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UNITED STATES
DEPARTMENT OF THE INTERIOR
GEOLOGICAL SURVEY

URANIUM, THORIUM ISOTOPIC ANALYSES AND URANIUM-SERIES AGES OF CALCITE AND OPAL, AND STABLE ISOTOPIC COMPOSITIONS OF CALCITE FROM DRILL CORES UE25a#1, USW G-2 AND USW G-3/GU-3, YUCCA MOUNTAIN, NEVADA

By

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ABSTRACT

Fracture and cavity filling calcite and opal in the unsaturated zone of three drill cores at Yucca Mountain were analyzed for uranium and stable isotope contents, and were dated by the uranium-series method. Stable isotope data indicate that the water from which the calcite precipitated was meteoric in origin. The decrease in ^{18}O and increase in ^{13}C with depth are interpreted as being due to the increase in temperature in drill holes corresponding to an estimated maximum geothermal gradient of 43° per km. Of the eighteen calcite an opal deposits dated, four of the calcite and all four of the opal deposits yield dates older than 400,000 years and ten of the remaining calcite deposits yield dates between 26,000 and 310,000 years. The stable isotope and uranium data together with the finite uranium-series dates of precipitation suggest complex history of fluid movements, rock and water interactions, and episodes of fracture filling during the last 310,000 years.

INTRODUCTION

Tertiary ash-flow tuffs of Yucca Mountain in southwestern Nevada are being considered as possible hosts for a repository for high-level radioactive waste. Initial examination of exploratory cores revealed that fractures and

cavities are commonly coated and in some cases are completely filled with secondary calcite and opal together with occasional trace amounts of manganese and iron oxides. Apparently these secondary minerals were precipitated from percolating groundwater that became saturated with calcite and silica by leaching wall rocks and fault gouge materials. The purpose of this study is to use the stable isotopic composition of the calcite to determine the origin of the fluids passing through fractures, and to attempt to determine the ages of calcite and opal samples by the uranium-series dating method.

EXPERIMENTAL PROCEDURES

Samples of calcite and opal were collected from the unsaturated zones of three drill holes at Yucca Mountain, Nevada: UE25a#1, USW G-2 and USW G-3/GU-3 (fig. 1). Geologic descriptions and results of geophysical measurements of the drill holes are reported by Hagstrum and others, 1980; Maldonado and Koether, 1983; and Scott and Castellanos, 1984. The thickness of the calcite and opal deposits sampled from these cores vary between about 1 mm and 1 cm. The calcite and opal coating was chipped or scrapped from the bed rock surfaces and fragments showing little or no wall rock contamination were hand-picked for subsequent analyses. Samples containing opal were checked for fluorescence under ultraviolet light. Opals exhibiting yellow-green light emission under ultraviolet stimulus are referred to as uraniferous opal in contrast to other silica which produced no noticeable fluorescence. Samples which were predominantly calcite were separated from occasional uraniferous opal fragments by hand-picking under ultraviolet light illumination. Samples that contained mixtures of calcite and uraniferous opal were collected together and were subjected to acid treatment for separation of calcite from opal, as discussed later.

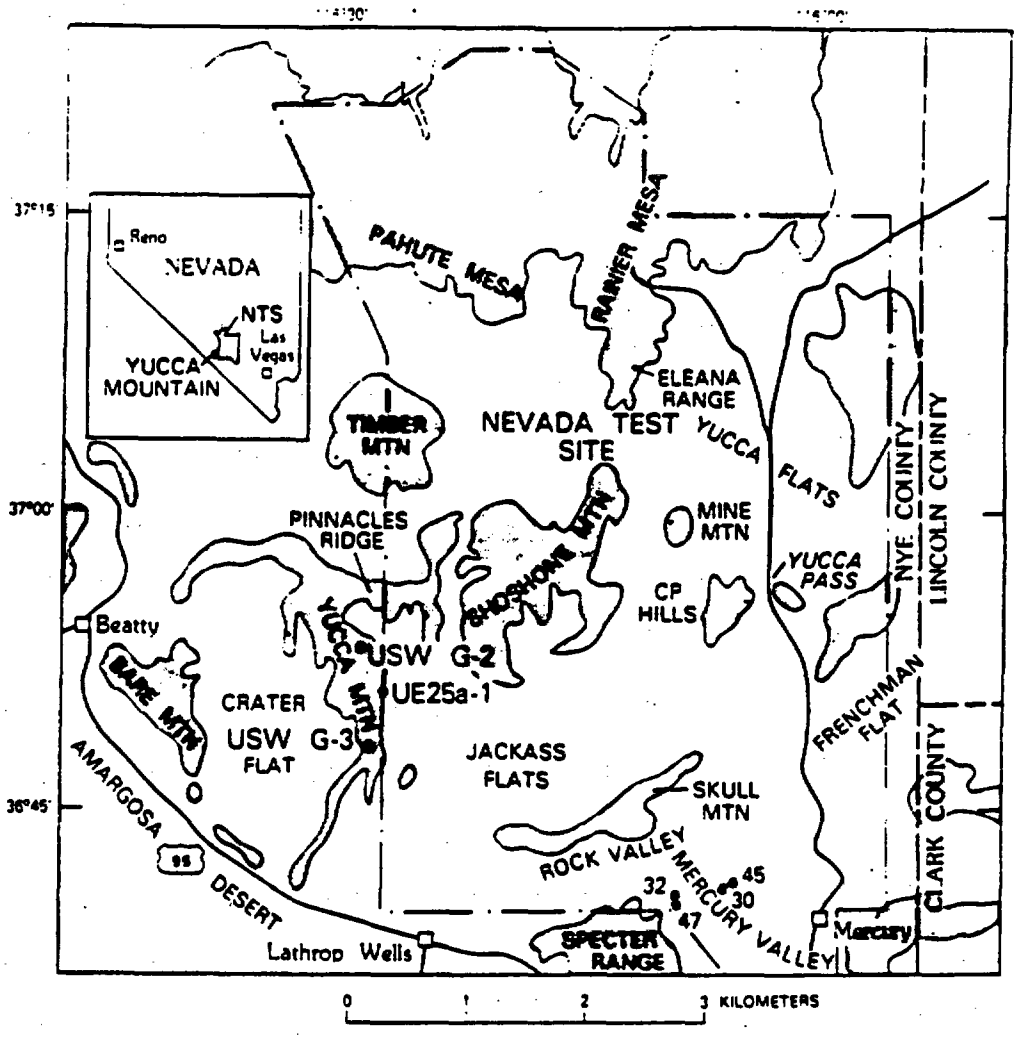

Figure 1. Location of Yucca Mountain, the Nevada Test Site and localities of drill holes.

Portions of calcite from the veins were dissolved in 100 percent phosphoric acid and the released CO_2 gas was analyzed in an isotope ratio mass spectrometer for carbon and oxygen isotopic compositions. The values are reported in the familiar delta notation as the $^{13}C/^{12}C$ or $^{18}O/^{16}O$ ratio in the sample relative to the ratios in a standard such that $\delta^{13}C$ or $\delta^{18}O$ = $(R_{sample}/R_{standard} - 1) \times 1000$ where $R = {}^{13}C/{}^{12}C$ or $^{18}O/^{16}O$. Standard mean ocean water (SMOW) is the standard for oxygen and a belemnite from the Peedee Formation in North Carolina is the standard for carbon. Using these techniques, we obtained a $\delta^{18}O$ value of +7.20 and a $\delta^{13}C$ value of -5.00 for NBS-18 carbonatite, in agreement with other laboratories.

Pure calcite samples selected for uranium-series analysis were ground to a fine powder and heated for a period of about 8 hours at 900°C to convert $CaCO_3$ to CaO. The samples were then dissolved in 8 F (formal) solution of HCl and spiked with ^{236}U, ^{228}Th, and ^{229}Th standard solution. Uranium and thorium isotopes were isolated and purified using an anion-exchange procedure described by Szabo and others (1981). Isolated and purified uranium was electroplated onto a platinum disc. Purified thorium was extracted in a small amount of 0.4 F thenoyltrifluoroacetone (TTA) in benzene and evaporated onto a stainless steel disc. Both discs were counted in an alpha spectrometer.

Mixtures of calcite and opal, or calcite and wall-rock were heated for a period of about 8 hours at 900°C converting calcite to CaO, then were separated by dissolving CaO in dilute nitric acid solutions (0.25 to 1.0 F HNO_3, depending on sample size). The weighed sample was added to continuously stirred solution of nitric acid in small portions to prevent the slurry turning basic. The final acidity was adjusted to about pH 2, and the soluble and insoluble fractions were separated by centrifuging.

The soluble carbonate fraction was spiked with ^{236}U, ^{228}Th, and ^{229}Th and evaporated to a smaller volume. Uranium and thorium were coprecipitated with oxyhydroxides of iron and aluminum by addition of concentrated NH_4OH, then the precipitate was dissolved in 8 F HNO_3 solution. The acid-insoluble fraction (opal and/or wall-rock particles) was spiked with ^{236}U, ^{228}Th, and ^{229}Th and dissolved by repeated refluxing with concentrated HF-$HClO_4$ mixtures. After taken to dryness, the residue was dissolved in 8 F HCl solution. For both fractions, the dissolved uranium and thorium were separated and purified by anion-exchange, and the discs were prepared and measured as described above.

RESULTS AND DISCUSSION

Uranium-series and stable isotopic data for fracture-filling calcite and opal samples from drill holes UE25a#1, USW G-2 and USW G-3/GU-3 at Yucca Mountain are shown in tables 1, 2, and 3. Errors reported for the uranium and thorium measurements are 1 σ propagated errors. All δ values are accurate to ± 0.15 (2 σ).

The uranium concentrations of the calcite, acid-insoluble residues, and uraniferous opal samples vary from about 0.01 to 58 ppm (fig. 2). The uranium concentrations of the carbonate samples range between about 0.01 and 5.0 ppm with the exception of sample 348.8-B which has an unusually high uranium content of 33.3 ppm (table 2). The uranium contents in the residue fractions consisting mainly of secondary silica and bedrock minerals vary between about 0.7 and 7 ppm and the uranium concentrations of the uraniferous opal samples range between about 15 and 58 ppm.

The calculated uranium-series dates of the fracture filling calcite and opal samples from drill holes at the Yucca Mountain area are listed in table 4. The dates of calcite free of acid-insoluble residue and of opal samples

Table 1. Analytical data of fracture filling calcite and acid-insoluble wall rock material from drill hole UE25a#1 at the Yucca Mountain area

Sample depth (m)	Material	Uranium (ppm)	$^{234}U/^{238}U$ (activity ratio)	$^{230}Th/^{232}Th$ (activity ratio)	$^{230}Th/^{234}U$ (activity ratio)	$\delta^{13}C$ (‰)	$\delta^{18}O$ (‰)
34	Calcite	0.767 ±0.015	1.17 ±0.02	2.37 ±0.07	1.02 ±0.04	-1.52	+20.00
87	Fault gouge	5.18 ±0.10	1.09 ±0.02	0.989 ±0.030	1.06 ±0.04	n.a.	n.a.
283	Calcite	5.03 ±0.10	1.47 ±0.02	22.2 ±0.7	1.04 ±0.04	-6.33	+17.60
611	Calcite	3.43 ±0.07	1.29 ±0.02	72 ±3	1.19 ±0.05	-5.41	+15.60

n.a. not applicable.

Table 2. Analytical data of fracture filling calcite, uraniferous opal and acid insoluble residue fractions from drill hole USW G-2 at the Yucca Mountain area

Sample depth (m)	Fraction	Percent carbonate	Uranium (ppm)	$^{234}U/^{238}U$ (activity ratio)	$^{230}Th/^{232}Th$ (activity ratio)	$^{230}Th/^{234}U$ (activity ratio)	$\delta^{13}C$	$\delta^{18}O$ (‰)
280	Calcite	>99	0.500 ±0.010	1.032 ±0.015	12.0 ± 2.4	1.023 ±0.041	-8.35	+19.21
302	Calcite	n.d.	n.d.	n.d.	n.d.	n.d.	-7.90	+19.31
346.7	Calcite	64	0.405 ±0.008	1.167 ±0.018	4.29 ±0.21	0.915 ±0.037	-7.43	+18.22
	Residue	0	6.85 ±0.14	1.135 ±0.017	2.43 ±0.10	0.965 ±0.049	n.a.	n.a.
346.8	Calcite	n.d.	n.d.	n.d.	n.d.	n.d.	-7.37	+18.30
348.7	Calcite	>99	0.073 ±0.006	1.02 ±0.03	4.6 ±0.5	0.73 ±0.06	-7.47	+18.19
348.8-A	Calcite	97	0.136 ±0.004	0.937 ±0.028	10.7 ±1.6	1.010 ±0.040	-6.93	+18.13
348.8-B	Calcite	63	33.3 ±0.7	1.026 ±0.015	94 ±15	0.093 ±0.037	n.d.	n.d.
	U Opal	0	57.8 +1.2	1.031 +0.015	232 +34	1.027 +0.031	n.d.	n.d.
359-A	Calcite	61	1.21 ±0.06	1.020 ±0.015	261 ±80	0.795 ±0.032	-6.82	+17.98
359-B	Calcite	75	0.644 ±0.013	0.965 ±0.014	36 ±11	0.811 ±0.032	n.d.	n.d.
	U Opal	0	27.0 +0.5	1.068 ±0.016	234 ±70	1.04 ±0.04	n.d.	n.d.
361	Calcite	n.d.	n.d.	n.d.	n.d.	n.d.	-6.56	+17.77

n.d. not determined

n.a. not applicable

Table 3. Analytical data of fracture filling calcite, uraniferous opal and acid insoluble residues from drill hole USW G-3/GU-3 at the Yucca Mountain area

Sample depth (m)	Fraction	Percent carbonate	Uranium ppm	$^{234}U/^{238}U$ activity ratio	$^{230}Th/^{232}Th$ activity ratio	$^{230}Th/^{234}U$ activity ratio	$\delta^{13}C$ (‰)	$\delta^{18}O$ (‰)
63	Calcite	95	0.553 ±0.011	2.26 ±0.03	35 ±4	1.00 ±0.03	-7.06	-20.23
	Residue	n.a.	0.66 ±0.04	1.37 ±0.10	7.6 ±1.5	1.94 ±0.19	n.a.	n.a.
131	Calcite	52	3.02 ±0.06	1.43 ±0.02	84 ±40	0.216 ±0.009	-5.11	+20.16
	U Opal	n.a.	35.0 ±0.7	1.13 ±0.02	473 ±190	1.15 ±0.05	n.d.	n.d.
147	Calcite	n.d.	n.d.	n.d.	n.d.	n.d.	-5.58	+20.04
159	Calcite	n.d.	n.d.	n.d.	n.d.	n.d.	-5.44	+20.28
318	Calcite	95	0.0836 ±0.0017	0.991 ±0.020	2.58 ±0.13	1.10 ±0.06	-5.10	+19.11
	Residue	n.a.	1.98 ±0.11	0.73 ±0.07	4.89 ±0.73	1.27 ±0.13	n.a.	n.a.
331	Calcite	87	0.36 ±0.01	1.06 ±0.04	10 ±5	0.24 ±0.02	-4.54	+18.73
	U Opal	n.a.	14.9 ±0.4	1.05 ±0.03	153 ±61	1.13 ±0.06	n.d.	n.d.

n.a. not applicable

n.d. not determined

Table 4. Calculated uranium-series ages of fracture filling calcite and opal from drill holes at the Yucca Mountain area, Nevada

Drill hole	Depth (ft)	Uranium (ppm)	Calculated age ($\times 10^3$ years)
		Calcite	
UE25a#1	34	0.77	3^{+80}_{-30} (a)
GU-3	63	0.56	227±20
GU-3	131	3.0	26±2
USW-G2	280	0.50	>400
UE25a#1	283	5.0	310^{+70}_{-60}
GU-3	318	0.084	>400
GU-3	331	0.36	30±4
USW-G-2	346.7	0.40	190±20 (b)
USW-G-2	348.7	0.073	142±30
USW-G-2	348.8-A	0.14	>400
USW-G-2	348.8-B	33	280±70
USW-G-2	359-A	1.2	170±18
USW-G-2	359-B	0.64	185±18
UE25a#1	611	3.4	>400
		Uraniferous Opal	
GU-3	131	35	>400
GU-3	331	15	>400
USW-G-2	348.8-B	58	>400
USW-G-2	359-B	27	>400

a. Isochron-plot corrected using analytical data of fault gouge.

b. Isochron-plot corrected using analytical data of both acid-soluble carbonate and acid-insoluble residue.

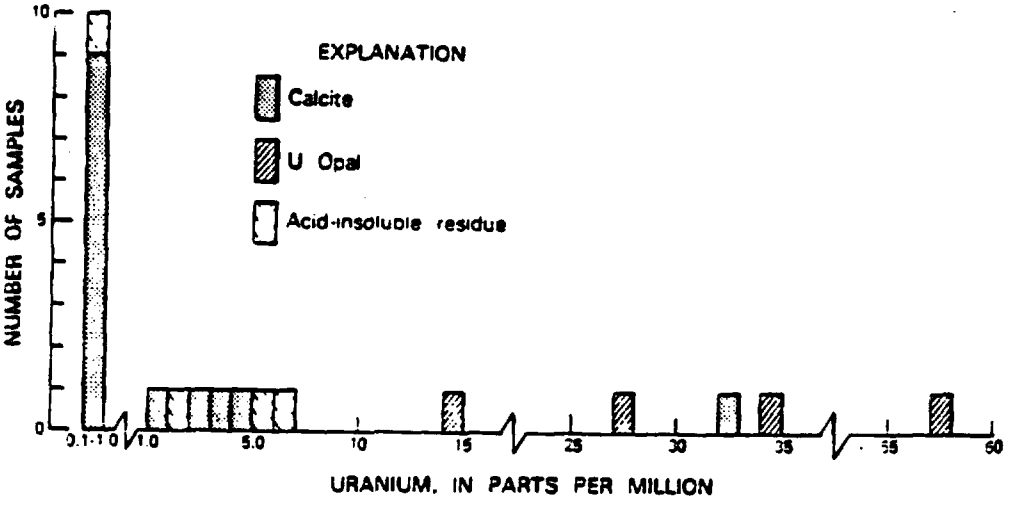

Figure 2. Histogram of uranium concentrations in calcite, uraniferous opal and acid insoluble residues.

Table 4. Calculated uranium-series ages of fracture filling calcite and opal from drill holes at the Yucca Mountain area, Nevada

Drill hole	Depth (m)	Uranium (ppm)	Calculated age ($\times 10^3$ years)
		Calcite	
UE25a#1	34		310^{+80}_{-50} (a)
GU-3	63	0.56	227±20
GU-3	131	3.0	26±2
USW-G2	280	0.50	>400
UE25a#1	283	5.0	310^{+70}_{-60}
GU-3	318	0.084	>400
GU-3	331	0.36	30±4
USW-G-2	346.7	0.40	190±20 (b)
USW-G-2	348.7	0.073	142±30
USW-G-2	348.8-A	0.14	>400
USW-G-2	348.8-B	33	280±70
USW-G-2	359-A	1.2	170±18
USW-G-2	359-B	0.64	185±18
UE25a#1	611	3.4	>400
		Uraniferous Opal	
GU-3	131	35	>400
GU-3	331	15	>400
USW-G-2	348.8-B	58	>400
USW-G-2	359-B	27	>400

a. Isochron-plot corrected using analytical data of fault gouge.

b. Isochron-plot corrected using analytical data of both acid-soluble carbonate and acid-insoluble residue.

are calculated from their measured $^{230}Th/^{234}U$ activity ratios using standard radioactive growth and decay equations assuming that the authigenic calcite and opal had remained ideal closed systems with respect to the isotopes of uranium and thorium since their formation. In contrast, the dates for impure calcite samples were calculated using the results for the acid-soluble and acid-insoluble fractions in a pseudo-isochron-plot method described by Szabo and others (1981), and Szabo and Rosholt (1982).

Uranium-series dates of fracture-filling calcite samples range from 26,000 to >400,000 years and uranium-series dates of uraniferous opal samples are all older than 400,000 years. The dates are plotted against their corresponding depths in fig. 3. Depth intervals lacking dated samples, however, may not be real because we arbitrarily selected the thicker deposits best suited for dating. Two calcite samples yield an average date of 28,000 years, four calcite deposits yield an average date of about 170,000 years and four calcite deposits have an average date of about 280,000 years. In addition, four calcite and four uraniferous opal deposits are yielding minimum dates of precipitation of greater than 400,000 years. The distribution of the finite calcite dates suggests fluid movement and fracture and cavity filling in these drill holes between 26,000 and 310,000 years.

The distribution of uranium in uraniferous opal at 348.8 m depth in drill hole USW G-2 was determined by fission-track mapping. A photomicrograph of the uraniferous infilling in the host calcite is shown in fig. 4-A and the corresponding fission-track image is displayed in fig. 4-B. The dark areas in fig. 4-B correspond to localities of uraniferous opal, the lighter-colored area are the partially crystallized opal of lower uranium content, and the lightest area is the host calcite of the lowest uranium content. Bulk uranium analysis of the separated calcite (0.136 ppm U-sample 348.8-A, table 2) and of

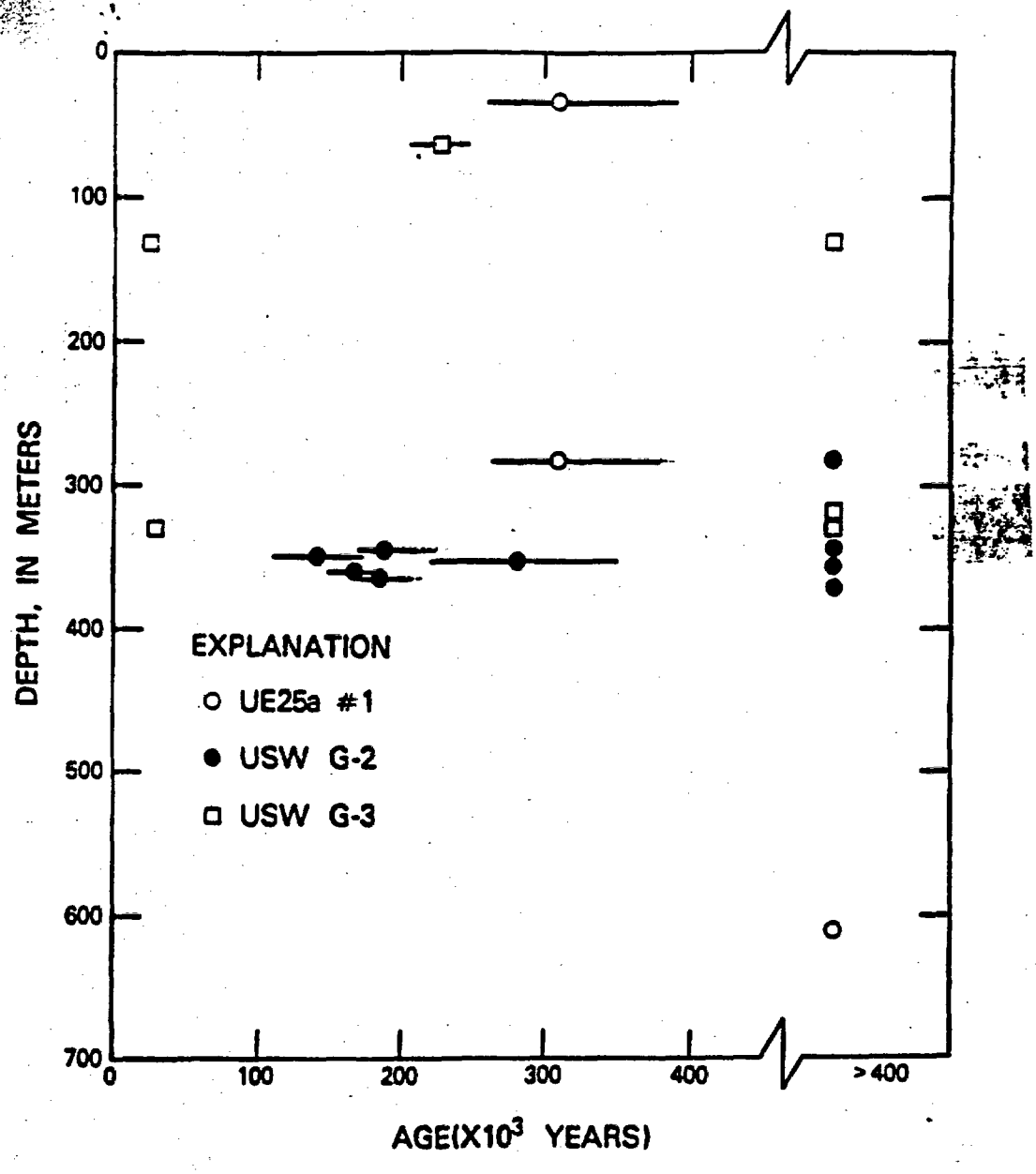

Figure 3. Calculated uranium-series dates ($x10^3$ years) of fracture and cavity filling calcite and opal in drill holes area plotted against corresponding depths in meters. Bars indicate experimental errors.

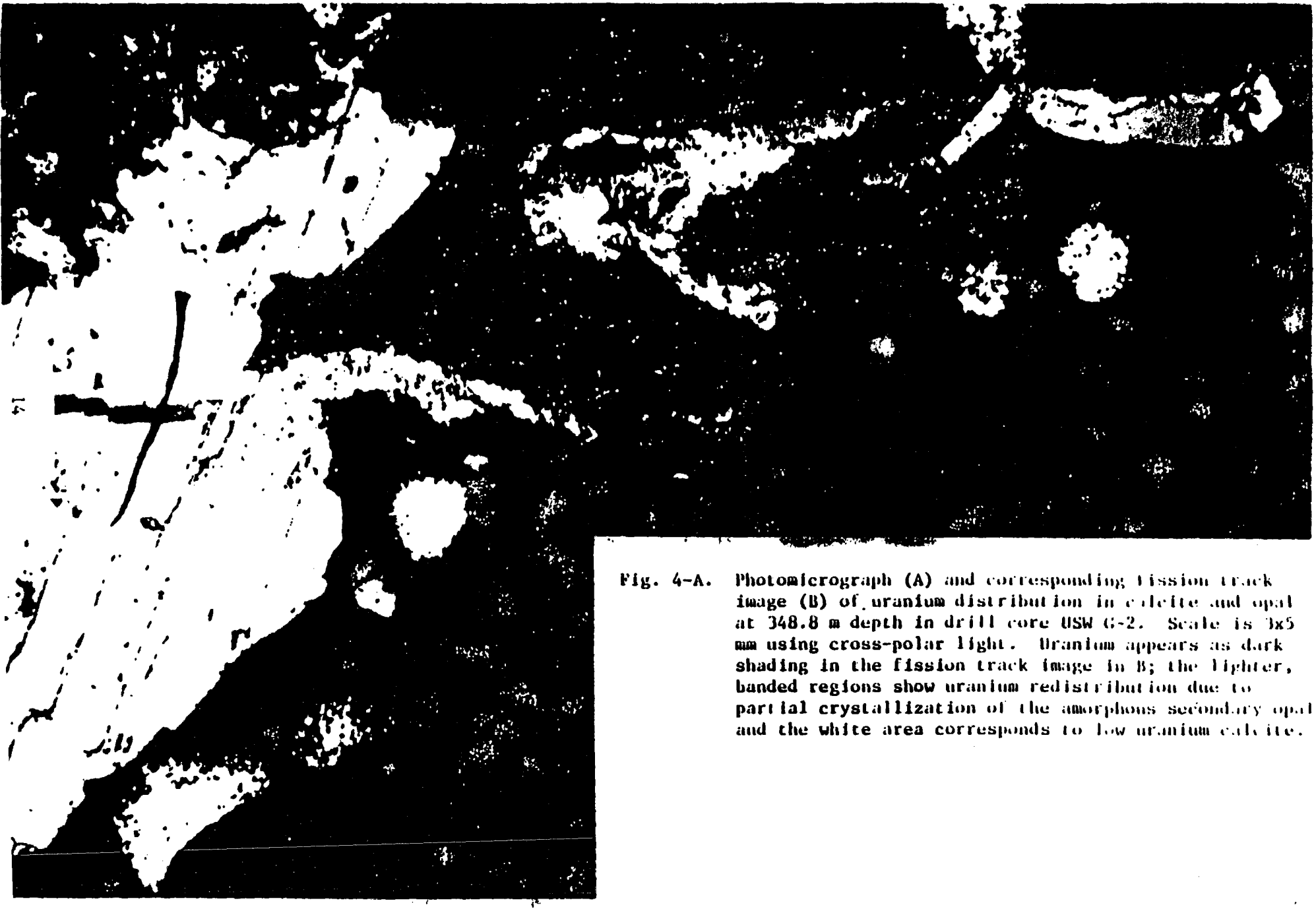

Fig. 4-A. Photomicrograph (A) and corresponding fission track image (B) of uranium distribution in calcite and opal at 348.8 m depth in drill core USW G-2. Scale is 3x5 mm using cross-polar light. Uranium appears as dark shading in the fission track image in B; the lighter, banded regions show uranium redistribution due to partial crystallization of the amorphous secondary opal and the white area corresponds to low uranium calcite.

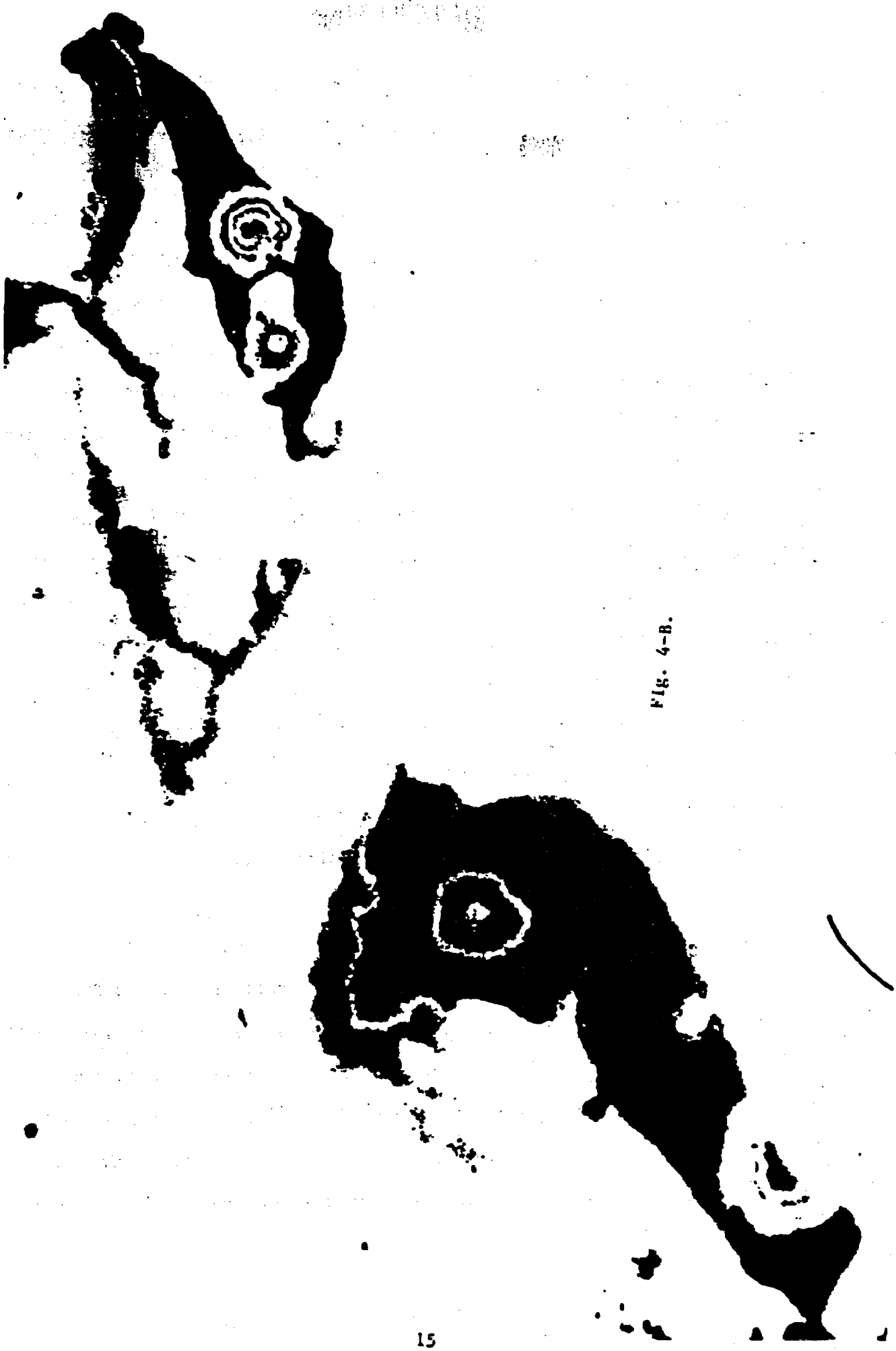

Fig. 4-B.

opal (57.8 ppm U-sample 348.3-3, table 2) confirms that nearly all uranium is in the silica phase. However, significant amounts of uranium may be lost from the amorphous opal by the process of crystallization.

Examination of the stable isotopic composition of calcites reveals no obvious correlation between the $\delta^{18}O$ values and dates obtained for the samples. The absence of such a correlation indicates that the samples have not been subjected to systematic diagenesis or that significant change with time in the isotopic composition of the fluids which precipitated the calcite had not occurred. Also because the isotopic composition of the fluids did not apparently change with time, the $\delta^{18}O$ value of the calcites can be used to constrain the isotopic composition and the origin of the fluid. The difference in $\delta^{18}O$ values of co-existing calcite and water at 20°C is about 30 per mil (O'Neil and others, 1969) so that the water in equilibrium at this temperature with the shallowest calcites with $\delta^{18}O$ of about +20 (fig. 5) would have $\delta^{18}O$ values near -10. This value is normal of modern meteoric water of this area and suggests that the source for the oxygen in these fracture-filling calcites is meteoric water. The fluids which precipitated the deeper, more ^{18}O-depleted calcites (fig. 5), had either lower δ^{18} values or experienced higher temperatures than those which equilibrated with the shallow samples.

The $\delta^{13}C$ values in drill core calcites of the Yucca Mountain range between about -4 and -8 (fig. 6). There are two probable sources for the carbon in these secondary calcites. One source is dissolved atmospheric CO_2 in the groundwaters which would produce $\delta^{13}C$ values near -1 in carbonates similar to those from the nearby Amargosa Desert (Kyser and others, 1981). The other source is reduced carbon from either organic matter or from the tuffs themselves; the δ^{13} value of these sources may approcah -20. The $\delta^{13}C$

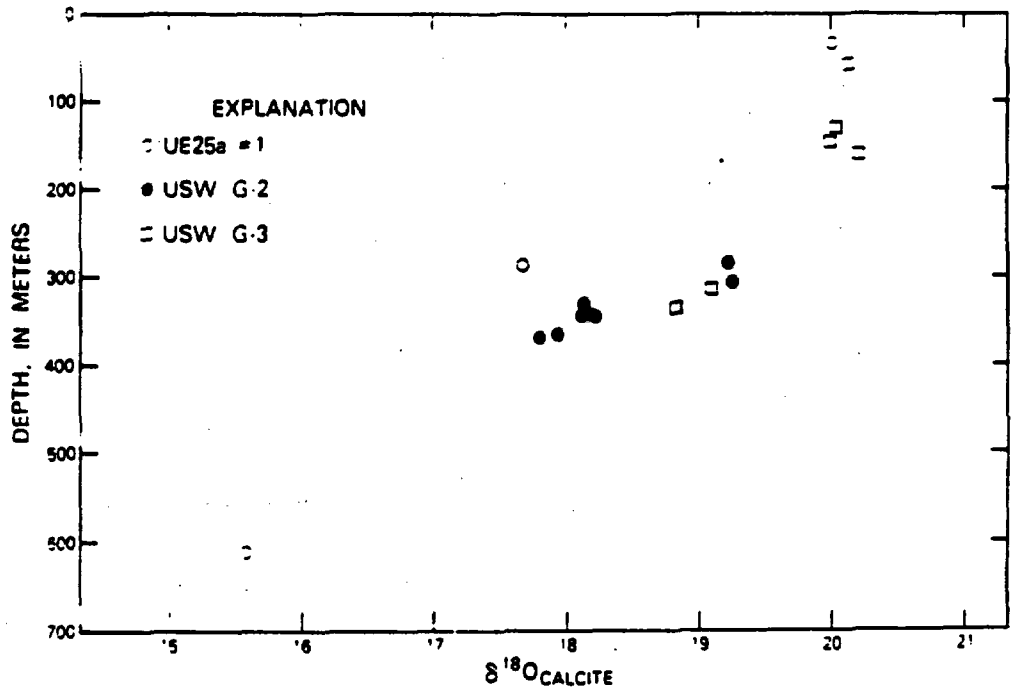

Figure 5. $\delta^{18}O$ values of calcite from drill holes and surface travertines are plotted against depth of sample.

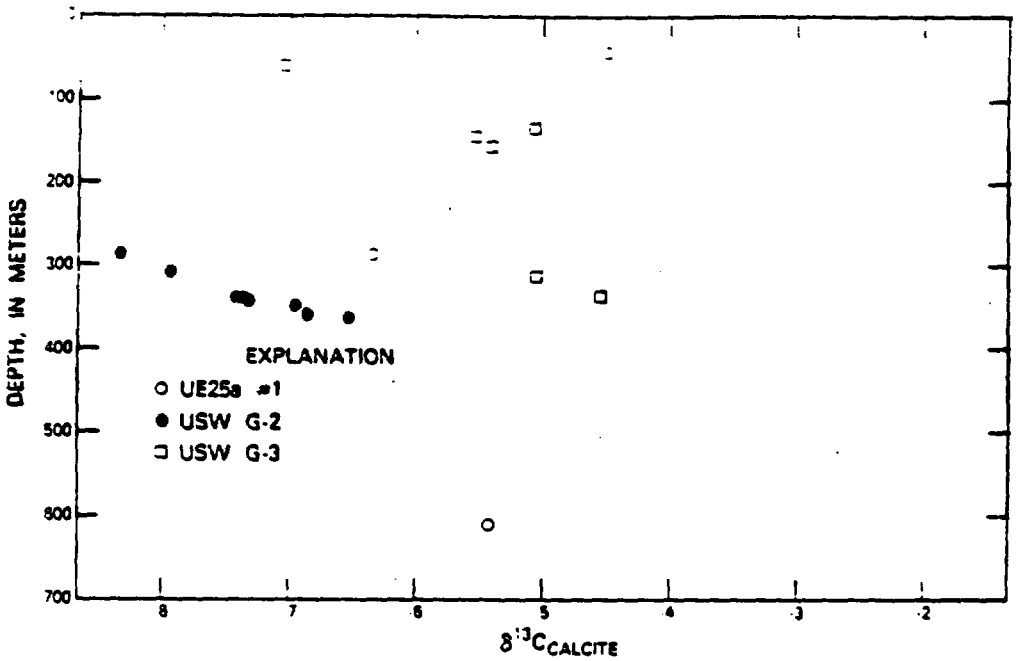

Figure 6. Relationship between the $\delta^{13}C$ value and depth of calcite from the Yucca Mountain area.

values of -4 to -8 per mil suggest that most of the carbon comes from dissolved atmospheric CO_2.

The $\delta^{18}O$ values decrease consistently with depth in all three drill holes (fig. 5). In contrast, the $\delta^{13}C$ values of calcites increase with depth (fig. 6). However, samples from drill holes USW G-3 have higher ^{13}C values than do those from USW G-2 at similar depths and the $\delta^{13}C$ values of drill hole UE25a#1 do not fit well the trends of the other drill holes even if $\delta^{13}C$ result of the shallowest sample (34 m, table 1) is disregarded. The general decrease of $\delta^{18}O$ and increase of $\delta^{13}C$ with depth of the calcites may result from several processes including (1) enrichment of ^{18}O and ^{12}C in the fluids which produced the shallowest samples through evaporation of ^{18}O-poor water and loss of ^{13}C-rich CO_2 from ascending deeper fluids, (2) depletion of ^{18}O and ^{12}C in the deeper fluids as a result of continued precipitation of calcite, (3) production of each calcite from fluids having different origins, and (4) increase in the temperature of precipitation of calcite with depth as the same fluid responds to the local geotherm.

Although the climate of the Yucca Mountain area is conducive to evaporation at the surface, the regular decrease in the $\delta^{18}O$ values of the calcites to depths of several hundred meters in conjunction with the high degrees of evaporation (>50%) needed to produce the required appropriate isotopic modification of the deep fluid suggests that this mechanism is unlikely. Similarly, the quantity of calcite that would have to precipitate from the shallow fluids to produce the appropriate $\delta^{18}O$ values of the deeper fluids should produce much greater variation of the $\delta^{13}C$ values than is observed. The regular increase in the $\delta^{18}O$ and decrease in the $\delta^{13}C$ values of the calcites with depth argue against distinct fluids for each calcite.

The isotopic fractionation in response to increasing temperature appears to best explain the depth-related trends in the stable isotope data of the calcites in these drill holes. Using the oxygen isotope fractionation factor between calcite and water as reported by O'Neil and others (1969) and assuming a $\delta^{18}O$ value for meteoric water of -9, the measured $\delta^{18}O$ values of the Yucca Mountain calcites suggest equilibration temperatures at about 20°C near the surface and at about 46°C for the sample at a depth of 611 meters (fig. 7). The corresponding enrichment of ^{13}C with depth in calcites from the same drill holes can also be explained by increasing the temperature at which the calcite precipitates. These estimates of the changes of temperature with depth correspond to a maximum geothermal gradient of 43°/km. This value represents an upper limit of the gradient because the descending meteoric water can also become ^{18}O-depleted and ^{13}C-enriched as a result of the precipitation of the carbonate.

Sass and others (1980) estimated thermal gradients of about 36°/km for depths to 470 meters in hole UE25a#1 and mean annual ground-surface temperatures of 14.8°C. A gradient of 45°/km was estimated for another hole (UE25a#3) about 12 km from UE25a#1 (Sass and others, 1980). These gradients agree well with that estimated from the isotopic composition of calcite in the drill holes although we assumed a surface temperature of precipitation of the calcite of about 20°C. Sass and Lachenbruch (1982) also report the possible downward percolation of ground water through both unsaturated and saturated zones at a rate on the order of 1 to 10 mm/year in the Yucca Mountain area. The stable isotope data presented here suggest that meteoric water moved downward from the surface along fractures and precipitated calcite in near equilibrium with the geotherm.

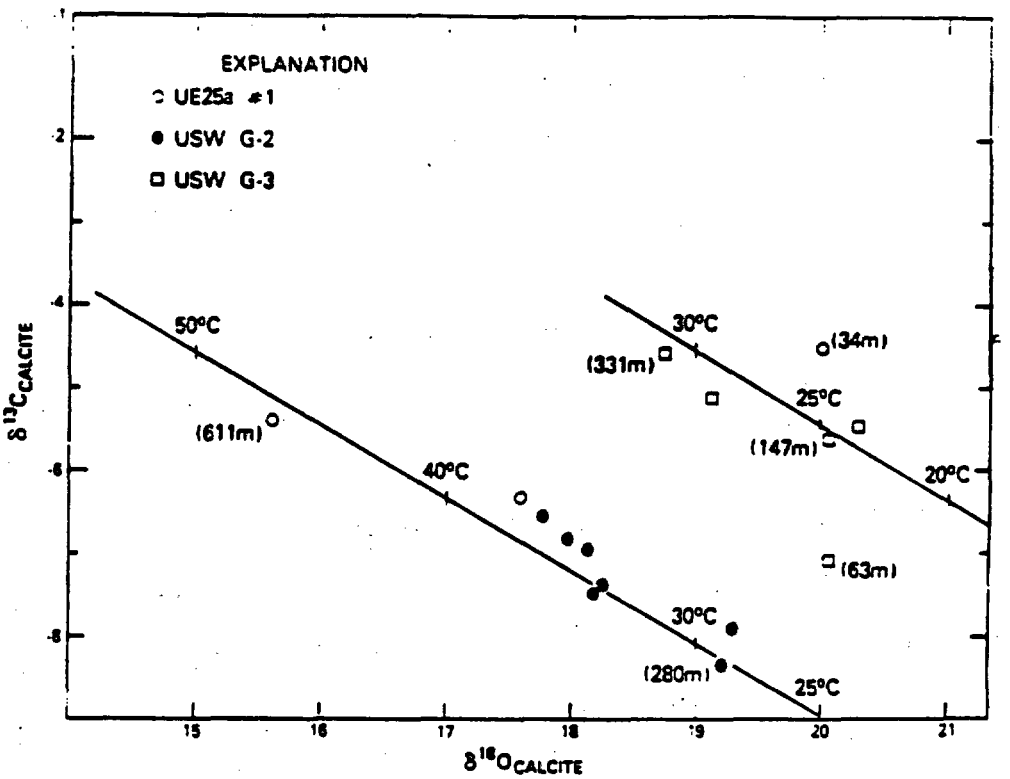

Figure 7. $\delta^{18}O$ versus the $\delta^{13}C$ values of calcite from Yucca Mountain. Lines and temperatures represent the calculated isotopic composition of calcite in equilibrium with meteoric water having a $\delta^{18}O$ value of -9 and $\delta^{13}C$ values of either -11.5 (lower line, hole USW G-2) or -8 (upper line, hole USW G-3). Fractionation factors for oxygen are those suggested by O'Neil and others (1969) and for carbon are those derived by Friedman 1970). The depth of some of the samples in meters are indicated in parentheses.

SUMMARY

Stable isotope data indicate that fracture and cavity filling calcite in Yucca Mountain drill cores precipitated from downward migrating meteoric water. The observed decrease in ^{18}O and increase in ^{13}C contents of calcite deposits with respect to depth (fig. 7) are interpreted as being primarily due to increase in temperature of the water moving downward along fractures in the unsaturated zone. The changes of temperature with depth, deduced from measured $\delta^{18}O$ values, correspond to a maximum geothermal gradient of 43°/km.

The percolating ground water leached uranium and other elements from wall-rocks becoming saturated with respect to calcite and silica. Subsequently, various amounts of dissolved uranium coprecipitated with calcite and opal. Some of these deposits in three drill holes were selected for dating by the uranium-series method. All dates are calculated by assuming a closed system, and single generation with rapid rates of accumulation of the fracture filling deposits. Of the eighteen samples dated, four calcite deposits and all four of the opal deposits are older than the detection limit of the method of about 400,000 years, and ten calcite deposits yield dates between 26,000 and 310,000 years (table 4). The finite dates of precipitation indicate fluid movement and fracture filling during the last 310,000 years.

Generalizing on a limited data base, the migration of meteoric water and subsequent calcite precipitation are probably tectonically controlled. Fractures caused by tectonic processes allowed ground water to move until the openings became obstructed by precipitating calcite. The obtained dates for calcite precipitation are then minimum ages for tectonic episodes. Another possible interpretation of the results is that water migration and calcite precipitation were climatically regulated. That is, ground water movement and calcite formation occurred during period of high rainfall. Three groups of

calcite dates may be recognized from our data which may correspond to pluvial periods at the Yucca Mountain area: two calcite deposits yield an average date of 28,000 years, four calcite deposits yield an average date of about 170,000 yers. and the average date of four other calcite deposits is about 280,000 yers (fig. 3). Alternatively, the finite calcite dates may be the result of open-system modification. In this explanation, all calcite deposits are presumed to be older than 400,000 years. Crystallization of opal then released additional uranium which was incorporated in some of the calcite deposits making them to appear too young. As presented in fig. 4-B, crystallized opal coexisting with calcite exhibits uranium redistribution and possibly uranium loss as a function of recrystallization, but there is no indication of secondary uranium uptake by the lowest-uranium containing host calcite (sample 348.8-A in table 2).

In conclusion, this preliminary investigation of calcite and opal deposits in fractures of Yucca Mountain drill cores combining uranium-series dating and stable isotope data show promise to warrant a more systematic sampling. Refinements of the procedure in a future study should include identification of possible multiple generation of calcite, determination of relationship betwen coexisting calcite and opal, and searching for evidence of secondary uranium migration by using thin-sections, luminescence observations and fission-track radiography.

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