THE DISTRIBUTION OF HELIUM AND RADIOACTIVITY IN ROCKS.

III. RADIOACTIVITY AND PETROLOGY OF SOME CALIFORNIA INTRUSIVES.

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ABSTRACT. As a part of an extensive petrologic study of granite intrusives in Southern California, helium and radioactivity determinations were made on minerals separated from three fresh specimens. The radioactivity of the accessory minerals is found to be from 100 to 1000 times higher than those of the common rock minerals. The helium retentive properties in decreasing order are hornblende, biotite, quartz, apatite, feldspar, sphene, and zircon. The assignment of Jurassic age is found to be the most consistent with the corrected helium age determinations and with field data.

INTRODUCTION.

THE naturally-occurring radioactive elements are unique in being important to both the geochemical and the geophysical aspects of experimental geology. Chemically and mineralogically, they are of interest as a part of the earth's multi-component chemical system. We wish to learn more of their interactions within the magma, the course of their separation in various minerals, and the chemical reactions involved in their concentration in crystallized rocks and veins.

Physically, the heaviest elements are important because of their spontaneous nuclear disintegration, which generates relatively huge quantities of heat, and which provides a clock mechanism for determining the time since crystallization.

Because Southern Californian granitic rocks had been studied by one of us (E. S. L.) in some detail in the field, the laboratory, and by spectrographic methods, and samples were being used for other geophysical investigations at Harvard University, it seemed desirable to determine the mineralogical associations of radioactivity in these rocks, and at the same time to make helium determinations in order to ascertain the relative helium retentivity of various minerals.[3, 4, 5]

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[4] Keevil, N. B.: 1940, Proc. Am. Acad. Arts & Sci. *73*, 311-359.
[5] Keevil, N. B.: 1941, Am. Geophys. Union, N. R. C., II, 501-503.

The Lakeview tonalite seemed well suited for such an investigation because of the ease with which eight of the more common minerals could be separated. For comparison, two other granitic rocks from the batholith of Southern California were included.

To study the distribution of radioactivity within granitic rocks seemed most important because earlier results from radioactivity determinations and studies of the occurrence of radioactive minerals have indicated their concentration in such rocks and their associated pegmatites. While petrographic investigations have supplied some information, more accurate knowledge of the chemical and mineralogical relationships seemed desirable. A comprehensive program has, therefore, been undertaken to supplement limited petrographic tools with sensitive analytical instruments for the purpose of studying the petrology of the very minor constituents. For general purposes, petrographic methods prove most useful, but in the case of radioactive elements the analytical sensitivity of atom-counting instruments is unsurpassed. This opens a new and promising field in the very low concentration range. The petrologic significance of such investigations is not limited entirely to the radioactive elements, for these are closely allied with other heavier elements, such as tantalum, vanadium, tungsten, and the rare earths, which separate with them in late magmatic processes, and form isomorphous and mixed minerals according to their spatial similarities (ionic size) rather than according to strictly chemical or periodic relationships.

GEOLOGY AND PETROGRAPHY.

The three rocks examined came from the area a few miles south and southeast of Riverside in the northern part of the Elsinore quadrangle and are from the northern part of the great batholith of Southern California, which is at least 400 miles long and 70 miles wide. In Southern California the following sequence of events indicates that the batholith is probably late Jurassic or early Cretaceous in age:

Deposition of Triassic sediments (fossiliferous)
Deformation and mild metamorphism
Erosion
Accumulation of a great pile of volcanic rocks
Deformation and mild metamorphism

Intrusion of the complex batholith in a large number of separate injections. Early injections were largely or completely crystalline before the next major magma was injected, indicating a considerable interval of time between injections

Erosion

Deposition of late Cretaceous sediments

The batholith of the Sierra Nevada, which is probably a northern extension of the batholith of Southern California, has been considered by Hinds to be of late Jurassic age.[6]

The batholith of Southern California in Mexico, only 200 miles south of the area studied, has been reported by Böse and Wittich[7] to intrude fossiliferous rocks of lower Upper Cretaceous age and to be unconformably overlain by rocks of the late Upper Cretaceous age.

The three rocks selected for study in this paper are all from bodies that were intruded rather early in the successive intrusions that make up the complex batholith—probably a little before the middle of the succession.

El 38-134 is from the Lakeview tonalite, one of the earliest intrusive bodies. It was collected from a small quarry four miles north-northeast of Perris, on the north slope of the northeastern of three hills surrounded on the topographic map of the Elsinore quadrangle by the 1700-ft. contour.

El 38-29 is from the Bonsall tonalite, which is much like the Lakeview tonalite, except that it carries abundant flattened inclusions. It is slightly younger than the Lakeview. The specimen was collected from shaft 2 of the Los Angeles aqueduct, two miles west of Valverde.

El 38-23 is from a small granodiorite dike, 250 feet across, which cuts and is believed to be satellitic to the Bonsall tonalite. Collected from a small quarry which is one quarter of a mile northwest of Bench Mark 1651, which is two miles west-southwest of Valverde.

[6] Hinds, N. E. A.: 1934, The Jurassic age of the last granitoid intrusives in the Klamath Mountains and Sierra Nevada, California, Am. Jour. Sci. Vol. 27, p. 182-192.

[7] Böse, E. and Wittich, E.: 1913, Informe relativo a la exploracion de la region norte de la costa occidental de la Baja California, Parergones del Inst. Geol. de Mexico, Vol. 4, p. 307-529.

Description of the Samples.

The percentage of the various minerals present in each rock was determined by Rosiwal determinations on one or two large, thin sections (area 500 to 600 mm.). Such determinations made on rocks as coarse-grained as the two tonalites may be several per cent in error, due to the fact that the thin sections do not sample the rocks. Mineral analyses were therefore made on a part of the powder by separating into fractions, each of which is made up of a relatively pure mineral and by estimating the impurities in the samples with the microscope. By this method the probable error in the estimate should be about one per cent.

The mineral compositions of the three rocks are given in Table 1.

TABLE 1.

Mineral Compositions (weight per cent) of three rocks from California.

	Lakeview tonalite E1 38-134	Bonsall tonalite E1 38-29	Granodiorite dike E1 38-23
Quartz	20	22	23
Orthoclase			19
Plagioclase	49	52	52
Biotite	18	15	4
Hornblende	13	9	
Magnetite	1	1	1
Zircon	0.02		
Apatite	0.2		
Sphene	0.04		
An. content of plagioclase	49	37	30

To avoid loss of helium only that part of the sample coarser than 40-mesh was used in the mineral separation. Clean separations were, therefore, not possible, and, in particular, some inclusions of the accessory minerals, apatite and zircon, remained in the silicate minerals. The magnetite separations were especially impure.

All three of the rocks were rather fresh but contained a little sericite, epidote, and chlorite. All contained very small amounts of pyroxene, chiefly hypersthene.

Preparation of Samples.

The rocks were carefuly sampled to avoid the entrance of radioactive contamination. Chips were selected from the centers of large specimens in each case, and pulverized in steel mortars previously cleaned with a "blank" rock, and followed by a portion of the rock to be sampled. The mineral separations were made with bromoform, methylene iodide, and an electromagnet. Washing was done with acetone. The separatory funnels and glassware used were new and the bromoform was fresh from the manufacturer. Care was taken to avoid contamination. To test the amount of contamination in sampling and in mineral separation, blank determinations were made, with negative results.

The compositions of the mineral separates were estimated with the microscope and are given in Table 2. In the samples made up of quartz and oligoclase, determinations of total SiO_2 were made to determine the amount of quartz.

TABLE 2.

Composition of mineral separates in weight per cent.

	Quartz	Orthoclase	Plagioclase	Biotite	Hornblende	Magnetite	Chloride		
El 38-134									
Quartz	95		5						
Plagioclase (1)	2		98	tr.	tr.				
Plagioclase (2)	5		95						
Plagioclase (3)	1		99						
Biotite	1		1	97	tr.		1		
Hornblende			1	1	97			Epidote 1	
Magnetite	1		3	2	38	55		Epidote 1	
Apatite	2							Zircon tr.	
Zircon								Apatite 1	Sphene 1
Sphene								Apatite 2	Zircon 2
El 38-29									
Biotite	4		7	89					
El 38-23									
Quartz and feldspar	45		55						
Quartz and feldspar	52		48						
Feldspar and quartz	20		80						
Biotite			5	95					

HELIUM, RADIOACTIVITY, AND AGE DETERMINATIONS.

The results of helium, radioactivity and helium index determinations on the three rocks are shown in Tables 3 and 4. In the Lakeview tonalite (Table 3) the radioactivity of the quartz and feldspar is very low and a small part of this is no doubt due to inclusions of zircon and apatite. The radioactivity of biotite is about twice that of feldspar, that of hornblende over 10 times, that of apatite over 100 times, and that of zircon and sphene about 1000 times that of feldspar and quartz. The data are summarized in Table 5.

The radioactivity values obtained for the three granitic rocks are not necessarily characteristic of the batholith, since relatively small samples were used and sampling errors due to inhomogeneity are bound to occur. The values found for the minerals are probably more representative of the rock, since larger samples were ground and mixed for mineral separation. The values of the radioactivities of the rocks calculated from the results on their mineral constituents and their mineral compositions are therefore considered to be more reliable.

TABLE 3.

Helium-radioactivity-age determinations on the minerals from a specimen of Lakeview tonalite (SLR 38-134) from near Perris, Riverside County, California.

Sample	Helium content x 10^5 cc./gm.	Rate of He production alphas/mg./hr.	Helium Index
Rock (measured)	2.37 ±0.23	0.95 ±0.24	77±20
Rock (calculated)	1.48	0.62	73
Quartz	0.40 ±0.01	0.167±0.002	74± 3
Plagioclase (1)	0.79 ±0.02	0.63 ±0.05	39± 4
Plagioclase (2)	0.32 ±0.01	0.225±0.01	44± 4
Plagioclase (3)	0.31 +0.03	0.23 +0.015	41+ 5
Biotite	1.17 ±0.04	0.393±0.019	91± 5
Hornblende	5.1 ±1.3	0.782±0.099	200±55
Magnetite		0.61 ±0.06	
Apatite	79.6 ±4	56.2 ±5	43± 4
Zircon	305. ±15	401. ±25	23± 2
Sphene	264. ±13	266. ±13	30± 2

TABLE 4.

Helium-radioactivity-age results on tonalite and granodiorite from Southern California.

	Helium content x 10^5 cc./gm.	Rate of He production alphas/mg./hr.	Helium Index
Bonsall Tonalite (El 38-29)			
Rock	0.59 ±0.02	1.01 ±0.1	18± 5
Biotite	0.60 ±0.02	0.12 ±0.01	150±15
Granodiorite (El 38-23)			
Rock	0.92 ±0.03	0.43 ±0.04	66± 7
45% quartz, 55% feldspar	0.21 ±0.01	0.125±0.004	51± 6
52% quartz, 48% feldspar	0.14 ±0.01	0.118±0.01	36± 5
20% quartz, 80% feldspar	0.34 ±0.02	0.15 ±0.01	70± 7
95% biotite, 5% feldspar	0.75 ±0.05	0.44 ±0.05	52± 7

TABLE 5.

Distribution of Radioactivity in California Granodiorites.

Sample	Activity Index	Radium Content x 10^{13} gm./gm.	Thorium Content x 10^{6} gm./gm.	Percent of total rock activity	Relative Radioactivity compared to biotite
		Lakeview Tonalite			
Tonalite	0.8	4.9	4.0	100	2.0
Quartz	0.17	1.06*	0.62*	5.4	0.4
Plagioclase	0.23	1.0*	1.5*	18.0	0.6
Biotite	0.39	3.36*	0.51*	11.7	1.0
Hornblende	0.78	3.9*	4.17*	16.2	2.0
Magnetite	0.61	3.25	3.05	0.9	1.6
Apatite	56.2	300	283	17.9	140
Zircon	401	2150	2000	12.8	1030
Sphene	266	1420	1340	17.1	680
		Bonsall Tonalite			
Tonalite	0.7	3.7	3.5	100	6.7
Felsic fraction	1.01	5.4	5.1	85	8.4
Biotite	0.12	0.65	0.6	2	1.0
		Riverside Granodiorite			
Granodiorite	0.43	2.3	2.2	100	1.0
Quartz	0.08	0.43	0.40	13	0.2
Feldspar	0.17	0.91	0.86	75	0.4
Biotite	0.44	2.35	2.2	11	1.0

* Calculated from experimental values of Th/Ra, others from Th/U = 3.3.

TABLE 6.

Helium Indices for Some Mesozoic and Late Paleozoic Rocks.

Rock	Location	Geological Age	Helium Index, M. y.	
Diabase*	Nevada	Cretaceous	47	Ave. 60
Granodiorite	Nevada	Jurassic	56	
Diabase*	Nevada	Jurassic	77	
Granodiorite	Near Valverde, California	Jurassic or Cretaceous	66	Ave. 70
Tonalite	Near Valverde, California	Jurassic or Cretaceous	65	
Tonalite	Near Perris, California	Jurassic or Cretaceous	77	
Camptonite dike*	Vermont	Triassic	77	
Diabase*	Maine	Triassic	72	
Lamprophyre	Eustis Mine, Que.	Triassic	52, 77	
Diabase*	North Star Mine, Calif.	Triassic	77	
Diabase*	Conn.	Triassic	78	Ave. 70
Diabase*	N. J.	Triassic	80	
Diabase*	Nova Scotia	Triassic	86	
Basalt	Watchung, N. J.	Triassic	44	
Basalt	Oldwick, N. J.	Triassic	59	

Rhyolite	N. Attleboro, Mass.	Pennsylvanian	47	
Granite	Fitchburg, Mass.	Carboniferous	43	
Granite	Chelmsford, Mass.	"	68	Ave. 70
Granite	Rockport, Mass.	" (?)	100	
Granite	Quincy, Mass.	"	62	
Monchiquites	Scotland	"	100	

* W. D. Urry (corrected values).

The helium indices for the rocks are in general agreement with the values obtained for other Mesozoic and late Paleozoic rocks (Table 6). The Riverside granodiorite was found to have a lower helium index (66) than the Lakeview tonalite (77), which is consistent with field observations, but the intermediate Bonsall tonalite gave spurious results.

The results obtained on the individual minerals show that the helium index for the rock as a whole cannot be used generally as a reliable means of determining the sequence of geological age. Earlier published results by the helium age method had indicated this, and both Urry and Keevil had pointed out the desirability of studying similar-type rocks. During the past few years, as results have accumulated on separated minerals, it has been necessary to exercise much caution in interpreting the results of helium and radioactivity analyses.

The highest value of the helium index was obtained for the hornblende separated from the Lakeview tonalite, variable results being obtained for quartz, feldspar, and biotite, and low values for apatite, zircon, and sphene. Similar variations in the helium indices for common rock minerals have been found in other rocks. Low results are generally attributed to escape of helium from the mineral, but possible contamination of the sample used for radioactivity determinations by minerals such as zircon may be responsible for a part of the error in some cases.

If these factors could be evaluated, helium ages could be calculated from helium indices. Methods of determining these correction factors are being explored by one of us (N. B. K.), but no rigorous system has yet been devised. However, approximate helium ages can be calculated from the results given in Tables III and IV, by correcting the helium indices on the basis of present knowledge of helium retentivities. By the use of reasonable values of the retentive factor, given in column two of Table 7, an average helium age of 185 million years is calculated.

Helium radioactivity results on a large number of hornblende concentrates have shown that unaltered hornblende retains about 95 per cent of its radiogenic helium. The average of three results for quartz separated from late Paleozoic granites was found to be 0.33, and for feldspar, 0.25. Femic concentrates high in biotite have indicated a probable value of 0.5 for the helium retentivity of this mineral.

The result, 185 ± 50 million years, places the intrusives between the Carboniferous and the Jurassic. Geological data outlined in a previous section indicate a post-Triassic and pre-late Cretaceous age. The probable age indicated by the combined field and laboratory attack is Jurassic. However, the limits of experimental error and the possible entrance of errors, such as contamination by magmatic helium, do not exclude the possibility of Cretaceous age. The variations in the results for quartz, feldspar, and for biotite and the high result for hornblende might be due to varying contamination by small quantities of extraneous helium. In support of this view is the lack of relationship between degree of alteration and helium index, but against it, is the absence of excessive concentrations in particular minerals as found in the few observed instances of helium contamination. (The ratio of helium to radioactivity in the accessories is not excessive.)

TABLE 7.

Results of Helium Age Calculations.

Mineral	Retentivity Factor	Helium Index	Helium Age
Quartz	0.33	60	180
Feldspar	0.25	40	160
Biotite	0.5	97	190
Hornblende	0.9	200	220
		Average Value	185 ± 50

Taking into consideration all factors, a probable age of 135 m.y. is indicated in the oldest of the three rocks investigated. A plausible grouping on the basis of intrusive relationships would then be

Riverside granodiorite	120
Bonsall tonalite	125
Lakeview tonalite	135

With these values, helium retentivities of the various minerals may be calculated from experimental values of the helium index (Table 8). The average retentivity factors for quartz, biotite, and feldspar are higher than the average values for specimens from other rocks. This may be due to the fact that these granitic rocks are younger and less altered than those studied previously.

TABLE 8.

Helium Retentivity in Minerals from California Granitic Rocks.

Sample	Helium Content x 10^5 cc./gm.	Per Cent of rock Helium	Helium Index	Retention Factor
	Lakeview Tonalite			
Tonalite	1.9	100	75	0.56
Quartz	0.4	5.4	74	0.55
Plagioclase	0.3	10.1	41	0.30
Biotite	1.2	15.1	91	0.67
Hornblende	5.1	46.6	200	1.48
Apatite	80	11.2	43	0.32
Zircon	305	4.3	23	0.17
Sphene	264	7.3	30	0.22
	Bonsall Tonalite			
Tonalite	0.6	100	30*	0.24
Felsic fraction	0.6	74	18	0.14
Biotite	0.6	15	150	1.2
	Riverside Granodiorite			
Granodiorite	0.92	100	66	0.55
Quartz	0.1	8.5	40*	0.33
Feldspar	0.3	79	55*	0.46
Biotite	0.75	10.5	52	0.43
	Average Values			
Quartz (2)				0.44
Feldspar (3)				0.30
Biotite (3)				0.77

* Estimated values (see Table 4).

DISCUSSION OF RESULTS.

The helium indices of the three California granitic rocks, when compared with values for other rocks, indicate a Mesozoic or late Paleozoic age. The relatively fresh character of the California rocks would tend to cause higher values of the helium index, so the possibility of Paleozoic age is unlikely. This is in accordance with observed geological relationships. However, the variations in the helium index from rock to rock and mineral to mineral show that the helium index is of little use in indicating either the relative age or the geological ages of these rocks. Helium ages calculated from approximate values of helium retentivities provide more reliable information. When corrections are applied for leakage from the various minerals, a Jurassic age is indicated. Geologically the intrusives are known to have formed between the Triassic and late Cretaceous.

If it is assumed that no extraneous helium was present, the possibility of early Cretaceous age seems to be excluded by the helium age results. Indirect geological criteria are not entirely in accord with this delimitation.

Apart from the problem of age relationships, the results on the Lakeview tonalite, and on the other rocks, are most enlightening from the standpoint of the mineralogical distribution of the radioactive elements. There has been some question, in the past, as to whether the radioactive and other heavy elements are to some extent distributed in a random fashion throughout the rocks, or are concentrated in the very minor constituents. In highly radioactive rocks it has been quite clear that the femic minerals—biotite, zircon, and so on—are generally consistently more radioactive than the felsic minerals, but in the less radioactive rocks the radioactivities of the common rock minerals are more uniform. In the latter type of rocks little or no evidence of any localization of radioactive elements has been found. The detailed study of the Lakeview tonalite, however, supports the view that the radioactive elements are associated with certain rare minerals, and that the activities of the common minerals are probably due to the presence of minute inclusions.

These rocks are only one third as active as the average granitic rocks, and the contrast in activity between the common minerals is not great.

The minor constituents, on the other hand, were highly active, and it is quite probable that inclusions of small grains of apatite, zircon, and sphene account for much, if not all, of the radioactivity of the common minerals. It is interesting to note that zircon, even though present to only two one-hundredths of one per cent, accounts for thirteen per cent of the total radioactivity of the rock. The addition or subtraction of one small crystal may then cause an appreciable error in radioactivity determinations.

This is an important consideration in the alpha-helium method of age determination where the helium and the radioactivity measurements are made on separate portions of sample. The non-uniform distribution of radioactivity is also of significance in the problem of diffusion of helium through minerals.[4]

Here the localization of radioactive disintegration may disrupt the structure, and cause concentration gradients of helium, both of which factors would cause migration of helium during geological time.

CONCLUSIONS.

1. The distribution of radioactivity in Southern Californian intrusives is non-uniform, the bulk of the radioactive elements having separated with the early minor constituents, zircon, apatite, and sphene.

2. The helium retentivities of the constituent minerals in decreasing order were found to be hornblende, biotite, quartz, apatite, feldspar, sphene, and zircon.

3. Consideration of field relationships and corrected helium ages indicate a Jurassic rather than a Cretaceous age for the earliest intrusives in the batholithic complex of Southern California.

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