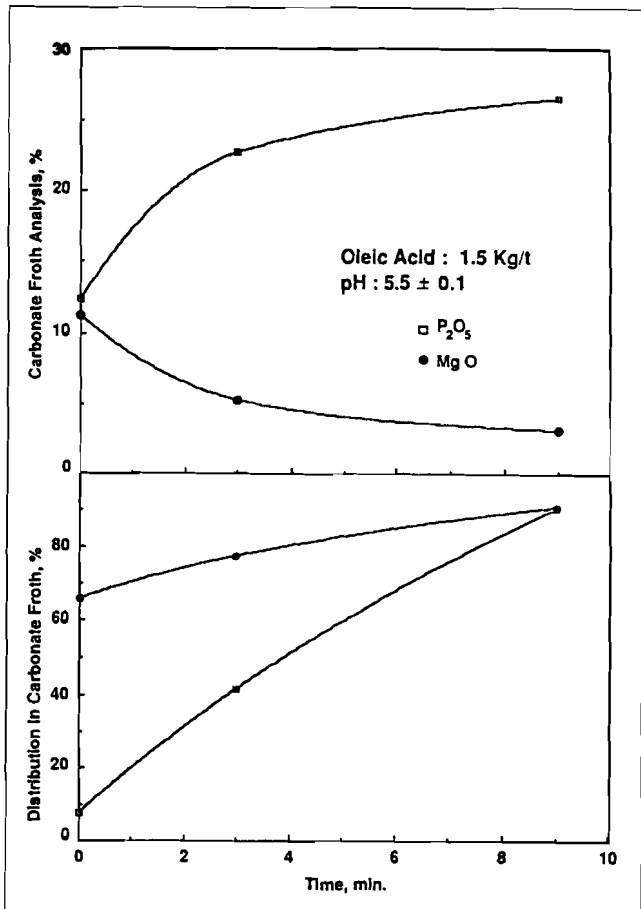


Fig. 8—Effect of collector conditioning time on carbonate flotation.

4. For practical reasons and cost-effectiveness, H_2SO_4 was the choice pH modifier for carbonate flotation.

Phosphate flotation

Phosphate flotation studies were conducted on the highly siliceous, carbonate-free pulp resulting from the carbonate flotation step. Initial tests were performed under alkaline conditions (pH 8.5) as practiced by the Florida phosphate industry, using oleic acid as collector and sodium silicate as silica depressant. Results of these phosphate flotation tests made at two collector and depressant levels are given in Table 5.

As experienced in commercial practice, oleic acid flotation in the alkaline pulp produced a rougher phosphate concentrate with a high silica content. In practice, such a product is further processed by strong acid scrubbing (de-oiling) to remove fatty

Na ₂ SiO ₃ kg/t	Oleic Acid, kg/t	Product	Weight, percent	Analysis, %			Distribution, %		
				P ₂ O ₅	Insol	MgO	P ₂ O ₅	Insol	MgO
0.00	1.5	Carbonate	6.1	8.74	9.30	12.34	5.6	0.9	70.9
0.25	0.5	Phosphate	44.2	19.16	38.00	0.60	89.7	25.8	24.9
		Tailings	49.7	0.90	96.05	0.09	4.7	73.3	4.2
		Composite	100.0	9.45	65.07	1.06	100.0	100.0	100.0
0.00	1.5	Carbonate	5.9	9.89	8.20	11.88	5.8	0.6	67.4
0.50	0.3	Phosphate	36.7	21.34	31.53	0.61	78.6	18.1	21.5
		Tailings	57.4	2.76	93.34	0.20	15.6	81.3	11.1
		Composite	100.0	10.02	65.82	1.04	100.0	100.0	100.0

Conditions
 Carbonate Flotation: 3-step addition of 0.5 kg/t oleic acid. No collector conditioning.
 Phosphate Flotation: 3-minute conditioning with Na₂SiO₃ and oleic acid at pH 8.5.

acid coatings, followed by cationic flotation of the siliceous gangue (double flotation) in order to produce a marketable-grade product of 30% to 31% P₂O₅, 0.7% to 1.0% MgO, and

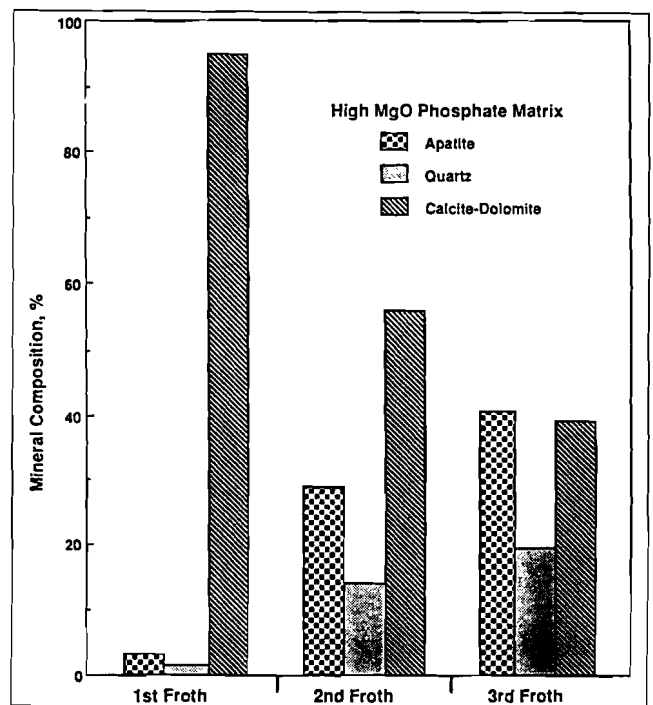


Fig. 9—Effect of collector stage addition on the mineralogic composition of carbonate froth.

8% to 10% insol. A combination of these steps with carbonate flotation would result in a multi-stage flotation process when beneficiating a high MgO/siliceous phosphate matrix. The addition of a carbonate flotation step in this process would further complicate an already complex flotation scheme.

As a result, a novel, more selective, simple, and cost effective phosphate flotation procedure was developed at MRI. This technique, which is of a proprietary nature pending a patent application, circumvents the need for the de-oiling and silica flotation steps and yields commercial grade phosphate concentrates in one step. The results of two tests made with the MRI procedure are presented in Table 6.

Product	Weight percent	Analysis, %			Distribution, %		
		P ₂ O ₅	Insol	MgO	P ₂ O ₅	Insol	MgO
Test 1							
Clean Phos. Conc.	27.9	30.68	2.80	0.75	84.4	1.2	21.6
Cleaner Tailings	2.1	16.80	45.02	0.59	3.4	1.4	1.3
Rough. Phos. Conc.	30.0	29.73	5.69	0.74	87.8	2.6	22.9
Carbonate Froth	5.6	10.81	7.39	12.57	6.0	0.7	73.1
Silica Tailings	64.4	0.98	96.58	0.06	6.2	96.7	4.0
Composite	100.0	10.15	64.33	0.97	100.0	100.0	100.0
Test 2							
Clean. Phos. Conc.	27.3	31.41	2.92	0.70	83.8	1.2	18.2
Cleaner Tailings	1.6	14.66	51.59	1.54	2.3	1.3	2.3
Rough. Phos. Conc.	28.9	30.49	5.58	0.75	86.1	2.5	20.5
Carbonate Froth	6.4	11.55	10.81	12.53	7.3	1.2	76.4
Silica Tailings	64.7	1.05	93.98	0.06	6.6	96.3	3.1
Composite	100.0	10.22	63.11	1.16	100.0	100.0	100.0

The data show that cleaner concentrates analyzing about 31% P₂O₅, 0.7% MgO, and less than 3% insol were produced with attendant P₂O₅ recoveries of about 84%. The rougher tailings produced were low in P₂O₅. The tests also showed that the cleaner tailings products could be recycled either to the rougher phosphate or carbonate flotation steps depending on their carbonate content.

Selectivity in carbonate/phosphate flotation

Several factors appear to be responsible for the effectiveness of the No-Conditioning Process in selectively separating

the carbonate gangue constituents from high MgO phosphate matrix without the use of a phosphate depressant and without collector conditioning. The fact that selective fatty acid flotation of carbonate minerals from francolite can be achieved at pH 5.5 regardless of the type of pH regulator (Table 4) points to the importance of hydrogen ion concentration in determining the surface properties and collector adsorption behavior of the minerals. In the acidic pulps the carbonate minerals (dolomite and calcite) react with the inorganic acids. This chemical reaction results in continuous dissolution or removal of surface contaminants and ensures the availability of clean surface suitable for fatty acid adsorption (Johnston and Leja, 1978).

In addition, during dissolution, C₂ microbubbles are generated on the carbonate particle surfaces leading to enhanced oleic acid adsorption at the solid/liquid/gas interface.

These are favorable conditions for particle/bubble attachment and flotation of carbonate minerals.

On the other hand, the reaction of the acids with the apatite particles may produce high concentrations of orthophosphate ionic species in the surface water layers surrounding the mineral particles. The phosphate-rich water layer is reported to be strongly hydrogen bonded to apatite and therefore depresses its flotation (Bertolucci et al., 1968).

Prolonged conditioning of the pulp appears to remove these depressant layers from the apatite surface and results in increased flotation, as shown in Figs. 8 and 9. The results also indicate that minimum contact was essential to achieve selective flotation of the carbonate minerals and depression of apatite. According to Johnston and Leja (1978), oleic acid can adsorb much faster on dolomite than on apatite because CO₂ evolves from the mineral surface and disturbs the hydrogen-bonded phosphate-rich water layer. Moreover, higher oleate adsorption on dolomite than on francolite has been reported by Moudgil and Chanchani (1985).

Conclusions

The research has demonstrated that the MRI No-Condi-

tioning Process is effective for selectively floating the carbonate minerals from a high MgO phosphate matrix obtained from the W.R. Grace Co.'s Four Corners Mine. In the phosphate flotation step that followed, high quality phosphate concentrates containing about 31% P₂O₅, 0.7% MgO, and 3% insol were obtained by a unique phosphate flotation procedure. The technique obviates the need for de-oiling and silica flotation steps used in current phosphate practice.

The selectivity in carbonate/phosphate separation, in slightly acidic media, may be attributed to preferential dissolution of the mineral surface layers that results in changes in adsorption kinetics and preferential flotation of the carbonate minerals.

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