MINERALOGICAL INVESTIGATIONS IN THE DEBRIS OF THE GNOME EVENT NEAR CARLSBAD, NEW MEXICO[1]

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ABSTRACT

Mineralogical studies on the debris formed during the underground Gnome nuclear explosion in a salt horizon of the Salado formation near Carlsbad, New Mexico have shown that significant quantities of olivine and kirschsteinite are present in the water insoluble fraction. Particles containing these minerals are generally strongly radioactive and suggest the minerals have had a role in localizing the radioactive fission products. The size (10–500 μ) and spherical shape of the silicate particles suggest that they were immiscible droplets in the salt melt which preferentially dissolved the fission products before crystallizing. The subsequent crystallization excluded the actinides from within the crystallites, leaving them on the crystallite boundaries.

INTRODUCTION

The Gnome underground nuclear explosion was one of a series of events in the Plowshare program, directed toward an evaluation of the peaceful uses of atomic energy. One of the objectives of the program was to determine the feasibility of recovering the actinides generated by the nuclear device. This program was termed the Coach feasibility study. The purpose of the investigation described in this paper was to determine any systematic association between the mineral assemblages in the debris and specific activities. It was determined early that the water-soluble fractions of the rock were devoid of actinides. If the mineralogy of the insoluble fraction could be defined, the extraction chemistry could be directed along the correct directions.

ENVIRONMENT

The Gnome experiment involved the detonation of a 3.1 ± 0.5 kiloton device at a depth of 1184 feet underground in the Salado formation near Carlsbad, New Mexico. It was detonated on December 10, 1961.

The region affected by the detonation was composed of about 89% halite, 7% polyhalite, 1% anhydrite and 3% silt and clay by weight (Rawson, *et al.*, 1964). The impurities occur primarily as separate beds interlayered in the halite, although some are part of the salt beds. The bulk chemical analysis of the rock is given in Table 1. The nearest limestones and dolomites are well out of the range of the area affected by blast and need not be considered.

The blast produced a cavity averaging around 123 feet in diameter.

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[1] Work performed under the auspices of the U. S. Atomic Energy Commission.

TABLE 1. CHEMICAL COMPOSITION OF GNOME CORE COMPOSITES (PRESHOT)[1]

Element	Wt % in III[2]	Wt % in V[3]
Al	0.19	0.35
Ca	1.11	2.65
Fe	0.01	0.01
K	1.28	2.56
Mg	0.86	1.66
Na	34.64	28.44
Si	0.47	1.01
Cl	53.65	46.59
SO_4	4.86	13.70
H_2O (260°)	0.56	1.07
H_2O (350°)	1.07	1.96
	98.14	98.93

[1] After M. Nathans (1965).

[2] Simulated 10-m radius.

[3] Simulated 15-m radius.

The maximum cavity dimensions were developed in about 100 μsec. It is hypothesized (Rawson et al., 1964) that the cavity was lined with a liquid layer at this time. Radial fractures developed in the walls within the first minute, and some of the fluids were injected into these cracks. The lack of radioactivity in the injected melt indicated it had not yet reacted with the fission products. With increasing time the melt collected in the lower part of the cavity, and finally the cavity imploded and the roof collapsed, forming a breccia-like mass in the pool region. The melt also reacted with lead shielding and iron from the support structure, as well as with the SiO_2 sand used as backfill. The maximum shock pressure may have been 400 kilobars. The lack of mineralogical pressure indicators suggests that the melt was in existence after the shock wave passed. The maximum temperature in the cavity was high enough to vaporize a large mass of the rock. The temperature of the melt is difficult to establish, but it may have reached as high as 1800° C. (Rawson et al., 1964).

SAMPLING AND SAMPLE PREPARATION

The samples used in the isotope recovery feasibility studies were taken from drill cores recovered from the pool region of the debris. The portions of these samples which showed the highest γ activity were used in the mineralogical studies.

The samples were leached in water and the insoluble residues were divided into four fractions, using bromoform and a magnetic separator.

The insoluble residues ranged from 4% to 60% of the initial samples. A typical analysis of the water-insoluble fraction is given in Table 2. One or more grains representative of a distinct grain type, based on physical features such as color or texture, were hand picked from the bulk and cemented on the end of a short glass capillary already mounted in a 1/8-inch brass pin. This configuration was then placed directly into a standard Debye-Scherrer x-ray camera. Where larger quantities of a particular particle type could be collected quickly, standard mounts were used. Before x-ray patterns of any samples were made the mounts were checked in α, β and γ counters for their radioactivity. The small grains required long count intervals, up to 30 minutes for β counts, and x-ray exposures up to 16 hours. Even with these long exposures, many patterns

TABLE 2. CHEMICAL ANALYSIS OF WATER INSOLUBLES FROM THE GNOME MELT[1]

Element	Per cent	Element	Per cent
Al	5	Mn	0.02–0.2
B	0.02–0.2	Na	0.1–1
Ba	0.003–0.03	Ni	0.003–0.03
Ca	20	Pb	2
Cr	0.005–0.05	Si	15
Cu	0.01–0.1	Sr	0.01–0.1
Fe	4	Ti	0.5
K	Not determined	Zr	0.01–0.1
Mg	12		

[1] Spectrochemical analysis by Lawrence Radiation Laboratory.

did not yield any visible lines. This result suggested the samples were glass.

RESULTS OF X-RAY ANALYSIS

The water-soluble minerals were inactive and were not included in the analysis. The minerals identified in the insoluble residue are summarized in Table 3. The minerals showing significant radioactivity were olivine [(Fe, Mg)$_2$SiO$_4$], and kirschsteinite (CaFeSiO$_4$). The olivine is the more common, and its physical features vary considerably. Where the grains are black or dark gray, the radioactivity is high. Some olivine is white and devoid of any radioactivity. It is not always possible to distinguish the dark olivine and kirschsteinite on the basis of physical appearance, although the latter generally has a more pitchy luster. The two minerals are commonly intimately mixed in a given grain. These mixed grains always show significant radioactivity. Usually the grains are spherical or elliptical in shape with smooth surfaces, and always finely polycrys-

talline. The grains commonly appear scoriacious, indicating they were once molten and gas-filled.

The olivine group of minerals contains divalent cations which may be Mg^{2+}, Fe^{2+}, Ca^{2+}, Mn^{2+}, and even Pb^{2+}, Zn^{2+}, and Ni^{2+}. Occasionally Cr^{3+} is present in small amounts. Continuous isomorphous substitution occurs between forsterite (Mg_2SiO_4) and fayalite ($FeSiO_4$), fayalite and tephroite (Mn_2SiO_4), and fayalite and kirschsteinite ($CaFeSiO_4$). Only partial replacement occurs along other joins, *i.e.*, almost no substitution occurs between forsterite and larnite (Ca_2SiO_4), although an intermediate compound, montecellite ($CaMgSiO_4$), exists.

By measuring the d-spacing of the 174 line, assuming the olivine to contain only Fe and Mg, it was possible to determine the Mg/Fe ratio

TABLE 3. MINERALS IDENTIFIED IN INSOLUBLE RESIDUES OF RADIOACTIVE GNOME DEBRIS

Abundant		Rare
Quartz	Lead	Maghemite
Magnetite	α Iron	Brucite
Gypsum	Silicon	Barite
Anhydrite	γ Iron	Laurionite [(Pb(OH)Cl)]
γ CaSO$_4$	Hematite	Pb$_4$O$_3$Cl$_2$
Olivine	γ Fe$_2$O$_3$	Montebrasite [(LiAlPO$_4$(OH)]
Kirschsteinite	Lepidochrocite	Talc
Mica clay	Goethite	Biotite
	Galena	

of the olivine based on the determinative curve of Jambor and Smith (1964). These measurements lead to 88 ± 2 atomic % Mg in both the black and white olivines. The kirschsteinite was initially identified by comparing its measured lattice constants reported by Sahama and Hytönen (1957) for a naturally occurring kirschsteinite of composition Ca:Fe:Mg:Mn $= 95:74:22:4$. At a later date the powder pattern was compared with some synthetic $CaFeSiO_4$ for further confirmation.

Several of the iron oxides have been found in the samples, and magnetite (Fe_3O_4) is actually abundant. Although radioactivity is commonly associated with magnetite grains, usually it is high only when olivine or kirschsteinite are also present. Thus, iron as an oxide evidently is not directly associated with the radioactivity, an idea also supported by the lack of significant radioactivity differences between the magnetic and relatively nonmagnetic fractions. More details concerning the relationships among the iron oxides have been described elsewhere (Nathans, *et al.*, 1965).

One strongly radioactive grain proved to be montebrasite [LiAlPO$_4$·(OH)]. This grain was one of a kind, and its significance is unknown. Its extreme rarity did not warrant further study.

Among the nonradioactive minerals the occurrence of quartz (SiO$_2$) is most interesting. Occasionally some associated radioactivity was observed, but this condition was rare. The quartz grains were peculiar in that they were mostly bounded by well-defined crystal faces. Morphologically, these faces suggest the crystals grew as high-temperature quartz, and are now pseudomorphs.

PROBE ANALYSIS

The electron microprobe was used to examine the particles listed in Table 4. An attempt was made to detect the presence of any actinides, establish associations among elements to corroborate the x-ray analysis, and establish the presence or absence of elemental gradients within particles.

The microprobe analyzer was operated in two different ways. The defocused electron beam, approximately 25 μ in diameter, was held on one spot on the sample and an elemental analysis was performed at that spot. Upon completion of the analysis, the sample was moved to another spot and this was repeated. In this manner a grid defined the network over which the particle was sampled. These data are summarized in Table 5.

Microprobe analyses were also performed using an electron beam of 1 to 2 μ and scanning a linear traverse of predetermined length. This same linear traverse was reexecuted for each element analyzed. Here the distribution of elements over the particle may be followed through grain boundaries. At the same time, one may observe the trends of different elements along the identical traverse and see which elements are asso-

TABLE 4. β/γ ACTIVITY OF PARTICLES IDENTIFIED BY X-RAY DIFFRACTION
AND SUBMITTED FOR PROBE ANALYSIS

Particle number	Identification	β/γ
22	CaFeSiO$_4$ (Kirschsteinite)	19.2
23	MgFeSiO$_4$ (Olivine)	18.4
24	PbS, CaFeSiO$_4$	37.4
25	MgFeSiO$_4$, SiO$_2$	48.5
27	MgFeSiO$_4$	22.2
28	MgFeSiO$_4$	25.0
29	MgFeSiO$_4$	16.4
30	MgFeSiO$_4$	26.3
31	MgFeSiO$_4$, CaFeSiO$_4$	28.3

TABLE 5. ELEMENTS FOUND IN SAMPLES BY STUDYING THE SPECTRUM
ON A GRID WITH A 25 μ BEAM SPOT

Element	Sample								
	22	23	24	25	27	28	29	30	31
Mg	x[1]		x	x	x	x	x	x	x
Al	x	x	x	x	x	x	x	tr[2]	x
Si	x	x	x	x	x	x	x	x	x
Fe	x	tr	x	x	x	x	x	x	x
Ca	x	x	x	x	x	x	x	x	x
Pb			tr	tr		?[3]	tr	tr	tr
Cl	tr		tr	tr	tr	tr	tr	tr	tr
P		x		tr					
S		x	tr	tr	tr	tr		tr	tr
K		tr			x	?	tr	tr	tr
Cu		x	?	tr	x	x			
Zn	?	x	?	tr	tr	x	?		
Ba		x							
Ti	tr	?	tr	tr	tr	x	tr	tr	tr
Sb				tr					
Ni				?					
Mn	?				tr	?	?		
Cr	?								

[1] x = major constitutent.

[2] tr = trace elements.

[3] ? = not enough lines appeared to render positive identification.

TABLE 6. DISTRIBUTION OF ELEMENTS OBTAINED BY LINE PROFILE—BEAM WIDTH 1–2 μ

Element	Sample							
	22	23	24	25	27	29	30	31
Mg	x		x	x	x	x	x	x
Al	x	x	x	x	x	x	x	x
Si	x	x	x	x	x	x	x	x
Fe	x	x	x	x	x	x	x	x
Ca	x	x	x	x	x	x	x	x
Pb			x			x	x	
Cl			x			x	x	x
P		x						
S		x	x	x			x	x
K			x		x	x	x	x
Cu		x	x		x			
Zn		x	x			x		
Ba		x						

TABLE 7. CATION ASSOCIATION WITH Si^{4+} IN PARTICLES OF
HIGHLY ACTIVE GNOME INSOLUBLE DEBRIS

Sample number	Cation(s)
22	Ca probably Fe
23	Al
24	Mg, Fe, Ca
25	Ca' Al, Fe, Mg
27	Ca, Mg, Fe (in part)
29	Ca, Mg, Al, Fe (in part)
30	Ca, Al, Fe (in part)
31	Ca, Fe, Mg

ciated and which are not. These data are summarized in Table 6. Those elements associated directly with Si^{4+} are listed in Table 7.

In general, the probe analysis indicated that:

(1) Fe in olivine grains is associated with Si in part only, suggesting that magnetic iron compounds are mechanically mixed with the olivines; (2) the actinide content was too low to be identified; and (3) there were no compositional gradients.

SUMMARY

From these data it is apparent that most α, β and γ activity is associated with the silicates olivine and kirschsteinite. Electron microprobe analysis suggests kirschsteinite is found with all olivine material, since Ca is associated with Fe and Mg at all times. The exact location of the actinides is still in doubt. Based on the size of their atomic radii, it appears unlikely that the actinides are trapped interstitially within the silicate crystallites.

The spherical nature of the olivine particles suggests that they formed as immiscible droplets in the salt melt. The actinides were probably dissolved while the silicates were still liquid. Subsequent crystallization left the actinides on the crystallite boundaries within the particle.

The absence of pyroxenes in the melt indicates no reaction of the olivine with the melt.[1] The anhydrous nature of the identified minerals certainly precludes formation of hydrous silicates, such as the amphiboles. Low pressures in the melt are suggested by the absence of coesite (SiO_2), sillimanite (Al_2SiO_5), diopside [$CaMg(SiO_3)_2$] and jadeite [$NaAl(SiO_3)_2$].

There were no elemental gradients within the particles, indicating that the outer glassy surface of the particles are compositionally identical to the crystalline interior. This fact strongly suggests that the outer surface of many particles is olivine glass.

[1] Ray Cooperstein remelted some of the unleached, post-shot samples and when they were allowed to cool in an open container, the only silicates formed were pyroxenes.

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