

HYDROTHERMAL ALTERATION OF A RHYOLITE FLOW BRECCIA NEAR SAN LUIS POTOSI, MEXICO, TO REFRACTORY KAOLIN

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Abstract—A deposit of kaolin clay, the site of the General Zaragoza Mine, located about 50 km northwest of San Luis Potosi, S. L. P., Mexico, has produced more than 250,000 tons of refractory clay by room and pillar mining methods during the last 32 years. The clay was formed by hydrothermal argillation of part of a fault block of rhyolite flow-breccia and probably welded tuff, presumably lower Tertiary in age. A silica-rich gossan now exposed at the surface caps the clay deposit.

Samples collected sequentially from the fresh rock to the most highly altered kaolin and studied by optical, X-ray, DTA, and chemical methods, show progressively:

(a) Bleaching of the breccia matrix accompanied by mobilization of much of the iron, which was concentrated locally as spots of reddish oxide in the least altered areas.

(b) Mobilization, and removal of considerable alkali and alkaline earth metals, in excess of that required to form montmorillonite; mobilization of silica, which was redeposited as fine anhedral quartz crystals disseminated within the montmorillonite and associated kaolinite-halloysite, or developed tripolitic, argillized rock peripheral to the clay ore body.

(c) Further desilication of the silica and clay yielding relatively pure, but poorly ordered, kaolinite-halloysite as the most intense end product of argillation. Much silica removed during intense argillation was reprecipitated as tripolitic clay, as minor cristobalite in microscopic globules and massive aggregates characteristic of colloform opal, and in vastly larger amounts as opal and chalcedony replacing the rock overlying the clay (forming a silica gossan).

(d) Minor amounts of alunite are present at two places in the deposit, which are interpreted tentatively as being the most probable loci of rising solutions.

INTRODUCTION

A HUGE deposit of hydrothermally argillized, refractory kaolinitic clay, commonly known as the General Morelos Zaragoza or the Arenal clay mine is located in the state of San Luis Potosi, Mexico, 4 km west of the 47 km post of the paved highway running north from San Luis Potosi to Zacatecas, Zac. The deposit has been worked since 1934, first by Sr. Ismael Gonzalez and subsequently by the late General Roberto Morelos Zaragoza, and has produced more than 250,000 tons of highly refractory clay (PCE 34–35). The clay is presently being trucked to the calcining plant of Refractarios A. P. Green, S. A., at San Luis Potosi, and thence to their manufacturing plants in Mexico City, and Monterrey, N. L.

GENERAL GEOLOGY

The clay deposit has been developed for about 250 m along the lower zones of a roughly rectangular, N–S trending, faulted block of rhyolite flow-breccia and probably welded tuff, approxi-

mately 4–5 km long and less than 1 km wide, Fig. 1. This volcanic parent rock, thought to be lower Tertiary in age, overlies shales tentatively assigned to the Jurassic. The clay deposit is stream-eroded along the east and south, and cut by high-angle faults along the west and north. Argillation occurred along a N–S elongate tabular zone, the base of which dips about 4° approximately N60°W.

The size and general shape of the deposit is shown in a schematic mine map, Fig. 2. Mine openings and rooms, driven in from the hillside, are typically 6–8 m wide and 6 m high, leaving pillars 8×8×6 m, Fig. 3. Argillation typically diminishes gradually downward, upward, and laterally (except where cut by faults). Clay generally grades laterally into less-leached parent rock, but upward continuously into secondarily silicified country rock, a silica gossan. This secondary silica was derived from the clay zone below while the parent rock was desilicated during argillation. Silica gossans are characteristic of hydrothermal clay deposits in Mexico, and may be used as a guide to ore in clay prospecting (Hanson and Keller, 1966).

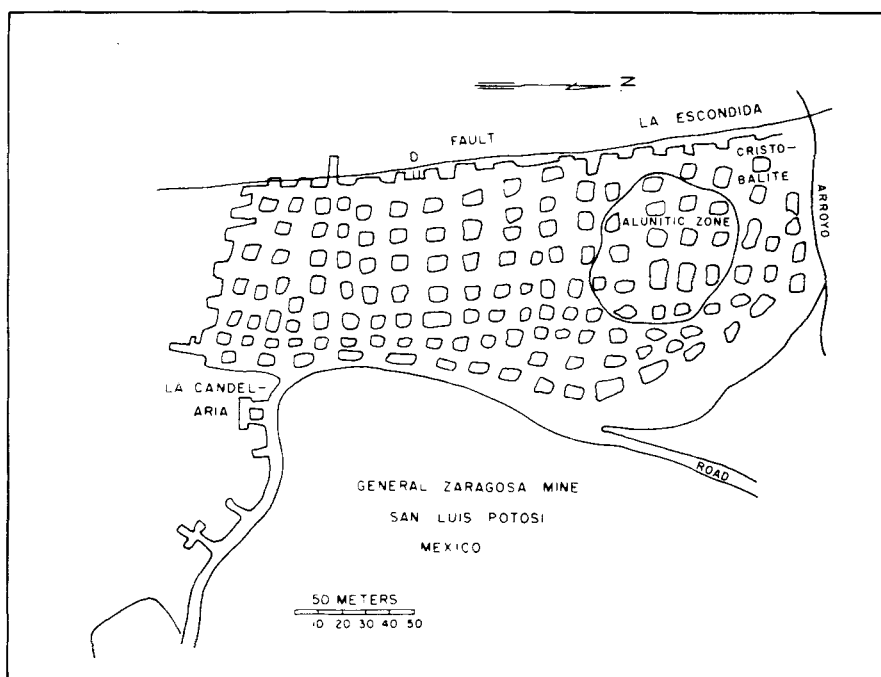


Fig. 2. Schematic map of General Zaragosa clay mine.

SEQUENTIAL ARGILLATION

The hydrothermally argillized zone was systematically sampled by collecting 20 specimens selected sequentially from fresh rock to high-quality clay, and from the silica gossan above the clay.

Parent fresh rock

The fresh rock is typically light pinkish gray, fine-textured, porphyritic rhyolite. It is porcelaneous to stony in luster suggesting that the rock may have been slightly altered, but no more so than many felsitic rocks ordinarily accepted as being fresh. Phenocrysts are fresh glassy sanidine and quartz up to 1×2 mm in cross section.

From thin-section traverses, the content of phenocrysts ranges from 15 to 32 per cent with a mean of 24 per cent. Sanidine predominates in the blocky phenocrysts whereas quartz is more abundant in finer, commonly triangular slivers ranging in size down to $40 \times 125 \mu$. About 5–10 per cent of the "phenocrysts" are micro-brecciated sub-angular rock fragments composed of sericitized microcrystalline feldspar and quartz. Swirls and parallel micro-laminae of devitrified, tightly compacted shards indicate some of the rock is probably welded crystal tuff. Other specimens show typical rhyolite flow structure, including tiny elongate vesicles lined with tridymite (identified by X-ray

diffraction of the pulverized rock). Minor kaolin-group mineral is recorded also in the diffractogram and confirmed by differential thermal analysis of the tuffaceous rock, although this was not recognized optically in the thin section.

Minor reddish pigmentation comes from tiny roundish to roughly hexagonal spots irregularly disseminated through the ground mass and along some feldspar cleavages.

A chemical analysis of the porcelainous rock specimen, "a" of Table 1, shows a lower alkali metal content and higher loss on ignition than an "average" (Daly, 1933) rhyolite composition. It is not inconsistent, however, with the composition of a slightly hydrated tuff that contains minor kaolin (recorded in the X-ray diffractogram).

Intermediate alteration zone products

Intermediate alteration between parent rock and end-product clay was studied in twelve samples selected from various transitional zones on outcrops and in the mine. Two of these were analyzed chemically, "b" and "c" in Table 1. Changes observed in the samples due to alteration may be summarized as: (1) mobilization (*i.e.* movement), segregation, and oxidation of iron, (2) montmorillonitization, (3) kaolinization, loss of alkali and alkaline earth metals, and extensive mobile redistribution of silica. Probably all three processes operated simultaneously, but the reactions ap-



Fig. 1. Looking west at 250-m long open face of General Zaragosa clay mine in San Luis Potosi, Mexico.



Fig. 3. Looking north along face of entries into clay deposit.

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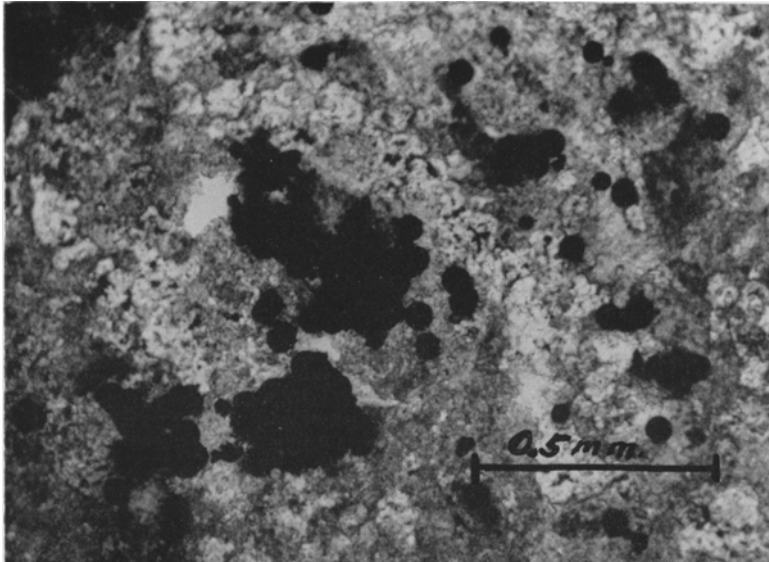


Fig. 4. Photomicrograph of iron oxide mobilized in early-stage alteration and redeposited in single and coalesced spheres.

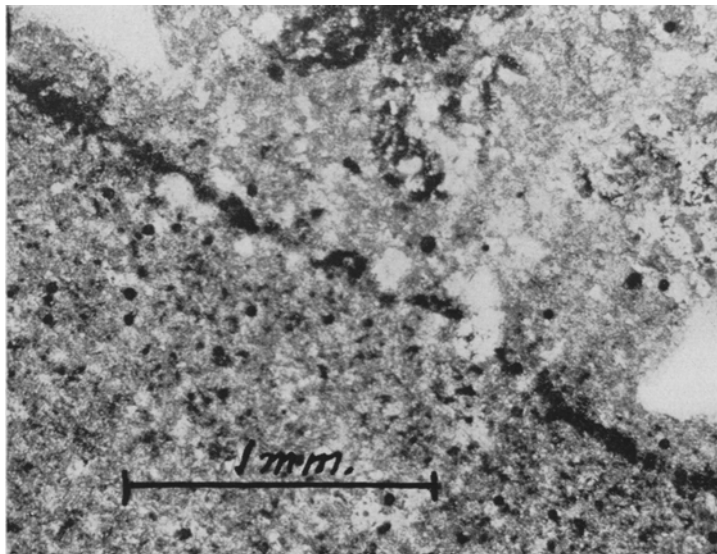


Fig. 5. Photomicrograph of line of iron oxide spots at the advancing front of alteration in the rhyolite.

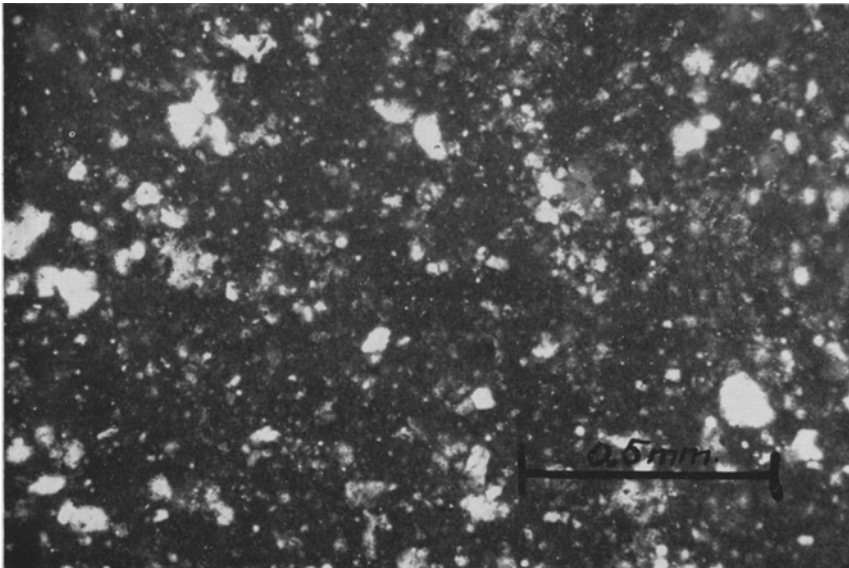


Fig. 6. Photomicrograph of fine-grained, birefringent, light-colored quartz grains disseminated in dark, low-birefringent clay.

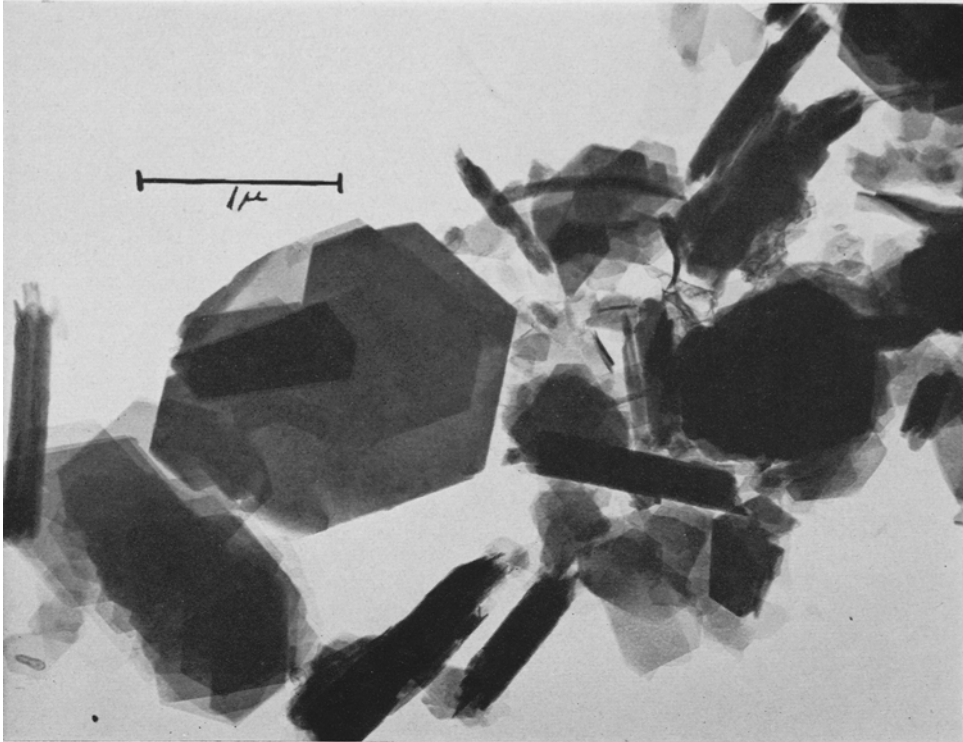


Fig. 7. Electronmicrograph of General Zaragosa clay. Note that both plate and scroll morphologies are present. Microscopy by Mr. Raymond Buck, Electron Microscope Laboratory, University of Missouri.

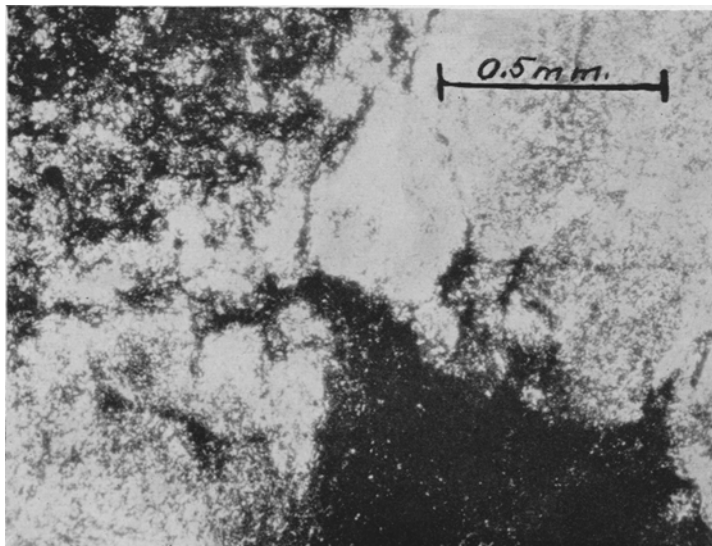


Fig. 10. Photomicrograph of alunite, light birefringent areas, in association with kaolin, dark areas. Crossed polars.

Table 1. Chemical analyses

	"a" (%)	"b" (%)	"c" (%)	"d" (%)
H ₂ O, 105°C On dry basis	0.59	3.08	0.18	0.38
SiO ₂	73.66	70.78	73.27	45.43
Al ₂ O ₃	16.85	15.83	17.04	39.78
Fe ₂ O ₃	0.92	2.29	1.53	0.09
FeO	0.12	0.06	0.10	none
TiO	0.06	0.06	0.35	none
CaO	0.21	0.18	0.25	0.20
MgO	0.11	0.18	0.11	0.13
Na ₂ O	0.86	0.25	0.77	0.25
K ₂ O	2.15	0.74	0.13	0.05
MnO ₂	none	none	none	none
SO ₃	0.05	0.07	0.07	0.02
P ₂ O ₅	0.03	0.08	0.13	0.03
Loss on ig	4.98	9.48	6.25	14.02

Analyses by Bruce Williams Laboratories, Joplin, Missouri. Cost of analyses defrayed by Research Council Grant, University of Missouri.

proached completion in the order given (iron first).

Hand specimens and thin sections show bleaching of the rock which apparently was accompanied by concentration of the iron into bands or zones or red-purple iron oxide $\frac{1}{2}$ -1 cm wide. In micro-scale, *i.e.* in thin section, the iron oxide was concentrated in tiny blebs or spheres which are grouped irregularly, Fig. 4. or as a wave front at the contact surface between "rock and clay," Fig. 5. This behavior of iron appears to be typical of early-stage hydrothermal argillation. It has been documented from occurrences at another large hydrothermal clay deposit at Guanajuato (Hanson and Keller, 1966), and at a kaolinized dike in Makhtesh Ramon in Israel, (Bentor, Field Trip, Int. Conf. Clay Mins., 1966), and has been used successfully in other field appraisals of hydrothermal argillation.

Clay-rich portions of thin sections of these rocks may show highly birefringent "needles" in moderate abundance within a clouded, dark, low-birefringent ground mass. Although the birefringent mineral resembles sericite optically, X-ray diffraction shows it to be montmorillonite or mixed-layer montmorillonite. Upon glycolation, various samples of the material yield strong 17 Å peaks, some of which are broadened by mixed layering, but scarcely any 10 Å response. X-ray shows that the microscopically unidentifiable dark ground mass contains quartz, estimated at 20 per cent content by subtracting 50 per cent, the approximate silica

content of montmorillonite, from 70 per cent, the total silica in "b", Table 1.

Other samples, illustrated by analysis "c", and thin sections show kaolinization, which presumably is a later and more intense stage of argillation than is montmorillonitization. The kaolinitic material is white, tripolitic, porous, and relatively low density. Kaolin is the dominant mineral in it, but this clay is accompanied by conspicuous (in thin section) irregular grains of chert-like quartz, Fig. 6.

In analysis "c", the presence of quartz is indicated by high total silica, 73.3 per cent, which is essentially the same as that in counterparts of fresh rock, 73.7 per cent and montmorillonitized material, 70.8 per cent. Although kaolin is desilicated relative to the antecedent rock, the silica has remained as quartz. The geochemistry of this association will be discussed later.

End-product kaolinite-halloysite zone

The end product of the hydrothermal alteration is a white, relatively pure kaolin composed of plates of poorly ordered kaolinite(?) and scrolls of halloysite (?), Figs. 7-9, and analysis "d", Table 1. Estimates from numerous electron-micrograph fields indicate that plates slightly exceed scrolls in amount. Interrelationships between morphology and nomenclature of the kaolin minerals are being reexamined (Brindley and de Souza Santos, 1966; Chukhrov and Zvyagin, 1966). The kaolin is uniformly very fine-grained, randomly oriented in this section, appearing much like flint clay. The kaolin occurs in the mine commonly as massive layers, separated by partings inherited from between flows. Some layers contain small lenses or bodies of kaolin which preserves relic structure of the brecciated rhyolite.

Cristobalite

In the extreme northwestern corner of the deposit, and in the La Candelaria portion of the southeast, both parts being peripheral to the deposit, cristobalite is developed in (and from?) microglobules of opal and coalesced globular aggregates of opal. Apparently the opal, which replaced clay (?) and rock, and filled voids, subsequently inverted, at least in part, to cristobalite. The occurrence of cristobalite with its precursor opal in the less-altered border zones of the deposit suggests a variation in intensity of activity: (1) low enough in vigor and temperature for opal to be deposited, but (2) interspersed with higher intensity to mineralize the opal to cristobalite.

Cristobalite has been observed in the "boiling mud" within the orifice of an active hot spring north of Ciudad Hidalgo, Michoacan, on the Mexi-

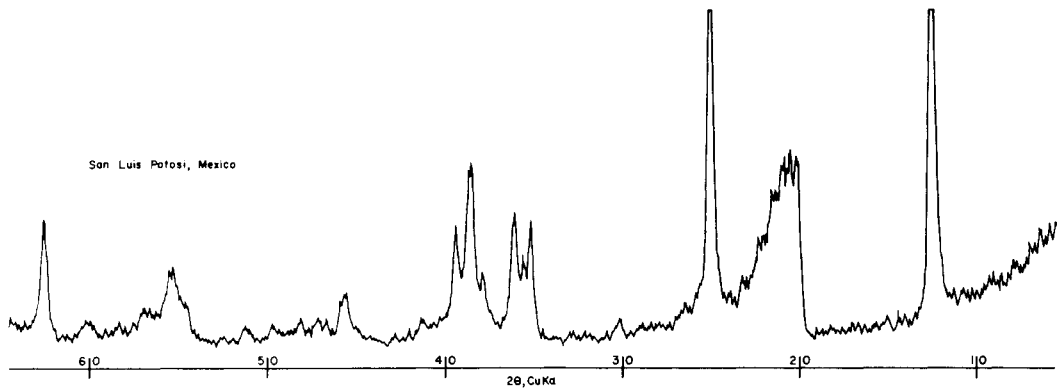


Fig. 8. X-ray diffractogram of first-quality, mixed kaolinite-halloysite from General Zaragosa mine.

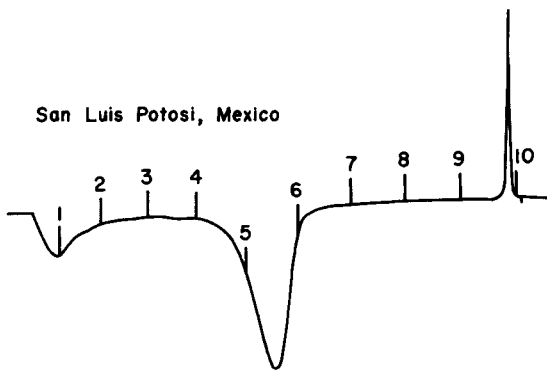


Fig. 9. Differential thermalgram of first-quality, mixed kaolinite-halloysite from General Zaragosa mine.

can volcanic axis, some 280 km south of San Luis Potosi.

Alunite

Small amounts of alunite, which may be almost indistinguishable to the unaided eye are mixed here and there with the kaolin in the San Luis deposit. Clay miners detect a "powdery", slightly harsher, feel from alunitic clay than from pure kaolinite. In the northern part of the deposit, as shown on the mine map, alunite may comprise from 5 to 50 per cent of the clay. It is microcrystalline in texture and intimately associated with kaolin, Fig. 10. Presumably the alunite was developed simultaneously with kaolinization by action of acid sulphate-rich hydrothermal solutions. Elsewhere in Mexico, alunite has been observed to be the dominant mineral, with kaolinite minor in quantity, in hydrothermally altered deposits (Knizek and Fetter, 1946). Alunitization of kaolin may develop as well, however, from cold, surface-water reaction of kaolin with iron-sulphide derived H_2SO_4 (Keller, Gentile, and Reesman, 1967).

Silicified cover rock—gossan

Intense silicification of the overlying flow breccia, presumably a part of the parent-rock series, has converted it into a pastel-colored, fine-textured, smooth, opalescent and chalcedonic silica rock commonly retaining partly corroded, relic breccia fragments. Other less-silicified parts of the covering rock show abundant small leached pore, oxidized iron minerals, and some kaolinite (in both DTA and diffraction pattern). Breccia fragments are in strong color contrast to one another and the matrix due to differential bleaching and oxidation. Tuff fragments retain laminar shard structure that has been blurred during replacement (mainly by opal). Cavities and interstices contain microcrystalline, and commonly undulatory and probably chalcedonic, quartz clusters. The silicified "gossan" extends as far as 5 m above the top of the clay body.

FACTORS OF ORIGIN

The basic geochemical controls of the argillic alteration, including zoning sequence, presumably are the ratio of H^+ to alkali and alkaline earth ions related to silica in the altering solution (Hemley and Jones, 1964; Garrels and Christ, 1965), and to the compositions of the parent rock and daughter mineral products (Keller, Kiersch, and Howell, 1955; Keller, 1956). Although no direct chemical measurements of the geologic solutions are available, indirectly the abundance of opal and cherty silica in the clay and gossan indicates high H_4SiO_4 in solution which, in turn requires a low K^+/H^+ ratio to produce kaolinite (in accord with stability diagrams of the system $K_2O-Al_2O_3-SiO_2-H_2O$, Garrels and Christ, 1965, and at higher temperatures, Fournier, 1967). The presence of alunite in two locations in the clay suggests that sulphide waters rose here and oxidized to acid sulphate

waters, presumably from surface contribution of oxygen.

One aspect of special mineralogical interest is the abundance of fine, cherty quartz and opal scattered through the kaolin in the tripolitic clay zone. Whereas kaolinization of rhyolite represents desilication, the deposition of secondary free silica within the kaolin obviously represents addition of silica. A probable explanation of the opposing reactions involves the differences between solubility of silica from silicate rocks and that of quartz, and the pronounced effect of temperature on silica solubility.

Silicate rocks, such as rhyolite and volcanic ash, when pulverized in distilled water at room temperature yielded 19–32 ppm dissolved silica (Keller and Reesman, 1963). The solubility of quartz under the same conditions was less than half the 32 ppm (Krauskopf, 1959). Alternately silicate could dissolve and silica precipitate. This effect could well be implemented by the temperature fluctuation of a hot spring area in Mexico due to seasonal variation in rainfall. During the drier parts of the year in Mexico the hot spring and surroundings may become hotter than when flushed with an excess of cool rain water. Under these hotter conditions hydrolysis would be accelerated by the higher temperature and concentrations of dissolved silica would increase greatly, as at Wairakei, New Zealand (Ellis and Mahon, 1967), and might well approach 300 ppm as found in hot springs elsewhere by White *et al.* (1957). During a rainy period, the influx of much more cool meteoric water in the relatively permeable flow breccia and tuff may cool the system enough to exceed saturation with silica, and either opal or fine quartz be deposited. Or, opal may have been deposited and then inverted to cherty type quartz.

The mineralogical data from the argillized samples at the San Luis deposit may be organized into a pattern of zoning, Table 2. Although such tabulation sets up arbitrary mineral categories, the actual

process is interpreted as being a continuous one resulting from a single hydrothermal episode. The most intense reaction, due to presumably hottest and most acid water, occurred at the alunite-producing source areas. As the water moved outward and upward it declined in temperature and acidity as it reacted with the rocks. Simultaneously, with the loss of H^+ ions during kaolinization of the rock the water picked up alkali and alkaline earth ions and silica. As the $[M^+]/[H^+]$ ratio and the concentration of dissolved silica thereby increased, montmorillonite was produced. Dissolved silica was precipitated in part with the clay, due to temperature drop, but most of the silica moved upward to replace and silicify the surficial rock—this was the most feeble of the geochemical and thermal reactions.

The fault on the west, a normal fault of approximately 50 m displacement, is post-kaolinite and definitely was not a source channel for the kaolinizing solutions. Some oxidized montmorillonite has formed in the fault zone, presumably by the action of descending surface water.

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Table 2. Argillic zonation, San Luis Potosi

	Hypogene source	Main deposit	Clay transition	Rock transition
Alteration Mineralogy	Kaolinite Alunite Cristobalite	Kaolinite Halloysite Sporadic quartz or opal	Kaolin Cherty quartz	Montmorillonite Quartz Opal
Geochemical Environment	Acid— SO_4 K fixed in alunite Silica in soln.	Low $[K^+, Na^+]/[H^+]$ Silica in soln. Temp. fluctuated		High $[Me^+]/[H^+]$ Silica in soln.

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Résumé—Un dépôt d'argile kaolinique, à la mine générale de Saragosse, qui est située à environ 50 km au nord-ouest de San Luis Potosi, S.L.P., au Mexique, a fourni plus de 250,000 tonnes d'argile réfractaire par des méthodes de mines souterraines au cours des 32 années passées. L'argile s'est formée par argillation hydrothermique d'une partie d'un block à faille de brèche fluidale de rhyolite et peut-être de tuf soudé, datant probablement du tertiaire inférieur. Un gossan riche en silice et maintenant exposé à la surface recouvre le dépôt argileux.

Des prélèvements tirés successivement de la roche fraîche jusqu'à la kaolinite la plus altérée et étudiés par divers moyens optiques, à rayon X, DTA, et chimiques, montrent progressivement:

(a) Une décoloration de la matrice de brèche qui s'accompagne d'une mobilisation d'une grande partie de fer, concentré localement en taches d'oxyde rougeâtre dans les parties les moins altérées.

(b) La mobilisation, et l'élimination d'une quantité considérable de métaux terrestres alcalis et alcalins, au-delà de ce qui est nécessaire à la formation de montmorillonite; la mobilisation de la silice, redéposée sous forme de cristaux de quartz anhédrique fins, disséminés à l'intérieur de la montmorillonite et de la kaolinite halloysite alliée, ou qui s'était formée en roche argilifère tripolitique à la périphérie du corps de minerai d'argile.

(c) Une désilication plus poussée de la silice et une argile donnant une kaolinite-halloysite relativement pure mais désordonnée comme produit final le plus intense d'argillation. Une grande partie de la silice éliminée au cours de l'argillation intensive a été reprécipitée sous forme d'argile tripolitique, de cristobalite secondaire en globules microscopiques ou en agregates massifs caractéristiques de l'opale colloforme, mais pour la plus grande partie sous forme d'opale et de chalcédoine en remplacement de la roche qui recouvrait l'argile (pour former un gossan de silice).

(d) On trouve de petites quantités d'alunite dans deux parties du dépôt, que l'on présume être les lieux les plus probables des solutions montantes.

Kurzreferat—Aus einer Lagerstätte von Kaolinton, im Bereich des General Zaragosa Bergwerkes ca. 50 km nordwestlich von San Luis Potosi S.L.P. Mexiko, sind im Laufe der letzten 32 Jahre durch Säulenabbauverfahren über 250,000 Tonnen Schamotteton gefördert worden. Der Ton ist durch hydrothermische Argillierung eines Teils eines Schollenbruchs von Rhyolith Fließbrekzien und vermutlich geschmolzenem Tuff, wahrscheinlich aus dem unteren Tertiär stammend, gebildet worden. Ein silikatreicher, eisenschüssiger ockerhaltiger Gossan, der jetzt an der Oberfläche freiliegt, deckt die Lagerstätte.

Proben, die fortlaufend aus dem frischen Gestein bis zum am meisten veränderten Kaolin entnommen und durch optische, Röntgen, DTA und chemische Methoden untersucht wurden, zeigen der Reihe nach:

(a) Ausbleichung der Brekzienmatrix mit gleichzeitiger Beweglichmachung eines Grossteils des Eisens, das örtlich in der Form von roten Oxydflecken in den am wenigsten veränderten Gebieten konzentriert war.

(b) Beweglichmachung und Entfernung beträchtlicher Mengen von Alkali- und Erdalkalimetallen, und zwar über den für die Bildung von Montmorillonit erforderlichen Bedarf hinaus; Beweglichmachung der Kieselsäure, die in Form feiner, allotriomorpher Quarzkristalle innerhalb des Montmorillonits und assoziierten Kaolinit-Halloysits eingesprengt abgelagert wurde, oder polierschiefriges, argillisiertes Gestein am Umfang des Tonminerals entwickelte.

(c) Weitere Entsilikatisierung der Kieselsäure und des Tones unter Bildung von verhältnismässig reinem, jedoch wenig geordnetem Kaolinit-Halloysit als ausgeprägtestes Endprodukt der Argillisie-

zung. Eine grosse Menge von Kieselsäure, die während der intensiven Argillisierung entfernt worden war, wurde zu einem geringen Teil als Cristobalit in mikroskopischen Tröpfchen und massiven für colloformen Opal charakteristischen Aggregaten, und in bedeutend grösseren Mengen als Opal und Chalcidon niedergeschlagen. Die letzteren Minerale ersetzen das den Ton bedeckende Gestein (eisenschüssiger, ockerhaltiger Gossan).

(d) Geringe Mengen von Alaunstein sind an zwei Stellen der Lagerstätte vorhanden, die vorläufig als die wahrscheinlichsten Ortlichkeiten steigender Lösungen angesehen werden.

Резюме—Залежи каолинитовой глины—местоположение шахты “Генерал Зарагоза”—находится при бл. в 50 км на северо-запад от Сан-Луис-Потоси, Мексика,—дали за последние 32 года добычу свыше 250 000 тонн огнеупоров, при работах по методам камерно-столбовой выемки. Глина образовалась гидротермальной аргиллизацией части сбросового целика риолитовой брекчии и возможно также сплавленного туфа, повидимому раннего третичного периода. Окрашенные окислом железа выходы жил, с крупным содержанием кремнезема, обнаженные теперь на поверхности, покрывает отложение глины.

Последовательно отобранные образцы от свежесбитой породы до наиболее измененного каолина и исследованные оптически, рентгенографически, ДТА и химическим методом, показывают прогрессивно:

(а) обесцвечивание жильной породы, брекчии с сопутствующей мобилизацией большого количества железа, которое сосредоточивалось местно в виде пятен красной окиси в наименее измененном участке;

(б) мобилизацию и удаление значительного количества щелочи и щелочноземельных металлов, превышающего количество, требуемое для образования монтмориллонита; мобилизацию кремнезема, который был переотложен в качестве тонких ангедральных кварцевых кристаллов, рассеянных в монтмориллоните, и сопутствующем галлоузитовом каолините, или-же развил трепельную, аргиллитную окружность породы на глинистом рудном теле;

(в) дальнейшее удаление кремнезема и глины, что дает сравнительно чистые, но не регулярно расположенные выемки каолинитогаллоузита в качестве наиболее интенсивного конечного продукта аргиллизации. Большое количество кремнезема, удаленного в ходе интенсивной аргиллизации, переосаждается как трепельная глина, как мелкий кристобалит в микроскопических шариках и как крупный агрегат, характерный для коллоформного опала и в значительно больших количествах в виде опала и халцедона, замешая породу залегающую над глиной (образуя силикатную железную шляпу);

(г) малые количества алунита присутствуют в двух местах в отложении, а это подается предварительной интерпретацией как наиболее правдоподобные участки восходящих растворов.