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# ANALYSIS OF ALPHA EMITTERS IN THE CORAL, *FAVITES VIRENS*, FROM BIKINI LAGOON BY SOLID-STATE TRACK DETECTION

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**Abstract**—A quantitative method for the non-destructive analysis of alpha emitters in $CaCO_3$ matrices by solid-state track detection (cellulose nitrate) was developed.

0.4 pCi/g in an area of 4 $mm^2$ can be measured routinely; smaller concentrations can be determined but with a lower resolution. Calibration methods used were (a) a Pu source of 0.15 $\mu$Ci in conjunction with polycarbonate and $CaCO_3$ absorbers of different thickness (2–30 $\mu$m) and (b) a powdered coral sample from Enewetak Atoll which had been radiochemically analyzed for plutonium radionuclides, $^{241}$Am and other long-lived fission and activation products.

Slabs of a coral, *Favites virens*, from Bikini lagoon were analyzed. A quantity of the alpha emitters detected in regions of the coral identified with growth during the years of nuclear testing (1954, 1956 and 1959) are found in small discrete spots. Thin sections cut parallel to the direction of coral growth give different patterns of distributions. No such "hot spots" are evident in any post-test year growth sections although plutonium and other long lived fission and activation products were measured in these sections by radiochemical techniques.

## INTRODUCTION

MANY applications of solid–state track detectors have been made in several fields of engineering and science (Fl75). Alpha-particle detection using cellulose nitrate as the solid-state track detector has been developed to the stage whereby distribution of alpha emitters at very low concentration levels can be determined. The efficiency for the solid-state alpha track detector (Kodak LR-115, Type II) was determined using a very thin $^{239}$Pu calibrated source in conjunction with alpha energy degraders of polycarbonate and calcium carbonate films and a powdered coral sample of known plutonium and americium activities.

A coral, *Favites virens*, collected alive from Bikini lagoon in 1972 was sectioned for analysis. Relatively high radionuclide levels, including plutonium radionuclides and $^{241}$Am, were detected in the coral growth sections corresponding to the nuclear testing years of 1954, 1956 and 1958 (No75). Since it has been demonstrated (Ba74) that coral skeletons are excellent samplers for suspended particulate material in the water column it was of interest to determine whether the radionuclides detected in the coral-growth increments were taken up as dissolved species or were taken up, in part, through particulate incorporation.

Through the application of the solid-state alpha detector to slabs (1 cm thick) and thin

sections (30 $\mu$m thick) of the coral it is possible to establish the concentration and distribution of the alpha emitters in relation to the textural and structural features of the coral. The resolution of the method is on the order of 5 $\mu$m (Fl75) consequently it is possible to determine whether the alpha-emitters are concentrated in the coral skeleton, in void filling cements, or in other organisms (algae or bryozoa). These results should be of importance in understanding the biogeochemical processes influencing the distribution of alpha emitting nuclides in the marine environment.

## METHODS

### *Alpha particle detection in natural materials using solid-state track detectors*

A particular advantage of using solid-state track detectors for analysis is the very high resolution in determining the location of the element being detected. With the solid state track detector technique it is possible to measure alpha emitter concentrations in 1 $\mu$g of calcite if the concentration is above 50 pCi/g.

Careful microscopic analysis, both of the solid-state track detector and the sample being analyzed, is required to take advantage of this very high resolution. Consequently, most analyses are done at 625× magnification using a high-quality research microscope and diligently recording the data in a manner to allow correlation between the alpha-emitter distributions and the structural features of the sample under study.

A disadvantage of using solid-state track detectors for alpha-emitter analysis is the inability to distinguish the different alpha-particle energies and consequently the inability to detect which isotope has produced the observed track. To be more specific, the data reported in this study are based on total alpha-emitter concentration which means the observed tracks are due to any alpha emitter contained in the sample $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Am, $^{210}$Po and others that might exist in the environment. Consequently in our analyses of the coral samples a certain proportion of the tracks detected are due to the naturally occurring uranium and thorium isotopes and their decay products. Listed in Table 1 are the concentrations of selected alpha emitters in the 1954–1955 coral-growth increment. The values were either determined by radiochemical or analytical techniques or are representative background values expected for some alpha emitters obtained from the literature. Background uranium and thorium effects are of concern when the concentrations of alpha emitters being measured are in the range of 1 pCi/g or less. Accordingly, at concentrations measured in hot spots in the coral, *Favites virens*, the uranium and thorium background is negligible.

### *Calibration of cellulose nitrate with respect to alpha-particle detection*

It is important to establish the efficiency of the alpha detector in each matrix to be analyzed. Kodak LR-115 was calibrated by using two standards to measure the efficiency of registering alpha particles in two matrices, polycarbonate and calcite. The standards used are: (a) a 0.015 $\mu$Ci $^{239}$Pu source electroplated on to a stainless steel planchet to give a homogeneous and thin (Ra75) source of plutonium and (b) an analyzed powdered coral sample containing the alpha-emitting radionuclides $^{238}$Pu $^{239,240}$Pu and $^{241}$Am.

(a) *Calibration of the Kodak film by use of the electroplated Pu source.* Kodak film LR-115, Type II is sensitive to alpha particles having energies of less than 4 MeV while the alpha particle energies of the plutonium and americium isotopes are in the range of 5–5.5 MeV. Alpha particles emitted from a surface of material (the thickness of which exceeds the range of the alpha energy considered) within which the radioactive isotopes are uniformly distributed will have energies

*Table 1. Alpha emitting radionuclides in 1954–1955 coral growth section*

| Radionuclide | Concentration (pCi/g) | Comments |
|---|---|---|
| $^{239}$Pu | 21.6 | Determined by alpha and mass spectrometry following radiochemical separation |
| $^{240}$Pu | 17.4 | As above |
| $^{241}$Am | 17.6 | Determined by gamma spectrometry |
| $^{238}$Pu | 2.18 | Determined by alpha spectrometry |
| $^{210}$Pb ($^{210}$Po) | 1.34 | Determined by alpha spectrometry |
| $^{238}$U, $^{235}$U (U) | 2.4 | Determined by neutron activation and fission track detectors |
| $^{232}$Th | 0.0008 | Expected coral background levels from Dodge and Thomson (1974) |
| $^{230}$Th | 0.0006 | As above |
| $^{228}$Th | 0.005 | As above |
| $^{226}$Ra | 0.027 | As above |

ranging from zero to the maximum alpha-particle energy emitted by the isotope under study. Two types of alpha-particle energy degraders, polycarbonate and calcite, were used in conjunction with the electroplated $^{239}Pu$ source to calibrate the Kodak LR-115 film for this range of alpha-particle energies. For both the polycarbonate and calcite energy degrader studies it is assumed that the $^{239}Pu$ which has been electroplated on a stainless steel planchet is exceedingly thin (i.e. all the alpha particles emitted are of 5.1 MeV energy). Further, when the results of these experiments are applied to determining the alpha emitter concentration in a calcite matrix it is assumed that (a) the difference in sensitivity of alphas in the range of 5.0–5.5 MeV to that of the 5.1 MeV energy of $^{239}Pu$ is negligible, and (b) the alpha emitters are homogeneously distributed within the calcium carbonate matrix.

(1) *Calibration of Kodak LR-115, Type II using polycarbonate film as an energy degrader of alpha particles*

Kodak film LR-115, with a thickness of 10 $\mu$m (designated as Batch A henceforth) was irradiated with a known dose of alpha particles of select maximum energies by using the electroplated $^{239}Pu$ source covered with polycarbonate films of different thicknesses (4, 8, 12, 20, 24, 28, and 30 $\mu$m). This results in alpha particle energies in the range of 0–5 MeV. The irradiated detectors were etched (2.5 N NaOH, 58°C, 70 min) and the number of tracks counted in an area of 1.2 $mm^2$ (same area each time) by using 625× magnification.

The detection efficiency of the combination of Kodak film LR-115 and the polycarbonate is plotted against the polycarbonate thickness (Fig. 1). It can be seen that the Kodak LR-115 film is not capable of detecting alpha particle energies above about 4.5 MeV and that the sensitivity of the film depends upon the alpha particle energy. The highest sensitivity occurs at 4.1 MeV.

Values of the stopping power for alpha particles in a polycarbonate matrix have been published (Be68). A relative stopping power of 2:1 for alpha particles in calcium carbonate as compared to polycarbonate films was calculated by using Northcliffe and Schilling's techniques (1970) in conjunction with Benton's polycarbonate alpha particle energy loss data (1968). Assuming that (1) the alpha particle emitters are uniformly distributed in a calcite matrix, (2) the relative stopping power for alpha particles in a calcite matrix compared to a polycarbonate matrix is 2 to 1 and (3) the calibration experiment for polycarbonate is valid (Fig. 1), the relationship between alpha particle tracks detected and the concentration of alpha emitters can be calculated. Thus, for calcite it was calculated that one track/0.04 $mm^2$/30 day exposure is equivalent to 40 pCi of alpha emitters per g-calcite using Kodak Batch A. Kodak Batch B (Kodak film LR-115, Type II thickness of 6 $\mu$m, 1974), gives a value of 104 pCi/g for one track/0.04 $mm^2$/30 day exposure. Etching conditions for Batch B are 2.5 N NaOH, 25 min, 58°C.

(2) *Calibration of Kodak LR-115, Type II using calcite film as an energy degrader of alpha particles*

Calcite films of various thickness between

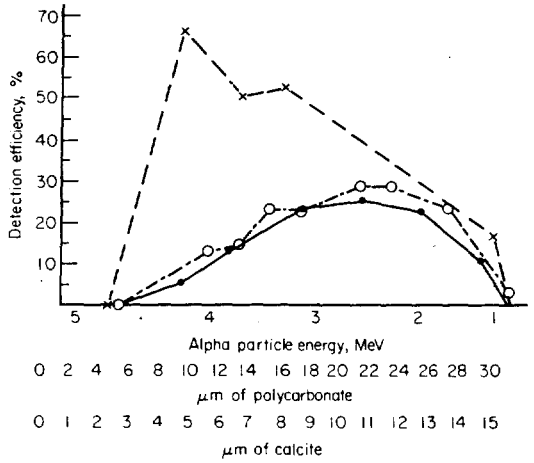


FIG. 1. Detection efficiency curves for Kodak LR-115, Type II cellulose nitrate using alpha particles from a $^{239}Pu$ source with polycarbonate and calcite energy degraders. 100% detection efficiency is one track per one alpha particle. ——— Batch A + polycarbonate, ——— batch B + polycarbonate —·—·—·— batch B + calcite.

2 and 15 $\mu$m were prepared by allowing calcite particles from a calcite suspension (particles less than 1 $\mu$m in diameter) to settle on the Kodak film. The thicknesses of the calcite were calculated by weighing the $CaCO_3$ (density of 2.7 g/cm$^3$) deposited on a known area of Kodak film. It is assumed that the stopping power of the air between the calcite grains is negligible. The curve labeled "Calcite" in Fig. 1 has been determined in this manner. The overlap of the two curves in Fig. 1 indicate that the relative stopping power of calcite to polycarbonate is about 2:1.

It can be calculated (Fig. 1) that 1 track/0.04 mm$^2$/30 day exposure is equivalent to 130 pCi/g when calcite is the energy degrader. Comparing this result to the value of 104 pCi/g for one track/0.04 mm$^2$/30 day exposure obtained from the polycarbonate experiments we find a difference of about 30%. The coral analyzed is aragonite; however, the calibration using calcite does not introduce significant error considering the other inaccuracies.

(b) *Calibration of the Kodak film by use of coral with known alpha emitter concentration.* An independent method for calibration of the Kodak film was accomplished by using a standard of known alpha emitter concentration in a calcium carbonate matrix (a powdered coral). The powdered coral sample contained $166 \pm 9$ pCi/g of plutonium and americium radionuclides consisting of $101 \pm 7$ pCi/g of $^{239}$Pu and $^{240}$Pu, $55 \pm 5$ pCi/g of $^{238}$Pu and $10.4 \pm 2$ pCi/g of $^{241}$Am. Batch A and Batch B of Kodak film LR-115 were exposed for 30 days to aliquots of this powdered coral sample giving a value of $174 \pm 17$ pCi/g based on Batch A polycarbonate calibration and $160 \pm 9$ pCi/g based on Batch B polycarbonate calibration. Both these values are within the precision ($166 \pm 9$) reported for the chemical method.

One track/0.04 mm$^2$/30 day exposure is taken as equivalent to 40 pCi alpha emitters/g-calcite for Batch A and 100 pCi/g for Batch B for all analyses discussed in this report. In contrast to the radiochemical data reported for this coral (No75) the activities reported here are not corrected for decay because concentration data on each individual radioactive isotope is not obtained by the solid-state track detection method.

## RESULTS

(1) *Analyses of slabs of the coral*

The first attempt at defining the alpha emitter distributions and concentrations by applying the solid-state track detector to a section of coral sample was done by using a slab of coral. The section selected contained the 1954–58 annual growth rings (No75). Cellulose nitrate film was applied directly to the slab and to three additional sections cut perpendicular to it (Fig. 2) and left for 30 days exposure. The analyses of detectors exposed to both sides of the slab show the following results (Figs. 3 and 4):

(a) Some of the alpha emitters in the 1954–1958 growth interval are concentrated in small volumes coinciding with damaged regions and holes in the coral texture (Figs. 3, 4b and c).

(b) "Hot spots" occurring in the coral growth bands equivalent to the years 1954 and 1956 are located within 4 mm$^2$ (Fig. 3). One "hot spot" was detected in a region of the coral we were unable to date using the annual band technique (No75) but it was apparently in a period of growth before 1954. It is possible that this spot designates the 1946 growth (the first nuclear test year at Bikini) but we cannot rule out possible chance contamination of the area during cutting operations.

(c) No "hot spots" were found on detectors exposed to post test year growth sections of coral.

(d) An alpha concentration of 800 pCi/g was obtained on very fine grained chalky material scraped from a void within the coral.

(e) A maximum concentration of 720 pCi/g was measured in 0.04 mm$^2$ of the 1954 growth band. This is an equivalent to 0.7 fCi of alpha activity.

(2) *Analyses of the thin sections of the coral*

To define more clearly the relationship between the coral fine structure and the alpha emitter locations, uncovered thin sections of the coral were prepared from the slabs cut

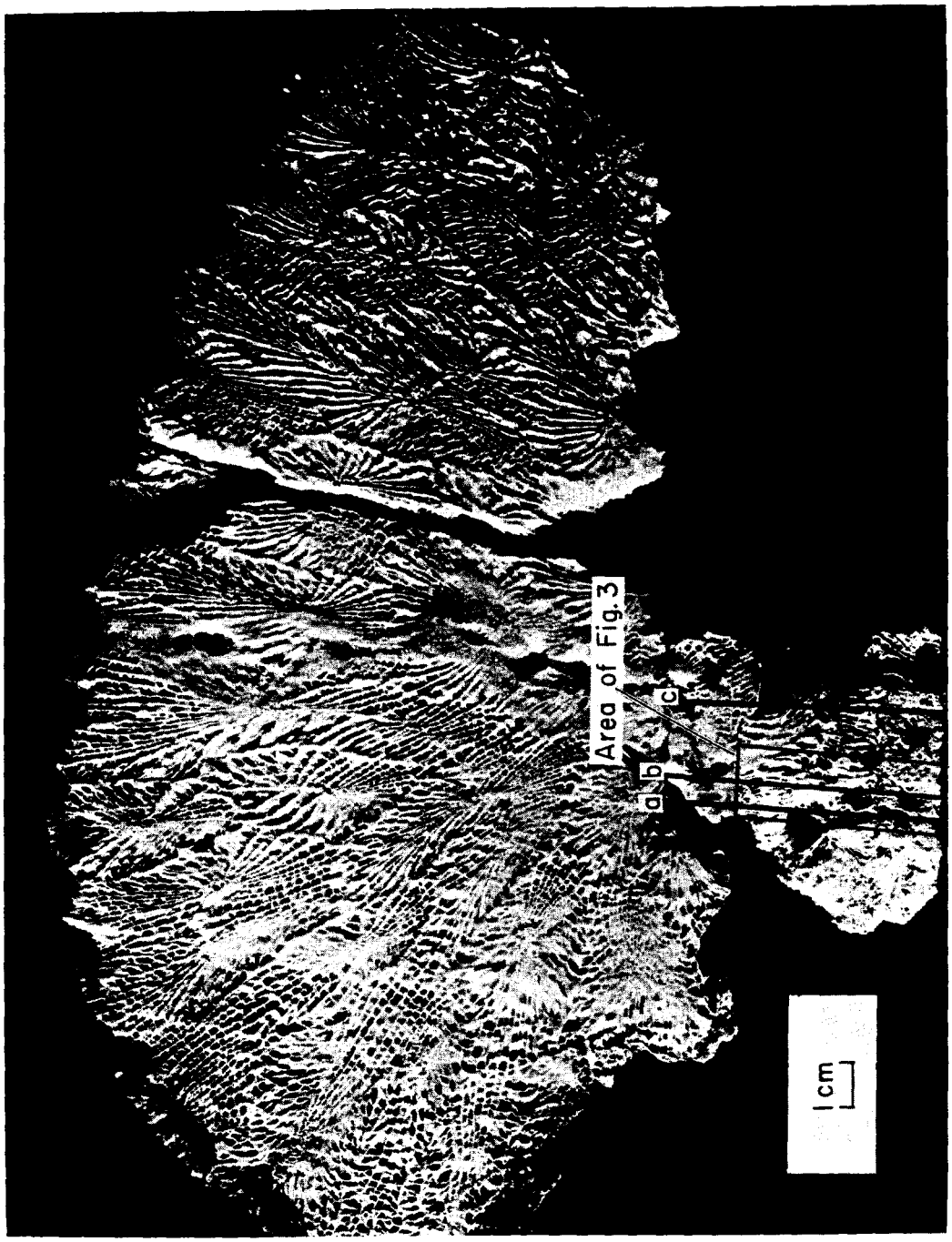


FIG. 2. Photograph of surface of coral *Favites virens* slab with outline of area analyzed and location of sections a, b and c (reflected light).


FIG. 3. Concentrations of alpha-emitters in a portion of the coral *Favites virens* slab (see Fig. 2). ——: boundary of areas counted. pCi/g less than 2, (a) 2–9, (b) 10–24, (c) 25–49, (d) 50–74, (e) 75–99, (f) 100–149, (g) 150–155.

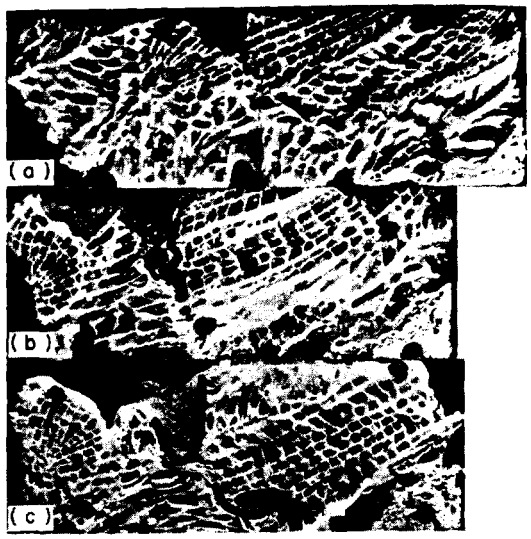


FIG. 4. Distribution of alpha-emitters in three thin slabs cut perpendicular to surface shown in Fig. 2. Scale is 1:1 (reflected light).

perpendicular to the surface A and shown in Fig. 4. The thin sections were covered by a Kodak film and after 30 days exposure the cellulose nitrate films were etched, studied and then replaced on to the coral thin section as close as possible to the same position as during exposure. This allows correlation between the alpha emitter location and specific petrographic features to within 5 $\mu$m. Figure 5 shows the results for thin section C. In actual practice the cellulose nitrate films and the thin sections were observed through the microscope simultaneously. At low magnifications both thin section and cellulose nitrate film were in focus, at high magnifications only one surface at a time could be focussed upon.

In material of the absolute fraction of alpha activity associated with the "hot spots", it is apparent that all the alpha activity within the coral is not homogeneously distributed. Rather, an analysis of the alpha "hot spots" in the 1954–1958 test year growth sections show:

(a) some of the alpha emitters are concentrated in small discrete ribbons rather than in broad bands,

(b) the alpha emitter distribution

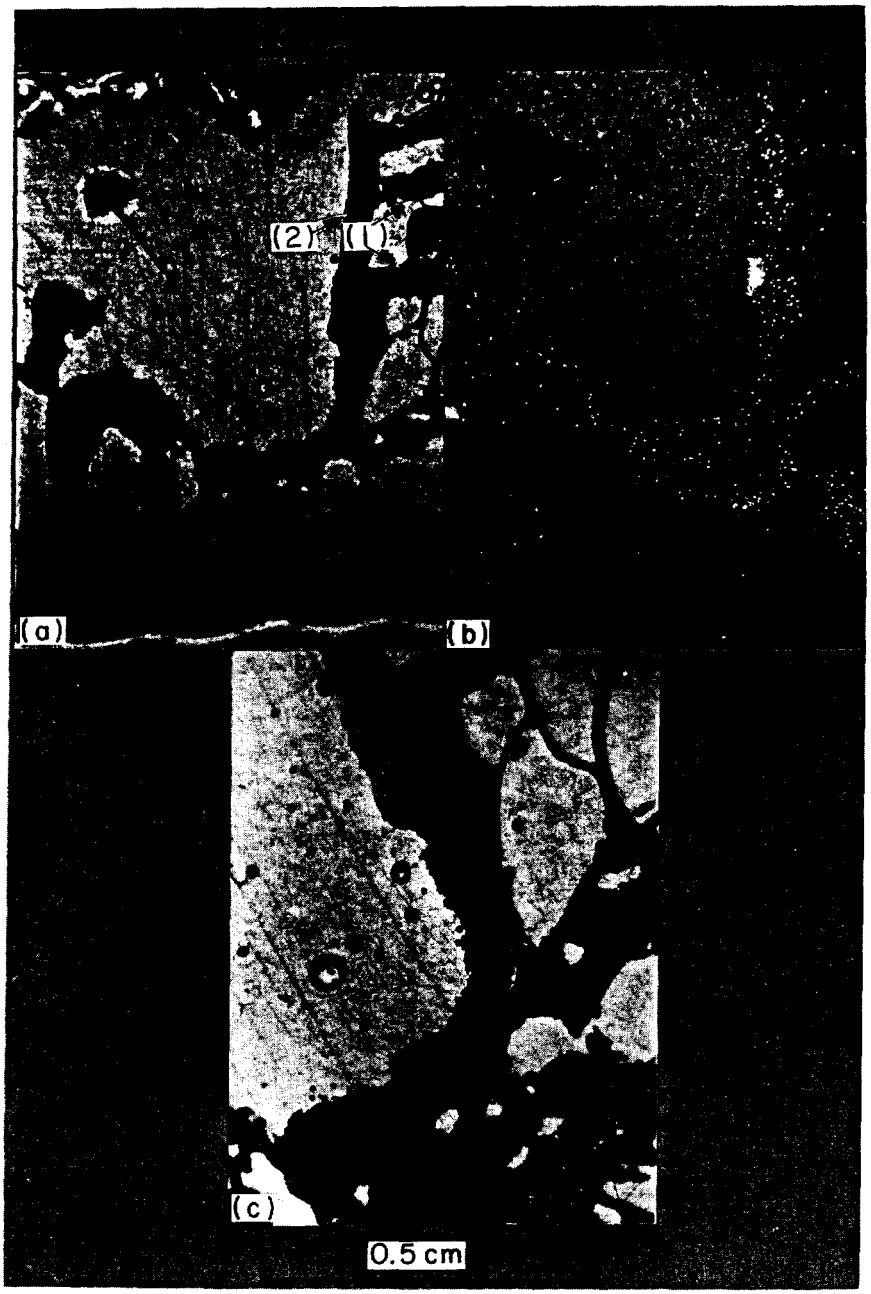


FIG. 5. 30 $\mu$m thin section (transmitted light) (a) Photomicrograph of thin section showing coral *Favites virens* texture (1) and Bryozoan-like texture (2). (b) Photomicrograph of cellulose nitrate detector at same scale as (a) showing the distribution of alpha-emitters in thin section c. Note that the high track densities coincide with non-coral material. (c) Photomicrograph of cellulose nitrate superimposed on portion of thin section c [different magnification than in (a) and (b)]. Left portion is composed of Bryozoan-like texture which shows many tracks. Upper right portion is coral and lacks tracks.

sometimes coincides with the annual growth bands (No75) but many times shows no correlation with the growth bands (Fig. 3),

(c) many high alpha emitter concentrations coincide with anomalous regions in the coral such as holes and/or infilling of other material (Figs. 3, 4 and 5),

(d) at least one of the voids contains fine-grained chalky material which contains very high concentrations of alpha emitters equivalent to the hottest spots found,

(e) although the alpha distribution does not always coincide with coral growth bands, it does correlate with areas which probably represent periods of coral growth interruption. Evidence for this interruption is obtained through thin-section analysis (Fig. 5) where it is seen that wherever there is high alpha-emitter concentration the coral structure is interrupted by a layer of unidentified skeletal debris with an algal or bryozoan like texture (J. Carew, personal communication).

## DISCUSSION

It is difficult to compare quantitatively results by this and the radiochemical method since it is almost impossible to obtain an average alpha concentration in a large volume of coral using the alpha-track technique. The lowest limit of detection using the track detectors is 50 pCi/g of alpha emitter in 1 $\mu$g of calcium carbonate. Since the average activity in the 1954–1955 growth section obtained by chemical analyses was 60 pCi/g (Table 1) we have no way of assessing if these "hot spots" account for most of the activity in the growth section (being diluted with very low activity coral to average 60 pCi/g) or, at the most, only 20% of the alpha activity in the growth section. To arrive at this latter figure of 20%, we need only assume the majority of the average 60 pCi/g alpha activity (at the 50 pCi/g level) determined radiochemically, would not be detected by the alpha-track technique.

The texture of material containing the higher levels of alpha radioactivity closely resembles the description of the types of small fallout particles resulting from near surface and water detonations at Bikini and Enewetak Atoll (Ad60). It is therefore quite probable that a number of the high activity alpha "hot spots" are merely evidence of small radioactive particulate inclusions containing, in addition to alpha active radionuclides, other fission and activation products. These radionuclides accumulated on the coral during periods of nuclear testing and subsequently incorporated by the coral. No such spots are in evidence in any post test year coral growth although plutonium radionuclides, other alpha emitters and some long-lived fission and activation products are easily measured in the coral growths by radiochemical techniques (No75). These observations suggest that particulate incorporation by corals occurs during extremely disturbed periods at the Atoll. During the periods of nuclear testing the coral incorporated trace elements, in this case alpha radioactivity, that are associated in large part with particulate debris as well as incorporating dissolved species from the water column. In periods of relative calm (post test years) the quantity of particulate material incorporated by this coral must decrease substantially which also suggests there is little resuspension of the radioactive sediments in the region of the lagoon where this coral was obtained.

Because of the alpha activities found in the coral (Table 1), it is very tempting to associate the "hot spots" with $^{238+239}$Pu and $^{240}$Pu and $^{241}$Am activity. It is unexpected to find high levels of $^{210}$Pb ($^{210}$Po) in the test year growth section; however we are presently in the process of attempting to analyze radiochemically several "hot" areas to determine what alpha emitters are present and how they are distributed throughout the "hot spot" regions.

## CONCLUSIONS

A quantitative method of non-destructive detection of alpha emitter concentrations in the range of 1.0–1000 pCi/g calcite with a resolution of 5 $\mu$m has been developed. The method was used to map the distribution of some alpha emitting radionuclides in the test year growth sections of a coral *Favites virens* from the Bikini lagoon.

This study has shown that application of the solid-state alpha track detector method is

a powerful tool for revealing the exact distribution patterns of alpha emitters in the range analyzed. Using the method in conjunction with thin section petrography shows that it is possible to assign the alpha activity to different matrices within the coral skeleton and structure, leading to better understanding of the factors which lead to the concentration of not only alpha emitters but trace elements as well in corals.

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