THE AMERICAN MINERALOGIST, VOL. 46, SEPTEMBER-OCTOBER, 1961

ISOTOPIC ANALYSES OF SINGLE GALENA CRYSTALS: A CLUE TO HISTORY OF DEPOSITION

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ABSTRACT

A detailed study of lead isotopes has been undertaken in order to evaluate variations in the isotopic composition of lead with respect to the geologic environment of a single lead deposit.

INTRODUCTION

For the purpose of this investigation a lead deposit was desired in which the number of geologic variables would be at a minimum, and which would be well exposed by mine-development workings. The Blanchard Property mine, located in the Hansonburg mining district of Socorro County, New Mexico, was selected for study. Numerous development adits have been driven into the galena-fluorite-barite ore body of the Blanchard Property mine. These adits are nearly at right angles to a number of subparallel, steeply dipping, mineralized fractures. The Number 1 entry was selected for the first detailed studies. Figure 1 presents a section of the Number 1 entry.

The mineral deposit chosen for this study is typical of the Hansonburg mining district and consists of galena, fluorite, and barite, with minor pyrite, chalcopyrite, and light yellow-brown sphalerite. The principal gangue mineral is quartz. Not only is the limestone host rock intensely silicified adjacent to the deposit but quartz crustification is present in the open spaces.

The ore controls of the deposit studied have not been established in detail, but in a general sense consist of a favorable bed, the Council Spring limestone (Pennsylvanian), a capping shale, and steeply dipping

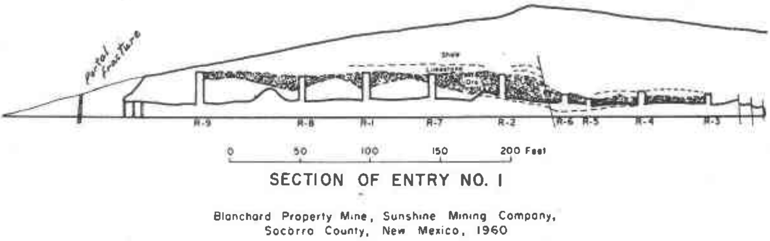

FIG. 1. Section of the No. 1 entry in which the galena samples were collected.

mineralized fractures that intersect the favorable bed. Much of the so-called bedded ore appears in the field to be the result of solution-cavity and/or breccia zone filling. Openings lined with coarse, museum-quality crystals of the ore minerals are plentiful both in the bedded ores and in the brecciated fracture zones. The deposit, in common with others in the Hansonburg district, exhibits a great uniformity of deposition with respect to mineralogy.

Sample collecting within the lead deposit was done initially to determine the gross variations in lead isotopes across a number of parallel fractures. To this end composite samples were taken at the portal fracture and at raises R-1 and R-2. These sample sites are illustrated in Fig. 1. Grab samples composed of a number of galena crystals and crystal fragments were taken. As an interesting local isotopic variation became apparent, a single crystal from each of the above three sites was singled out for detailed analyses.

SINGLE CRYSTAL ANALYSES

A large galena crystal from the portal fracture had the form of a cube cut in half along a diagonal plane. The diagonal surface of the crystal was attached to a large breccia fragment of silicified limestone. The crystal was sampled by cutting it into small cubes with a diamond saw. A corner sample (111), two samples (222 and 333), along a diagonal and an edge sample (211) were analyzed. The positions of these samples are indicated in Fig. 2. The isotopic compositions of the samples are listed in Table II. The corner sample (111) was found to be less radiogenic than the other two samples (222 and 333). The edge sample (211) was cleaved into three portions, as shown in Fig. 2. The crystal's outer surface or "skin" was found to be much less radiogenic than the inner portions (see Table II and Fig. 3).

Single galena crystals were selected also from raises R-1 and R-2. These crystals were sampled by cleaving. The portions taken are illustrated in Fig. 2 and comprise three samples from each crystal: an outer skin, the portion of the crystal immediately under the outer skin, and the core of the crystal. The isotopic compositions of these samples are listed in Table II, as are the results of the composite grab-sample analyses from the three sample locations.

The initial sample preparation was carried out at the New Mexico Bureau of Mines. This comprised separation of galena from the gangue and associated ore minerals and the sawing and cleaving of the single crystal samples. Following this initial preparation, the samples were sent to Toronto for analysis. They were dissolved in hydrochloric acid, and the lead precipitated as lead iodide. A Grignard reaction of lead iodide

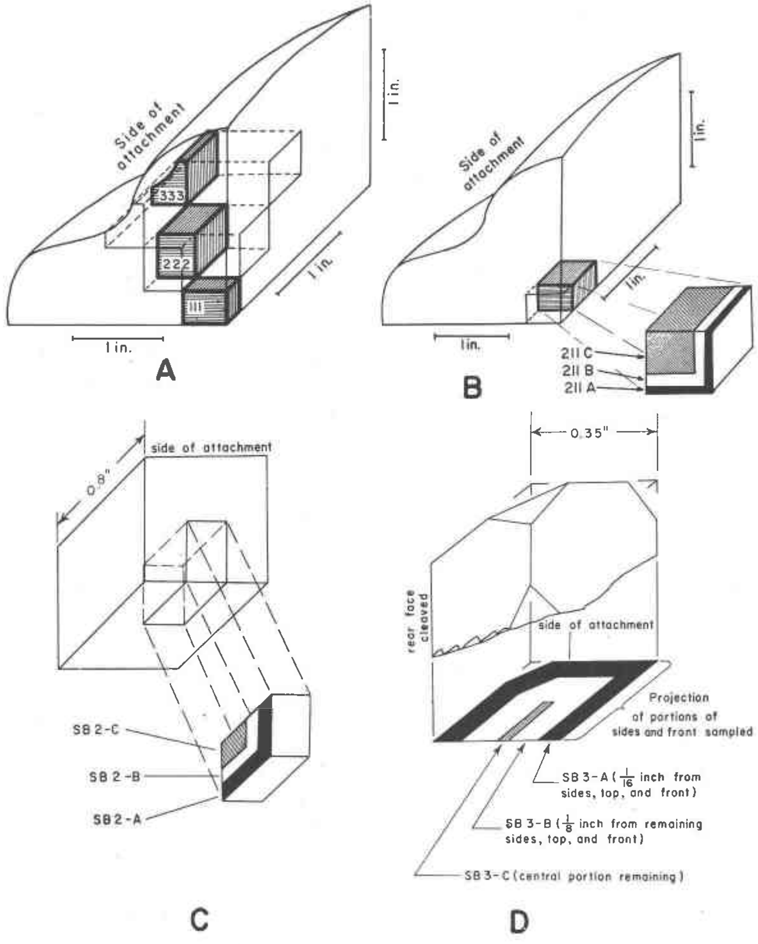

FIG. 2. Sample locations within single galena crystals. A, portal crystal, diagonal sampling; B, portal crystal, edge sampling; C, R-1 crystal; D, R-2 crystal.

and methyl magnesium bromide (CH_3-Mg-Br) at room temperature was used to produce lead tetramethyl. The lead tetramethyl was mass analyzed in the Nier-type 180-degree mass spectrometer operated by the Geophysics Laboratory of the University of Toronto. The analytical precision is shown in Table I.

The samples from the Number 1 entry show a linear relationship when plotted as Pb^{207}/Pb^{204} versus Pb^{206}/Pb^{204}. This is illustrated in Fig. 3, which includes the plots of 22 samples taken from lead deposits in the surrounding 5,000 square miles (Slawson and Austin, 1960). Figure 3 also presents a detailed plot of the single crystal analyses.

TABLE I. PRECISION OF DATA

Replica Analysis of Sample #1040

Date of run	Pb^{204}	Pb^{206}	Pb^{207}	Pb^{208}
July 20, 1959	1.255%	27.85%	20.05%	50.84%
	1.000	22.19	15.98	40.51
Nov. 11, 1959	1.248%	27.83%	20.08%	50.85%
	1.000	22.30	16.09	40.75
Jan. 28, 1960	1.253%	27.85%	20.06%	50.83%
	1.000	22.23	16.01	40.57
Feb. 22, 1960	1.250%	27.83%	20.14%	50.78%
	1.000	22.26	16.11	40.62
June 20, 1960	1.251%	27.83%	20.04%	50.88%
	1.000	22.25	16.02	40.67
July 21, 1960	1.250%	27.87%	20.03%	50.85%
	1.000	22.30	16.03	40.68
Sept. 1, 1960	1.253%	27.87%	20.08%	50.80%
	1.000	22.24	16.02	40.54
Sept. 13, 1960	1.248%	27.83%	20.05%	50.87%
	1.000	22.30	16.06	40.76
Sept. 13, 1960	1.256%	27.83%	20.05%	50.86%
	1.000	22.17	15.97	40.51
Oct. 12, 1960	1.248%	27.79%	20.03%	50.93%
	1.000	22.26	16.05	40.80
Mean *	1.251 ± 0.003	27.84 ± 0.03	20.06 ± 0.03	50.85 ± 0.04
	1.000	22.25 ± 0.04	16.03 ± 0.04	40.64 ± 0.11

Upper row for each run is percent, lower row for each run is ratio with respect to Pb^{204}.

* Sample variation: $S = \left[\frac{\Sigma(x-\bar{x})^2}{n-1}\right]^{\frac{1}{2}}$

INTERPRETATION

Since a linear isotopic relationship is obtained for all leads in both the deposit studied in detail and the surrounding deposits, a common history of origin is indicated for all the isotopic variations encountered. This apparent common history of origin lends much strength to relative age determinations based on local isotopic variations within the deposit, as it indicates that ore fluids with random discrete isotopic compositions have not been active in the region studied.

From the foregoing data (Table II) it appears that the lead isotope composition of the ore fluids definitely underwent measurable changes with time. The mineralizing solutions contained somewhat less radiogenic lead as time progressed, thus causing a decrease in the radiogenic content of the lead deposited.

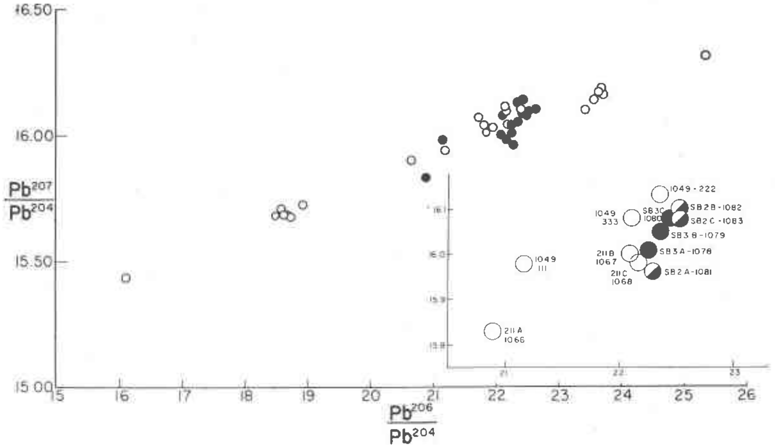

FIG. 3. Regional and detailed lead isotope data. Samples from No. 1 entry show as black circles on large graph.

Detailed Insert: Open circles are from the portal fracture,
Half-filled circles are from the R-1 raise,
Filled circles are from the R-2 raise.

Two theories currently in vogue attempt to explain changes in the isotopic composition of lead ores with continuing deposition. Russell and Farquhar (1960) have emphasized the suggestion that radiogenic lead probably was collected as a contaminant by isotopically homogeneous ore fluids when the fluids passed through whatever country rock intervened between the site of fluid generation and the site of ore deposition. Thus the early fluids that travel through a given fracture or similar permeable zone should extract relatively large quantities of radiogenic lead. Later fluids passing through the same conduits should encounter a decreasing amount of extractable radiogenic material adjacent to the fracture and thus will show corresponding decreases in their radiogenic content as time progresses.

The second theory (Eckelmann, *et al.*) advanced by the geochemists of the Lamont Geological Observatory, is based upon the assumption of an isotopically nonhomogeneous source for the mineralizing solutions. Thus when the ore solutions leave the source area, the readily mobilized radiogenic lead components leave the source in the early solutions, and the radiogenic content of the ore solutions should again decrease with increasing time. (Radiogenic lead, being interstitial and not in rock mineral structures, is believed to be more mobile than that lead tied into mineral structures during the formation of the rocks comprising the fluid source area and/or the wall rocks of the fluid conduits.)

TABLE II. DATA COLLECTED

Location		Toronto number	Data *			
			Pb^{204}	Pb^{206}	Pb^{207}	Pb^{208}
Raise R-2	SB3-A	1078	1.255% 1.000	27.94% 22.26	20.09% 16.01	50.71% 40.41
	SB3-B	1079	1.252% 1.000	27.98% 22.34	20.09% 16.04	50.68% 40.47
	SB3-C	1080	1.249% 1.000	28.02% 22.43	20.07% 16.07	50.66% 40.55
Raise R-1	SB2-A	1081	1.254% 1.000	27.95% 22.29	20.01% 15.96	50.79% 40.50
	SB2-B	1082	1.244% 1.000	28.02% 22.52	20.04% 16.10	50.70% 40.75
	SB2-C	1083	1.245% 1.000	28.04% 22.51	20.03% 16.08	50.68% 40.69
Portal fracture	211-A	1066	1.291% 1.000	26.96% 20.88	20.50% 15.88	51.24% 39.69
	211-B	1067	1.256% 1.000	27.75% 22.09	20.10% 16.00	50.90% 40.53
	211-C	1068	1.252% 1.000	27.74% 22.16	20.01% 15.98	51.00% 40.73
	111	1049	1.282% 1.000	27.13% 21.16	20.49% 15.98	51.10% 39.86
	222	1049	1.244% 1.000	27.80% 22.35	20.07% 16.13	50.88% 40.90%
	333	1049	1.254% 1.000	27.72% 22.11	20.16% 16.08	50.87% 40.57

* Upper row for each sample is percent, lower row for each sample is ratio with respect to Pb^{204}.

The favorable bed (Council Spring limestone) presumably received the ore fluids through the steeply dipping, highly permeable fracture zones that cut this bed. In dealing, however, with interpretations concerning the loci of deposition in a given favorable bed, the actual means of fluid entry into the favorable area is not significant. Probably the entire favorable bed was never fully permeated by ore-bearing fluids. Some areas may have been protected, as the result of sealing by early silica deposition. Other areas may well have received only early solutions and then have remained stagnant for long periods, receiving fresh ore solutions only intermittently as early deposition would change the permea-

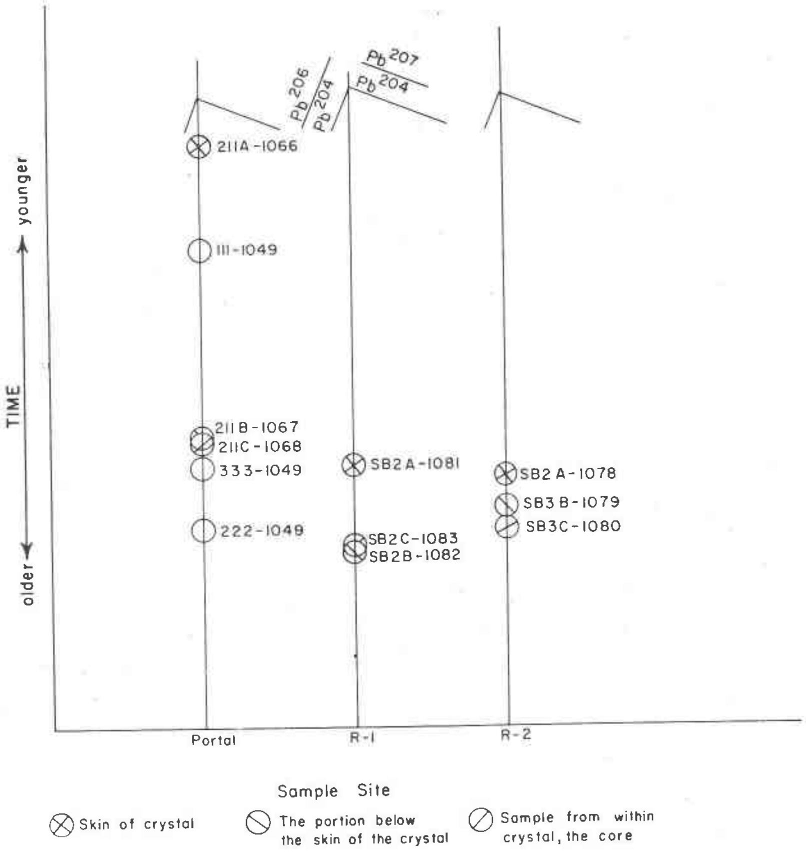

FIG. 4. Time correlation chart for various portions of three galena crystals collected in the No. 1 entry.

bility of surrounding areas. Thus local variations in physicochemical conditions created scattered depositional sites, the location of which shifted with time.

The first lead sulfide that was deposited in the area studied in detail had an isotopic composition about equal to that of sample 1082. At raise R-1 the single crystal studied grew fast enough to retain homogeneity betweeen its core (1083) and the portion immediately below the skin (1082). At raise R-2, however, the rate of crystal growth appears to have been somewhat slower, for the core (1080), the portion below the skin (1079), and the "skin" or outer surface (1078) of the crystal studied show a gradual and fairly uniform change in isotopic composition. The final deposition at sites R-1 and R-2 must have occurred at the same time, as is indicated by the similarity of the composition of the outer-most "skins" of crystals from both areas (1081 and 1078). The most reasonable implication is that physicochemical conditions were varying locally in such a fashion as to cause a temporary cessation of deposition at site R-1 after a brief, initial rapid deposition, whereas deposition continued rather steadily at site R-2.

The deposition of galena along the portal fracture appears to have started at about the same time as R-1 and R-2. The isotopic constitution of sample 1049–333 is not as radiogenic as that found at other places in the inside of the crystal. Owing, however, to the manner of sampling, 333 is not necessarily representative of the true, early core of the crystal. Deposition at the portal fracture site must have been very slow, or else very intermittent, as the outer portion of the crystal sampled is even less radiogenic than the "skins" from crystals at sites R-1 and R-2, thus indicating deposition at a later time. The "skin" of the galena crystal sampled at the portal fracture zone is much less radiogenic than any sample yet analyzed from the Hansonburg district. Thus, it must have been one of the last sites of deposition in the district. Figure 4 is a correlation chart for the three sample sites.

Regardless of the theory of origin adopted, one should keep in mind the ever-present possibility of renewed structural activity that either formed new, unleached fluid conduits or released new fluids into existing conduits, thus causing a resurgence of higher radiogenic lead content in the lead ores deposited later. The lack of cyclic radiogenic variations in the Hansonburg district indicates a stable structural environment following formation of the initial fluid conduits. The entire region appears to have been structurally stable with respect to fluid conduits throughout the time of ore deposition.

The data at hand are equally applicable to the concept of wandering or skipping loci of galena deposition within a fluid-saturated favorable bed, or to the concept of partial fluid saturation of the favorable bed; that is, the wandering of deposition reflects the lack of complete fluid saturation in the favorable bed. In the case of the samples from the portal fracture, the low-radiogenic skin or overgrowth is very definitely younger than the rest of the crystal. There is no evidence to support skeletal growth of such a delicate nature at any site examined in the district. In addition, the crystals from all sites are sufficiently fresh to preclude selective isotope removal through weathering and the sharpness of the crystal edges and corners indicates that leaching by hypogene fluids is unlikely. We can conclude that relative isotopic variations of single crystals within this district yield relative ages of deposition, and thus permit the wanderings of loci of deposition to be traced about the district with respect to time.

In view of the interesting possibilities of using isotopic data as a relative age determinant in the interpretation of the depositional history of a lead deposit, the authors are continuing their isotopic investigations in the Hansonburg district and surrounding vicinity.

ACKNOWLEDGMENT

Support for this project was provided by the New Mexico Bureau of Mines and Mineral Resources and the Geophysics Laboratory of the University of Toronto.

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Manuscript received November 26, 1960.