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## VARIATION OF STRONTIUM ISOTOPES IN TEKTITES

C. C. Schnetzler\* and W. H. Pinson, Jr. (MIT) [1963] *repl*  
 Department of Geology & Geophysics  
 Massachusetts Institute of Technology  
 Cambridge, Massachusetts

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ABSTRACT -- The rubidium, strontium, and strontium isotopic compositions have been determined in tektites from Indo-China, Philippine Islands, Java, Australia, Texas, Georgia, Massachusetts, and Czechoslovakia. The range of  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios in 18 tektites is from 0.7121 to 0.7223, indicating the results of Pinson *et al.* (1958), reporting no radiogenic strontium in tektites, were not of sufficient precision to study the problem definitively. This range of  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios is quite small compared to the range found in nature, in general agreement with previous studies of oxygen and silicon isotopes in tektites. Rb and Sr contents of tektites from a given locality are very similar, but differences exist between localities. There is indicated, however, a sympathetic variation of the  $\text{Sr}^{87}/\text{Sr}^{86}$  and Rb/Sr ratios; i.e., the tektite groups have age values of approximately 400 m.y. If tektites were derived from the Moon, and surface of the Moon is old (4.5 b.y.), the observed  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios could not have been developed by a chondritic, granitic, or tektitic surface material, but could have been generated by a basaltic type

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\*Present address: Theoretical Division, National Aeronautics  
 and Space Administration, Goddard Space Flight  
 Center, Greenbelt, Maryland.

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material. However, production of tektites from a basaltic type surface material during a high temperature fusion implies extreme fractionation in a direction contrary to expectation. A second possibility is that the Moon has undergone differentiation within the last 500 m.y.

AUTHOR

INTRODUCTION -- Several elements whose isotopic compositions vary for reasons other than radioactivity, i.e., due to physical-chemical fractionation, have been isotopically analyzed in tektites. These include hydrogen (Friedman, 1958), oxygen (Silverman, 1951; Taylor and Epstein, 1962, 1963), and silicon (Tilles, 1961). The values obtained were compared to the isotopic composition of different types of terrestrial and extra-terrestrial material in the hope of obtaining a clue to the origin of tektites.

Although the variation of strontium isotopes in nature is due to radioactivity, the half life of the decay of  $\text{Rb}^{87}$  to  $\text{Sr}^{87}$  is so long, approximately  $4.7 \times 10^{10}$  years (Glendenin, 1961), no measurable change has occurred in the  $\text{Sr}^{87}$  content since the tektites were formed from their parent material. This formation of the tektites is assumed to have occurred at the time dated by the K-Ar method, i.e., less than 35 m.y. ago (Zahringer, 1963). Also, as strontium is much heavier than hydrogen, oxygen, or silicon it should not be as subject to fractionation during the high temperature fusion (Taylor and Epstein [1963] found a slight increase in the  $^{18}\text{O}/^{16}\text{O}$  ratio during fusion of tektite glass). Thus, the strontium isotopic composition of the tektites now should be the same as the parent material at the time of fusion.

The relative isotopic abundance of  $\text{Sr}^{87}$ , conveniently expressed as the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio, in a given system is determined by the Rb/Sr ratio of the system, the initial abundance at an initial time, and the time elapsed since this initial time. Thus terrestrial materials, having different Rb/Sr ratios, different initial  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios, and different ages, have a wide range of  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios. In this respect the study of strontium isotopes may be more informative than the study of oxygen, hydrogen, or silicon which show a rather small range of variation in nature.

In addition, of course, a knowledge of the relative isotopic abundances of strontium, and the Rb and Sr elemental contents enable the calculation of an age value for the sample. In the case of samples such as tektites whose association or past geologic history are in doubt, however, a number of assumptions must be made and such age values must be interpreted with discretion.

Previous studies of strontium in tektites have been made in this laboratory, but have been plagued with analytical difficulties. Pinson et al. (1956) suggested that the relative Sr isotopic composition of a representative sampling of tektites from all known localities be measured; if the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios are identical in correspondence to their Rb/Sr ratios (all lying on the same isochron) this would prove their extraterrestrial origin, as this circumstance occurring in terrestrial rocks from different parts of the world would be highly improbable.

Later Pinson et al. (1957a) reported the rubidium and strontium, and strontium isotopic composition of three indochinites, two australites, and two philippinites. The  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios varied from 0.718 to 0.735, while the Rb/Sr ratios ranged from 0.39 to 1.08, but there was no apparent sympathetic variation between the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios and the Rb/Sr ratios. The computed age values, assuming an initial ratio of 0.712 (this was called "normal strontium" as it was the ratio found in sea water), varied from 180 to 730 m.y., with estimated errors of  $\pm 50\%$ , the large error being due chiefly to the poor precision in the  $\text{Sr}^{87}/\text{Sr}^{86}$  mass spectrometric determinations.

These values were later reported to be in error, and the corrected values for the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios were given as 0.712 (Pinson et al. 1958). The corrections were thought necessary due to suspected lack of mass discrimination in the mass spectrometer. In view of the questionable validity of the correction that was applied, the rather large errors in the precision of the measurements and in view of improvement in mass spectrometric techniques, it was thought a re-investigation of the strontium isotopic composition of tektites might prove fruitful. As the strontium isotopic composition is directly related to the Rb/Sr ratio, the elemental rubidium and strontium contents were also investigated.

### Techniques

Rubidium and strontium were determined mass spectrometrically by the stable isotope dilution method, and by X-ray fluorescence techniques. The strontium isotopic compositions were determined mass spectrometrically on a separate portion of "raw" (i.e. unspiked) strontium extracted from the tektites. The techniques used in this investigation for the mass spectrometric determinations are essentially the same as described by Herzog and Pinson (1955, 1956). The mass spectrometer used was a standard 6-inch radius, 60° sector, solid source, single collector Nier-type instrument. Amplification of the ion current was obtained by means of a vibrating reed electrometer.

The  $\text{Sr}^{84}$ ,  $\text{Sr}^{86}$ , and  $\text{Rb}^{87}$  spikes used in the isotope dilution analyses were prepared and calibrated as described by Pinson (1960, 1962).

The X-ray fluorescence unit used was a North American Phillips instrument with a tungsten target X-ray tube. The crystal was LiF and the detector was a scintillation counter. The standards used were tektites which had been analyzed for rubidium and strontium by isotope dilution. It was found that tektites from a given locality were similar enough in major element composition so that unknowns and standards from the same area could be compared with no matrix effects, and a precision and accuracy comparable to the mass spectrometric analytical work was achieved.

The powdered sample was packed into a nylon sample holder, covered with a 0.5 mil mylar sheet and put in the X-ray beam. The region of  $K\alpha$  radiation of rubidium and strontium was scanned to

determine the exact location of the peaks, and to pick the angles on both sides of the peaks where background could be obtained free of any interfering element emission. Operating conditions were: LiF crystal; pulse height analyzer: base = 41.5, window = 5.60; number of counts on peak and background = 102,400.

The goniometer was set at the proper angles and the times necessary to accumulate the fixed number of counts were measured at the peak and background position. The calculated counts per second (cps) for the backgrounds were subtracted from the peak intensities to give a net peak intensity. The intensity values obtained for the standard samples were plotted against the ppm strontium, obtained by isotope dilution analyses, to yield a calibration curve. Strontium values for the unknowns were obtained by interpolation from this calibration curve. The rubidium values in the standards were too similar (the Rb varied only from 111 to 129 ppm) to enable a curve to be drawn, so an average value of cps/ppm rubidium was calculated, and this value was used to calculate the rubidium contents of the unknowns.

It was found that only samples of quite similar major element composition could be compared in this manner. The indochinites and philippinites could be compared but samples from other localities gave spurious results when compared with samples from these two localities. This matrix effect could have been eliminated by the addition of an internal standard. However, it was found that the

tektites from a given locality were similar enough so that unknowns and standards from the area could be compared.

It was apparent from the earlier investigations of Pinson and co-workers that the variations of  $\text{Sr}^{87}$  in tektites are small, if present at all. It was also apparent that complete resolution between the large mass 88 peak and the smaller mass 87 peak ( $88/87 \approx 12$ ) was not obtained -- i.e., the 88 peak "tail" was enhancing the 87 peak. Improved vacuum conditions and re-alignment of the mass spectrometer have improved discrimination so that ratios of minimum ion current between the 87 and 88 peaks to the ion current at the 87 peak of 1/600 or better are obtained routinely. A small portion of a typical record are shown in Figure 1 to demonstrate the resolution obtained.

The precision of the strontium isotopic analysis runs can be estimated from replicate runs on several samples shown in Table 1. The Homestead meteorite runs were made of three separate portions of a powdered sample, each receiving independent chemical treatment. The  $\text{SrCO}_3$  standard runs were made on separate portions of a liquid sample and did not undergo any chemical purification before analysis. The standard deviation of a single analysis ( $\sigma$ ) of the  $(\text{Sr}^{87}/\text{Sr}^{86})_{\text{N}}$  ratio in both cases is  $\pm 0.0006$ , or approximately 0.085%.

An isotopic fractionation correction was applied to the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios on the assumption that the  $\text{Sr}^{86}/\text{Sr}^{88}$  ratio is a constant and equal to 0.1194. The run-to-run variations of the  $\text{Sr}^{86}/\text{Sr}^{88}$  ratio from this value were attributed to instrumental fractionation, chiefly fractionation of the isotopes of different mass off the filament.

Half of any correction necessary to re-adjust the  $\text{Sr}^{86}/\text{Sr}^{88}$  ratio to 0.1194 was applied to the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio. The corrected, or normalized,  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios are listed in the tables in columns headed  $(\text{Sr}^{87}/\text{Sr}^{86})_N$ , while the ratios as actually measured are listed in columns simply headed by  $\text{Sr}^{87}/\text{Sr}^{86}$ .

The  $\text{SrCO}_3$  standard (Eimer and Amend Company lot #492327) was also used to monitor the accuracy of the isotope ratio determinations. Five laboratories have informally reported that they find a value of  $(\text{Sr}^{87}/\text{Sr}^{86})_N$  in the range of 0.708 to 0.709 (Hurley et al., 1963). The average value shown for this standard in Table 1 indicates there are no systematic errors between this laboratory and the other reporting laboratories.

Replicate analyses of a tektite by isotope dilution, and by X-ray fluorescence, indicate the rubidium and strontium results have a precision of approximately  $\pm 2\%$  (relative deviation of a single analysis). Inter-laboratory analyses of a standard sample indicate the accuracy of the isotope dilution analyses is about  $\pm 1\%$  (Pinson, 1960, p. 244). By comparison of the strontium and rubidium values for G-1 and W-1 obtained by the M.I.T. Geochronology Laboratory with the results obtained by other methods of **analysis** (Stevens, 1960, p. 97 and 99) a conservative estimate of the accuracy would be  $\pm 5\%$ .



### Results

The analytical results obtained in this investigation are presented in Tables 2 and 3. Eighteen tektites were analyzed for strontium isotopic composition and 27 tektites were analyzed for rubidium and strontium elemental contents.

Four philippinites, from three different sites, have  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios varying only from 0.7168 to 0.7182, and average 0.7173. As the reproducibility ( $\sigma$ ) of strontium isotope analyses appears to be approximately  $\pm 0.0006$ , the variation in the philippinites is only slightly greater than one would expect from measurement error. Seven philippinites, including the four analyzed for isotopic composition, have an average rubidium content of  $117 \pm 2$  ppm ( $\bar{\sigma}$ ) and an average strontium content of  $173 \pm 4$  ppm. The variations are greater than measurement error so some differences exist between the samples in rubidium and strontium contents. However, these differences are small and the Rb/Sr ratios are remarkably uniform, averaging  $0.68 \pm 0.02$  for the seven samples.

Four indochinites, from four different localities, also have homogeneous  $\text{Sr}^{87}/\text{Sr}^{86}$  values, ranging from 0.7182 to 0.7185 and averaging 0.7184. This variation is even less than one could expect from measurement error. The  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of indochinites seems to be slightly higher than the ratio measured in philippinites, but not enough samples have been analyzed to determine if this difference is real. There is no doubt, however, that the Rb/Sr ratio of the

indochinites differs significantly from the Rb/Sr ratio of the philippinites. Eight indochinites have an average Rb/Sr ratio of  $0.89 \pm 0.013$ . The rubidium and strontium contents average  $118 \pm 3$  ppm and  $132 \pm 2$  ppm respectively, the variation again only slightly greater than measurement variation. It should be noted that the average rubidium contents of the indochinites and philippinites are identical within experimental error but that the strontium content of the indochinites is significantly lower than the strontium content of the philippinites. It is thought that alternation of the order of analysis throughout the analysis period excludes any possibility of a systematic error.

Two australites have similar  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios of 0.7162 and 0.7172 while a javanite has a  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of 0.7182. The Rb/Sr ratios of the two australites are dissimilar however--0.47 and 0.71, and the Rb/Sr ratio of the javanite is 0.64, similar to the philippinites. Two other australites, which were not analyzed for isotope ratios have identical Rb/Sr ratios of 0.66. The four australites have an average Rb/Sr ratio of  $0.62 \pm .05$ , similar to the philippinites but exhibiting much larger variation. However, more sampling is needed of this important group of tektites.

The four North American tektites have an average  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of 0.7125, and the range exhibited -- from 0.7121 to 0.7130 -- could be attributed to analytical error. The Massachusetts and Georgia tektites contain quite similar rubidium contents (78 and 74 ppm)

respectively) and strontium contents (177 and 170 ppm respectively). The three Texas samples contain slightly less rubidium and strontium than the Massachusetts and Georgia samples, but the Rb/Sr ratios of the five are amazingly similar for samples from such widely separated localities.

The three moldavites analyzed gave the highest  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios of the tektites analyzed, averaging 0.7216. Again the range, 0.7208 to 0.7223, is about what one would expect from analytical error. The moldavites also have the highest Rb/Sr ratios, approximately one, of any group analyzed. Due to the small size of the sample available, one moldavite (T3314) could not be analyzed for rubidium and strontium. However, values of 130 ppm Rb and 136 ppm Sr were reported for this sample by Pinson et al. (1958).

An "amerikanite" from the Santa Mesa site in the Philippine Islands was analyzed for rubidium and strontium contents. "Amerikanites" are found intermixed with the true philippinites at this site and exhibit a sculptured tektite-like external appearance. However, they show an internal structure typical of ordinary obsidian, and are considered such by Beyer (1935). Certainly the rubidium and strontium contents of the sample analyzed are dissimilar to the philippinites and are more characteristic of obsidian than tektites. The major element contents of this sample (Schnetzler and Pinson, 1963) are also typical of obsidian and unlike tektites.

### Discussion of Results

The first conclusion that can be reached from this investigation is that the work of Pinson et al. (1958) was in error and that tektites do contain radiogenic strontium-- in the limited sense that their  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio is greater than "primordial" strontium (approximately 0.700 -- see following discussion) or "normal reagent" strontium (approximately 0.708 to 0.711).

However, many of their conclusions were valid and will not be discussed here. It should only be mentioned that the results of this investigation substantiate their conclusions that it is possible, from the viewpoint of  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios, for the tektites to have been derived from basalt and achondrites, but not from any chondrite yet measured for strontium isotopic composition.

1. Comparison of the variation of  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios in tektites with the variation of terrestrial materials.

The variation in the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio measured in tektites, from 0.7121 to 0.7223, is certainly real, i.e., not due to analytical error, as the spread is approximately 16 times the reproducibility ( $\sigma$ ). It is of interest to compare this variation with the spread observed in nature and, particularly with the variation one might expect assuming various terrestrial fusion models.

$\text{Sr}^{87}$  and  $\text{Sr}^{86}$  ratios and Rb/Sr ratios reported in the literature for various types of terrestrial and extraterrestrial materials are given in Table 4. A complete literature survey has not been attempted. Unfortunately it has been common practice in Rb-Sr dating to use only a few favorable minerals rather than the whole rock for analysis so

that data of interest to this investigation are sparse, or non-existent, for some rock types.

It can be seen that certain types of rocks show a very limited range of  $\text{Sr}^{87}/\text{Sr}^{86}$  values while others exhibit a considerable range. This is due to the magnitude of the Rb/Sr ratio, the range of the Rb/Sr ratio, and the range of the age of the samples analyzed. Granites have a very large variation in  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios. As all but one of the 21 granites in Table 4 are over 1000 m.y. old it is apparent that the variation shown is due primarily to the variation in high Rb/Sr ratios. The average  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of 0.85 is undoubtedly not a representative number for average granite but is a reflection of the great age of the samples analyzed.

Basalts, however, do not show this extreme range in  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios due primarily to their low Rb/Sr ratios. The basalts analyzed by Faure were Recent basalts, but due to their low Rb/Sr ratios little change in the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio would appear in older samples.

Nine obsidians analyzed by Pinson and Bottino (1961) show a limited  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio, ranging from 0.704 to 0.724, while the Rb/Sr ratios are quite high--from 1.2 to 31. The low  $\text{Sr}^{87}/\text{Sr}^{86}$  values are due to the young ages of the samples, and since obsidians generally devitrify quickly (geologically speaking) high  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios similar to that observed in granites would not be expected.

Limestones, both young and old, have low  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios due to their extremely low Rb/Sr ratios. The Rb/Sr ratio in limestones averages about 0.008 (Faure, 1961, p. 115).

The shales analyzed by Whitney (1962), all from the Hamilton Group of Middle Devonian age, show a considerable spread in both Rb/Sr ratio and  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio. Faure's (1961) analyses are on two composites of Paleozoic shales from the east and west coasts of North America. Variations are suppressed between such composite samples.

Chondrites have fairly high, varying  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios. Currently, analyses of stoney meteorites are being carried out at M.I.T. Variations from about 0.73 to 0.96 have been found in chondrites.

Achondrites have extremely low  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios which show almost no variation (See Table 4). Under the assumptions that 1) all terrestrial and meteoritic strontium was derived from a homogeneous parent material and thus had the same abundance of initial  $\text{Sr}^{87}$ , and 2) the meteorites have existed as closed chemical systems since their formation approximately 4.5 b.y. ago, the achondrites probably give us a good approximation of the initial  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio. As Patterson (1956) and his co-workers have demonstrated that these assumptions hold true in the case of lead, by analogy the assumptions should be true for strontium. The achondrites have extremely low Rb/Sr ratios ( $\sim 0.005$ ) so their initial  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio has remained essentially unchanged for  $4.5 \times 10^9$  years. The lowest value found for achondrites (0.700 - Gast, 1960) is accepted here as the primordial  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio.

From the above discussion it is obvious that the data is too meager to define the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios in the various rock types. The data does show, however, that a great deal of variation does exist.

The  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios in tektites given in Table 3 and the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios taken from the literature for various materials as given in Table 4 are plotted in Figure 2. The figure shows the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios for various materials from only 0.700 to 0.780, approximately the lower one-seventh of the range exhibited by the data in Table 4. The full range of the data is shown in the inset; the shaded portion of the inset is that part of the scale which has been expanded in Figure 2.

It can be seen that the variation of strontium isotopic composition, although measurable, is quite small compared to the variation observed in nature. This is in qualitative agreement with the constancy of oxygen isotope ratios (Taylor and Epstein, 1962; 1963) and silicon isotope ratios (Tilles, 1961) recently demonstrated in tektites. The strontium isotopic compositions are particularly homogeneous when compared to the isotopic compositions of the acid rocks most commonly mentioned as possible parents for the tektites: siliceous shales (Urey, 1959), granite (Mason, 1959) and granophyre (Lovering, 1960).

It has been suggested that tektites were formed by many independent fusions of terrestrial material in a random manner by such processes as lightning (Hawkins, 1960) or small meteoritic impacts (Spencer, 1933). The probability of fusing a certain type of

material should be in direct proportion to the extent of that material's occurrence upon the earth's surface. Such modes of production should produce tektites exhibiting a wide variation in Sr isotopic composition. However, in recent years it has become apparent that there are only a few independent tektite occurrences. K-Ar ages of tektites (Zahringer, 1963), chemical and specific gravity determination (Schentzler, 1961; Chapman et al., 1963) and aerodynamic evidence (Chapman, 1963) strongly indicate that there are a maximum of four known independent groups of tektites. These are the North American tektites, the moldavites, the Australasian tektites, and possibly the Ivory Coast tektites. Therefore, the tektites listed in Table 3 would represent only three large scale impacts. The variation of isotopic composition between groups could be quite small for several reasons. 1) Naturally, the smaller the number of impacts the greater the probability of striking the same general type of material; it could be merely chance that the different groups of tektites have similar and characteristic compositions. 2) Widespread surficial deposits of quite uniform compositions have been proposed for the parent material in an effort to explain the lack of chemical variation in tektites; soil (Schwarcz, 1962) and loess (Taylor, 1962) have been specifically suggested. The variations in isotopic composition of these particular materials are not known. However, the young sediments which cover much of the earth's surface represent a possible source that has been at least partially homogenized with respect to strontium isotopic composition. 3) The impact may have fused and mixed a very large



volume of material, incorporating a number of rock types, thereby suppressing variations. The larger the volumes fused, the closer the materials' composition would be to crustal abundances. Tektites are strikingly similar in chemical composition to estimates of crustal abundances (Suess, 1963), and it is interesting that the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios in tektites are quite similar to recent estimates of the crustal average (Faure, 1961). However, if a large volume of rather heterogeneous material was fused, an almost incredible degree of mixing must have occurred before the liquid parent material solidified into the individual tektites. The tektites of any particular group demonstrate over wide areas a surprising homogeneity with respect to strontium isotopic composition, and it is difficult to imagine such mixing during the short time the material was molten.

Variation of the strontium isotopic composition of tektites do not point specifically to a terrestrial or extraterrestrial origin, but emphasize again how completely homogeneous these objects are. Not enough is known of the strontium isotopic composition of various widespread surficial materials, or the degree of mixing which can occur during high temperature fusion by impact to indicate if such constancy is impossible to explain by a terrestrial impact origin.

## 2. Rb-Sr age values of tektites

It is interesting that the data presented in Tables 2 and 3 suggest a positive correlation of  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios with Rb/Sr ratios in the tektites. This is illustrated in Figures 3 and 4.

To quantitatively test the degree of association between the Rb/Sr ratios and the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios, the co-efficient of correlation

(Moroney, 1956, pgs. 289-291) was calculated. If all 18 samples are considered independent (Figure 3) the coefficient of correlation ( $r$ ) is +0.91 (when  $r=+1$  there is perfect direct relationship and when  $r=0$  there is no relationship --a random scatter). This coefficient of correlation is significant at less than the 0.1% level-- i.e., the probability of this high correlation coefficient arising by chance in this large a population is less than one in a thousand. If the samples are assumed to represent three independent groups (for reasons discussed above)--the australasian tektites, North American tektites, and moldavites--and the average values for each group is used (Figure 4), the correlation coefficient is +1.0. The small number of groups involved precludes a calculation of the statistical significance of this correlation but the Rb/Sr -  $Sr^{87}/Sr^{86}$  correlation seems more than coincidental or circumstantial. Certainly it is difficult to reconcile this correlation with tektite production within the last 35 m.y. from random materials of different ages, compositions, and geologic histories. It seems quite unlikely that the variables which determine the present  $Sr^{87}/Sr^{86}$  ratios (initial Sr isotopic composition, Rb/Sr ratios, and ages) would be just in the right combinations to cause unrelated groups of tektites to lie on a particular line.

This correlation suggests the tektites from different areas are related, and might have a common origin. Rocks formed at the same time from a common material (for example, by fractional crystallization from a magma) will develop with time, from a common  $Sr^{87}/Sr^{86}$  ratio,

different  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios in relation to their different Rb/Sr ratios. Such a series of rocks, when plotted on a graph of  $\text{Sr}^{87}/\text{Sr}^{86}$  vs Rb/Sr lie on a straight line which is an isochron. The slope of the isochron gives the time since differentiation from the common source, while the intercept of the line on the  $\text{Sr}^{87}/\text{Sr}^{86}$  axis (at zero Rb/Sr) is the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of the source material at the time of differentiation.

If the direct correlation of the Rb/Sr ratios with the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios in tektites is due to their derivation from a common source, and if they have remained closed systems to Rb and Sr since that time, age values can be calculated without having to assume a particular initial  $\text{Sr}^{87}/\text{Sr}^{86}$  value. The "least square" line through all 18 points shown in Figure 3 corresponds to the 310 m.y. isochron ( $\lambda_{\text{Rb}} = 1.47 \times 10^{-11} \text{ year}^{-1}$ ). The "least squares" line in Figure 4 through the points representing the averages of the three groups is the 400 m.y. isochron. It is interesting that the initial  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of 0.705 is within the narrow range of values reported for volcanic rocks of recent origin--that is, this initial ratio is typical of the source region of igneous rocks (Hurley et al., 1962)

The assumption that the tektites and/or their parent material have remained closed systems since their derivation from a common source presents a serious objection to the validity of the age value. If tektites were formed by high temperature fusion at a time dated by the K-Ar method (35 m.y. to less than 1 m.y. ago) then the assumption is that the Rb and Sr contents, and isotopic contents of the strontium in the parent material were not disturbed by this fusion. In a

sense we are looking through this "metamorphic" event to the time of formation of the parent material. The fact that the points do lie on an isochron suggests that extensive selective fractionation might not have occurred during the fusion process as its occurrence would add another variable to those already mentioned (initial  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios, parent  $\text{Rb}/\text{Sr}$  ratios, and age) which must be in a particular combination to cause unrelated samples to lie upon an isochron. In fact, in normal  $\text{Rb}-\text{Sr}$  dating, the isochronal relationship of samples is usually regarded as evidence of closed systems. In view of the assumptions involved, and the small number of groups of tektites, the most that should be stated at this time is that the data suggests the different groups of tektites, or their parent materials, have developed from a common type of source material. The initial  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of the source was 0.705, and the time since this development has been approximately 400 m.y.

Independent of the validity of a particular isochron age for tektites, it is clear that tektites have undergone differentiation within the last  $10^9$  years. Tilton (1958) pointed out that the  $\text{U}$  and  $\text{Th}$  in tektites have increased relative to  $\text{Pb}$  within the last tens of millions of years, while this investigation has demonstrated that  $\text{Rb}$  has increased relative to strontium within the last hundreds of million years. If after this differentiation a high temperature fusion took place one might expect  $\text{Pb}$  to be lost preferentially to  $\text{U}$  and  $\text{Th}$ , and/or  $\text{Rb}$  lost preferentially to  $\text{Sr}$ . Analyses would yield  $\text{U}-\text{Pb}$  ages that are too young and  $\text{Rb}-\text{Sr}$  ages that are too old; this might be an explanation for the quantitative disagreement in these ages. The

Rb-Sr ages however would be maximum values. The maximum age since the tektites obtained their present Rb/Sr ratio is obtained by assuming an initial ratio equal to the primordial value found in achondrites (0.700). The age calculated is approximately 700 m.y., making them distinctly younger than stony meteorites.

3. Comparison of the data with a possible lunar origin for tektites

Some investigators have suggested that tektites were ejected from the Moon's surface by meteorite impacts (Nininger, 1943; O'Keefe, 1960; and others). Most students of the Moon (for example, Urey, 1952, p. 30; Baldwin, 1949, p. 194) have concluded that the present lunar surface was formed soon after the origin of the Moon, and the Moon has remained relatively cool since that time. Also, the age of the Moon is generally assumed to be the same as the rest of the solar system - approximately 4.5 billion years. Assuming these conditions, it is possible to calculate the present-day  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio for various hypothetical lunar crustal materials.

Granitic material, with an average Rb/Sr ratio of 1 (Faure, 1961, p. 106) and an initial  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of 0.700 would have, after 4.5 billion years, a  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of about .890, far greater than that found in tektites. Chondritic material would have a present day  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of about 0.750, (which is approximately the ratio found in chondrites) again in excess of that found in tektites. If it is assumed that the surface of the Moon has had a chemical composition similar to tektites since its formation the  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio would be between 0.78 and 0.90, again far in excess of that found in tektites.

Conversely, the Rb/Sr ratio that would generate a  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio of 0.718 (average actually observed in tektites) in 4.5 b.y. from a primordial ratio of 0.700 is approximately 0.1. The common terrestrial material whose Rb/Sr ratio most closely resembles this ratio is basalt (see Table 4), and the marias have commonly been theorized as being covered with basaltic lava flows. Thus a basalt that had remained on the surface of the Moon for 4.5 billion years should have about the same  $\text{Sr}^{87}/\text{Sr}^{86}$  ratio as is observed in tektites. It is difficult, however, to reconcile such a radical change in Rb/Sr ratio (from 0.1 to between 0.44 and 1.1) and other radical changes in composition such as increase in K and Si with any mechanism that would throw the tektites from the Moon, especially high temperature fusion where the alkali elements would be expected to volatilize preferentially to the alkaline earth elements.

Perhaps a more reasonable hypothesis, if tektites come from the Moon, is that there has been differentiation of the Moon since the start of the Paleozoic era and that tektites are the results of fusion of this differentiated material or the ejecta from volcanos during this time. The recent spectroscopic evidence of gas escaping from Alphonsus crater and the existence of a temporary surface temperature of 1200-1500°K in this crater have raised serious doubts concerning the widely held theory that the Moon is cold, without any remnants of volcanic activity (Kozyrev, 1961). By comparing the number of craters on the marias with the number of meteorites of various sizes that fall on the Earth per year, Kreiter (1960) has concluded the marias are about 200 m.y. old. But the same data has been used by

most lunar investigators to conclude the marias are approximately 4.5 billion years old (Opik, 1960).

In short, the low Rb-Sr and U-Pb ages in tektites are difficult to explain if the tektites were derived from the Moon, if the conventional model of a cold undifferentiating body is accepted. Recent differentiation of the Moon is almost required if tektites were produced from its surface.

In conclusion, the slight variation in strontium isotopic compositions exhibited in tektites suggests that if they are terrestrial in origin they must be derived from a uniform widespread source material. The direct correlation of Rb/Sr ratios with  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios (i.e., the homogeneity of age values) is, however, difficult to reconcile with any terrestrial origin in view of their widespread geographic locations and varying K-Ar ages. As the isochron relationship is based upon only 3 points, the analysis of the fourth possible independent group, the Ivory Coast tektites, is quite critical to this discussion; unfortunately none have been available. It would also be interesting to look for possible uniform source materials which have apparent ages of approximately 300 to 500 m.y., using 0.705 as the initial ratio. The recent suggestions of soil (Schwarcz, 1962) and loess (Taylor, 1962) might be possibilities. Equally germane to this discussion would be the comparison of variations of strontium isotopes in tektites with that observed in known impactites, and the comparison of tektites with postulated source regions, such as the moldavites with the Ries crater, and the Ivory Coast tektites with the Bosumtwi crater (Cohen, 1961). Some of this work is already in progress.

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TABLE 1. Replicate  $(\text{Sr}^{87}/\text{Sr}^{87})_{\text{N}}$  Ratio Analyses

<u>Homestead Meteorite</u>	<u>SrCO<sub>3</sub> Standard</u>
0.7503	0.7070
0.7507	0.7076
0.7515	0.7071
	0.7084
Avg. = 0.7508	0.7085
	0.7076
$\sigma = \pm 0.0006_1$	Avg. = 0.7077
$E\% = \pm 0.08\%$	$\sigma = \pm 0.0006_3$
	$E\% = \pm 0.09\%$

Table 2 Rb and Sr Content of Tektites

Sample No.	Locality	Rb(ppm)	Sr(ppm)	Rb/Sr
PHILIPPINE ISLANDS				
T3379	Santa Mesa Site Rizal, Prov.	115	174	0.66
T3962	Santa Mesa Site Rizal, Prov.	111 110*	176 176*	0.63
T3964	Santa Mesa Site Rizal, Prov.	126*	175*	0.72
T3765	Pugad-Babuy Site Bulakan, Prov.	119 118*	191 188*	0.62
T3979	Pugad-Babuy Site Bulakan, Prov.	118*	174*	0.68
T3984	Pugad-Babuy Site Bulakan, Prov.	112 115*	164 168*	0.68
T3986	Busuanga Island	116 115*	157 157*	0.74
INDO-CHINA				
T3987	Kouang-Tcheou Wan	118 123 119 116*	138 138 133 129*	0.88
T3989	Kouang-Tcheou Wan	110*	132*	0.83
T3990	North Cambodia	116 116*	127*	0.91
T3991	North Cambodia	129 134*	134 139*	0.96
T3993	Annam	113 112*	130*	0.87

\*X-ray fluorescence analysis. All others by isotope dilution.

Table 2 (Con't)

T4219	Dalat, S. Viet-Nam	111 110*	125 125*	0.89
T4220	Dalat, S. Viet-Nam	124*	138*	0.90
T4218	Dalat, S. Viet-Nam	125*	138*	0.91
T4104	Java	98	153	0.64
AUSTRALIA				
T3313	Charlotte Waters, Central Australia	109	154	0.71
T4214	Nullarbor Plain	93	197	0.47
T4215	Nullarbor Plain	135*	205*	0.66
T4216	Nullarbor Plain	144*	218*	0.66
NORTH AMERICA				
T4106	Grimes Co., Texas	70	152	0.46
T4300	Grimes Co., Texas	64	134	0.48
T5129	Lee Co., Texas	63*	134*	0.47
T4271	Empire, Georgia	74	170	0.44
T4091	Martha's Vineyard, Mass.	78	177	0.44
CZECHOSLOVAKIA				
T4090	Nechov, Bohemia	146	136	1.07
T3314	"moldavite"	130†	136†	0.96
T4575	Ratiborova Lhota, Bohemia	136	143	0.95
"AMERICANITE"				
3967	Philippine Is.	181	98	1.85

\*X-ray fluorescence analysis. All others by isotope dilution.

†From Pinson et al. (1958).

TABLE 3 - STRONTIUM ISOTOPIC RATIO RESULTS

MPT No.	Locality	Date	86/88	87/86	(87/86)N	Scans	E 87/86
PHILIPPINE ISLANDS							
T3379	Santa Mesa Site Rizal Province	6/25/63	.1208	.7129	.7172	72	0.06%
T3962	Santa Mesa Site Rizal Province	5/14/63	.1200	.7153	.7171	54	0.03%
T3984	Pugad-Babuy Site Bulakan Province	7/23/63	.1200	.7150	.7168	60	0.03%
T3986	Bucugana Is.	6/3/63	.1199	.7167	.7182	54	0.05%
Average for Philippinites			.1202	.7150	.7173		
INDO-CHINA							
T3987	Kouang-Tcheou-Wan	7/24/63	.1207	.7144	.7183	66	0.03%
T3991	North Cambodia	7/11/63	.1194	.7182	.7182	66	0.03%
T3993	Annam	5/9/63	.1203	.7158	.7185	72	0.02%
T4219	Dalat, S. Viet-Nam	8/3/63	.1196	.7178	.7185	58	0.03%
Average for Indochinites			.1200	.7166	.7184		
JAVA							
T4104	Java	8/13/63	.1190	.7193	.7182	54	0.02%



MIT No.	Locality	Date	86/88	87/86	(87/86)N	Scans	E 87/86
AUSTRALIA							
T3313	Charlotte Waters Central Australia	8/1/63	.1204	.7142	.7172	72	0.06%
T4214	Mullarbor Plain	5/16/63	.1199	.7148	.7162	72	0.04%
	Average for Australites		.1202	.7145	.7167		
NORTH AMERICA							
T4106	Grimes Co., Texas	3/11/63	.1194	.7122	.7122	72	0.05%
T5129	Lee Co., Texas	4/2/63	.1196	.7114	.7121	72	0.03%
T4271	England, Georgia	8/16/63	.1197	.7119	.7127	66	0.04%
T4091	Martina's Vineyard, Mass.	6/6/63	.1199	.7115	.7130	72	0.02%
	Average for N. America		.1196	.7118	.7125		
CZECHOSLOVAKIA							
T4090	Nechov, Bohemia	5/28/63	.1203	.7196	.7223	72	0.02%
T3314	"moldavite"	8/15/63	.1199	.7194	.7208	64	0.05%
T4575	Ratiborova Lhota, Bohemia	3/12/63	.1193	.7221	.7218	62	0.04%
	Average for moldavites		.1198	.7204	.7216		

Table 4. Some  $\text{Sr}^{87}/\text{Sr}^{86}$  ratios and Rb/Sr ratios reported for terrestrial and extraterrestrial materials

Material and Reference	No. of Samples	Range $\text{Sr}^{87}/\text{Sr}^{86}$	Avg $\text{Sr}^{87}/\text{Sr}^{86}$	Range Rb/Sr	Avg. Rb/Sr
GRANITE					
Gast (1960) <sup>a</sup>	10	0.720 - 1.00	0.830	0.35 - 3.9	1.64
Fairbairn <u>et al.</u> (1961) <sup>b</sup>	11	0.732 - 1.25	0.868	0.25 - 6.0	1.78
BASALT					
Gast (1960)	5	0.705 - 0.711	0.708	-	-
Faure (1961) <sup>c</sup>	25	0.705 - 0.710	0.708	0.02 - 0.18	0.05
OBSIDIANS					
Pinson and Bottino (1961) <sup>d</sup>	9	0.704 - 0.724	0.714	1.2 - 31.	4.2
LIMESTONE					
Gast (1960)	5	0.704 - 0.712	0.709	-	-
SHALE					
Faure (1961) <sup>e</sup>	2	0.720 - 0.723	0.7215	-	-
Whitney (1962)	20	0.712 - 0.755	0.735	0.34 - 2.67	1.57
CHONDRITES					
Herzog and Pinson (1956)	2	0.753 - 0.755	0.754	0.33 - 0.36	0.345
Gast (1960, 1961)	5	0.739 - 0.960	0.796	0.26 - 1.37	0.54
ACHONDRITES					
Herzog and Pinson (1956)	1	-	0.703	-	-
Gast (1960, 1961)	3	0.700 - 0.703	0.701 <sub>5</sub>	0.002 - 0.005	0.003
SEA WATER					
Pinson <u>et al.</u> (1957b) <sup>f</sup>	1	-	0.712	-	0.016

- a. One age 200 m.y. - others 1000 - 2700 m.y.  
b. Ages from 1200 - 2200 m.y.  
c. Recent oceanic and continental basalts.  
d. Ages range from 0 to 60 m.y.  
e. East and west coast composite samples.  
f. Mid-Atlantic.

### Figure Captions

- Figure 1. Portion of a mass spectrometer isotope ratio analysis record. The 86 and 87 mass peaks are amplified 10 times in relation to the 88 mass peak.
- Figure 2.  $\text{Sr}^{87}/\text{Sr}^{86}$  in tektites, meteorites, terrestrial rocks and sea water. Inset shows the full range of measurements reported in the literature for granites and chondrites.
- Figure 3.  $\text{Sr}^{87}/\text{Sr}^{86}$  plotted against  $\text{Rb}/\text{Sr}$  for all eighteen individual tektites. Least squares line through these points is the 310 m.y. isochron.
- Figure 4.  $\text{Sr}^{87}/\text{Sr}^{86}$  plotted against  $\text{Rb}/\text{Sr}$  for averages of tektites from three groups. Least squares line through these points is the 400 m.y. isochron. The vertical and horizontal lines represent the range of the  $\text{Sr}^{87}/\text{Sr}^{86}$  and  $\text{Rb}/\text{Sr}$  ratios respectively, while the hash lines represent the value of the particular parameter for each sample. Numbers in parentheses are the number of samples from each area.

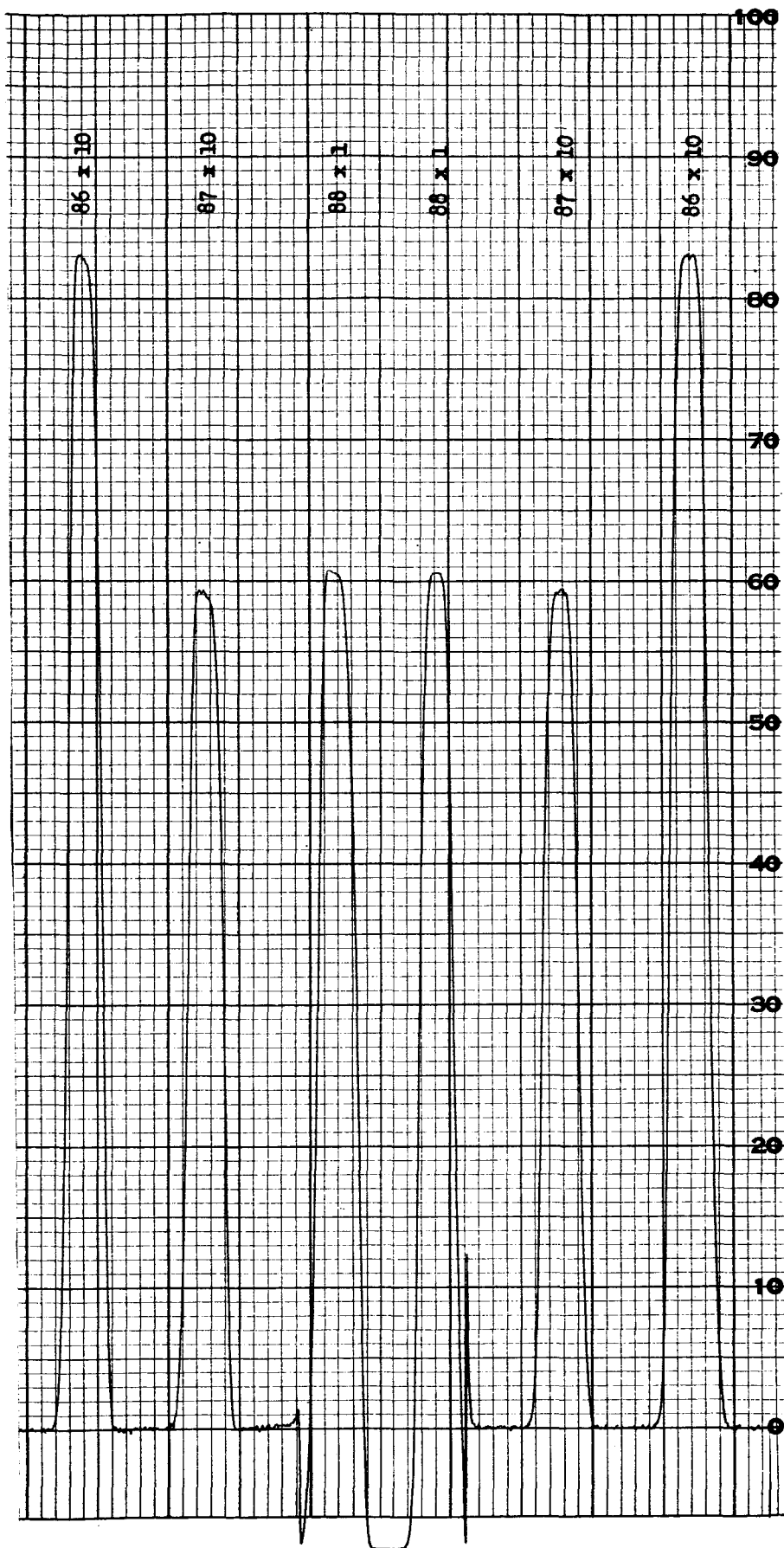
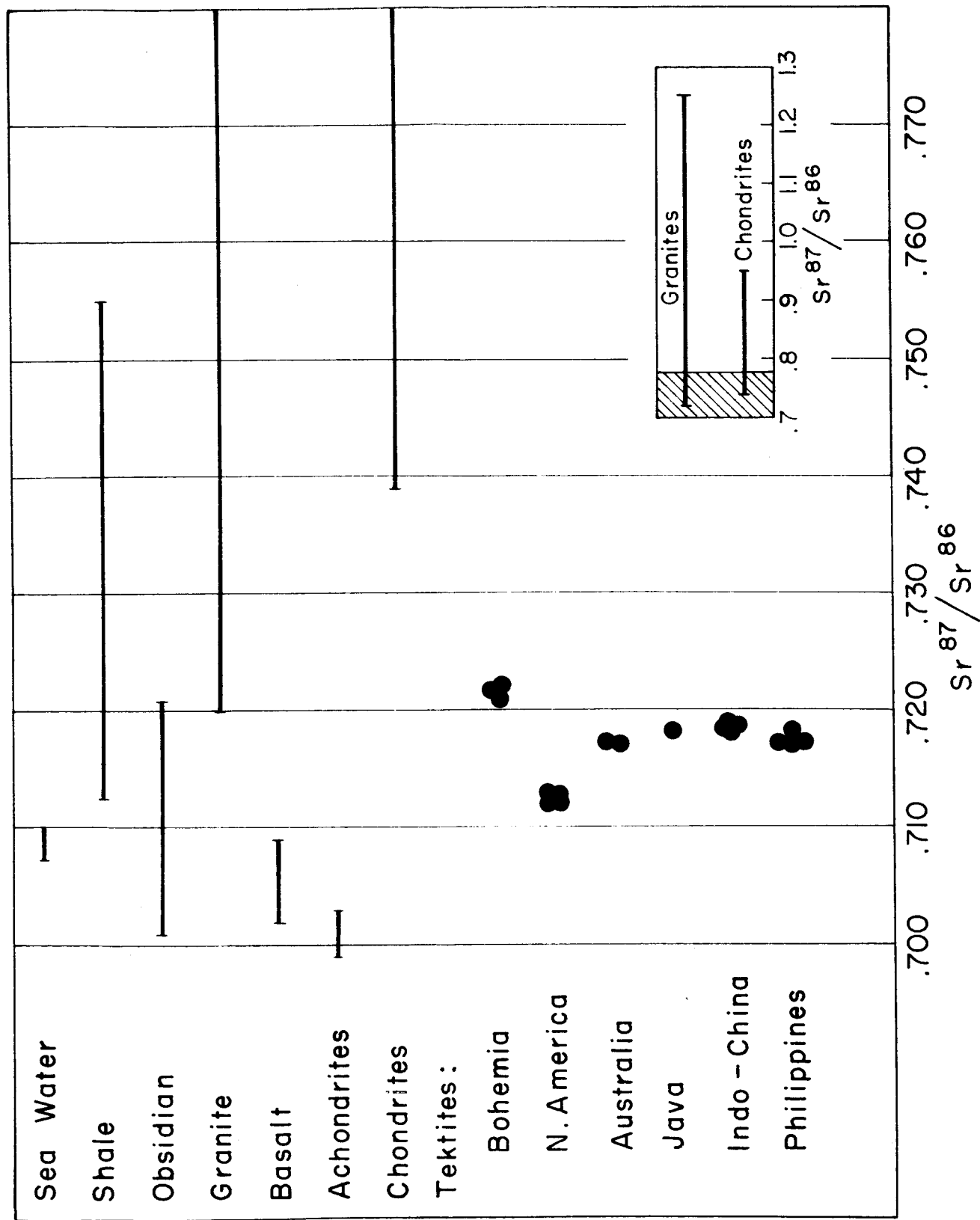


Fig. 1



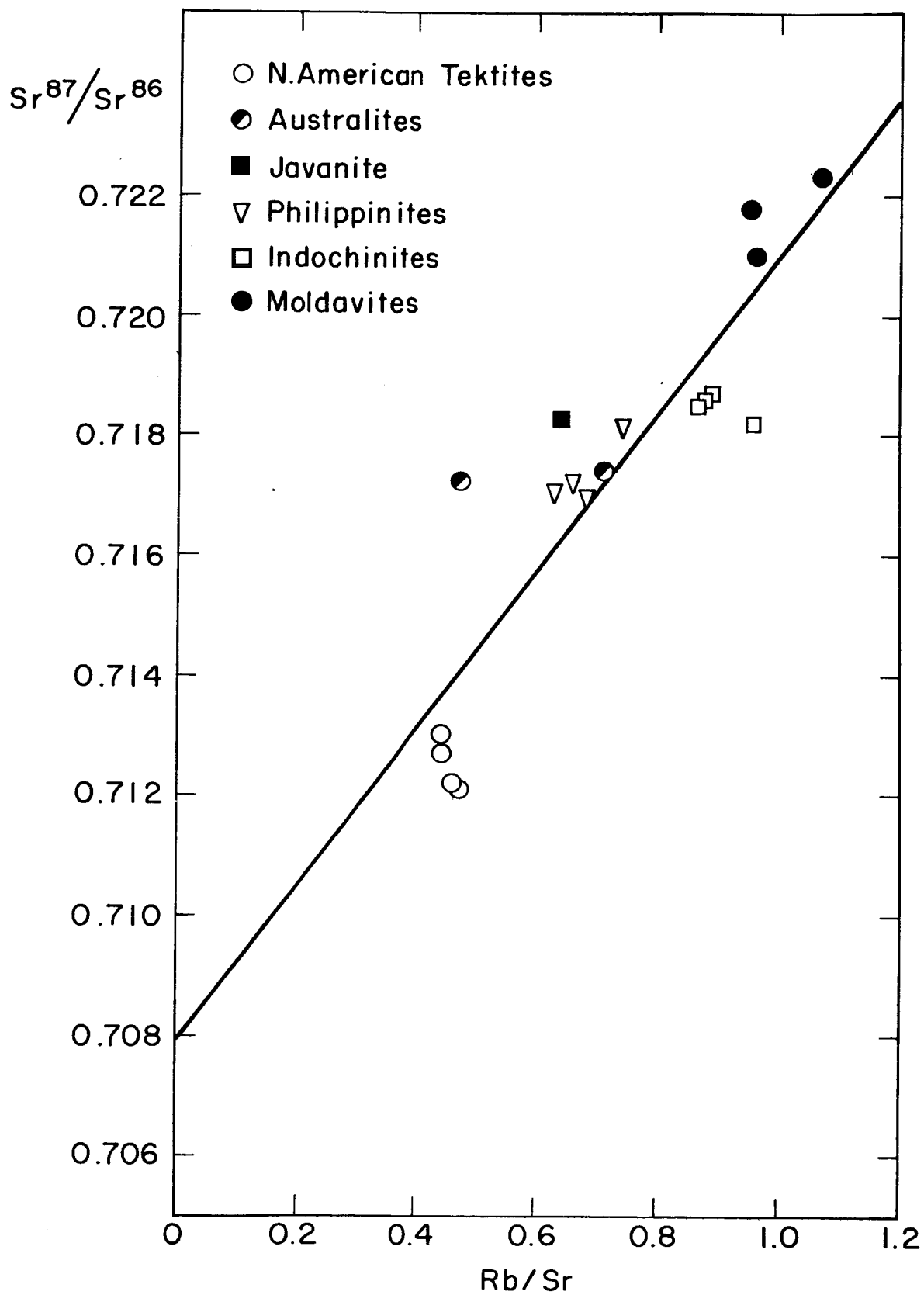


Fig. 3

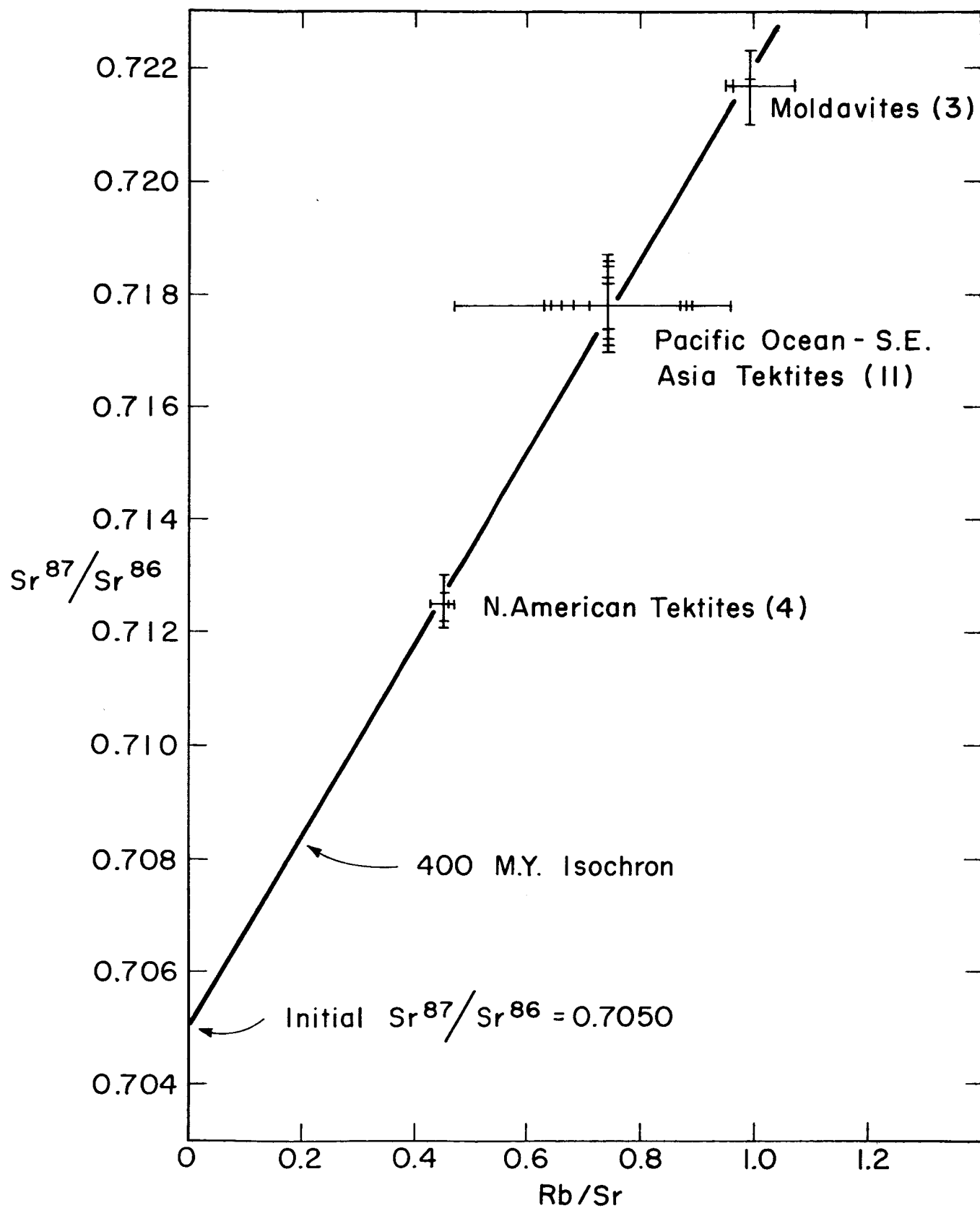


Fig. 4