

Fig. 4. Saturation of enzyme by synthesized RNA compared to viral RNA. The experiment was carried out as described for Fig. 1. The circles refer to the values obtained with RNA isolated from virus particles in the experiment of Fig. 1, and the triangles to the rates obtained with the RNA synthesized in the experiment of Table 2. Since in the latter case the tem-plate was labeled with  $P^{32}$ ,  $H^3$ -UTP at  $1 \times 10^{\circ}$  count/min for each 0.2  $\mu$ mole was used to follow the synthesis. Preparations and counting as in Fig. 1.

Table 2. Sixty-fold increase in synthesis of viral RNA. A 1-ml reaction mixture contained the following, in micromoles: HCl, pH 7.4, 84; MgCl<sub>2</sub>, 12.8; CTP, ATP, UTP, and GTP, 0.8 each. Then 160  $\mu$ g of enzyme protein and 4  $\mu$ g of Q $\beta$ -RNA as template were added. The specific activity of UTP<sup>32</sup> was adjusted so that  $1 \times 10^5$ count/min are equivalent to 25.4  $\mu$ g of RNA. The mixture was incubated at 35°C at the indicated intervals, 20  $\mu$ l were removed, and acid-precipitable RNA was assayed as described in Fig. 1. At the end of the reaction, the RNA was purified as follows: the volume was adjusted to 2 ml with TM buffer (10<sup>-2</sup>M tris,  $5 \times 10^{-3}M$  MgCl<sub>2</sub>, pH 7.5) Water-saturated phenol (2 ml) was then added, and the mixture was shaken in heavy-wall glass centrifuge tubes (Sorvall, 18 by 102 mm) at 5°C for 1 hour. After separation of the water phase from the phenol by centrifugation at 11,000 rev/min for 10 minutes, another 2 ml of TM buffer was added to the phenol, and the mixture was shaken for 15 minutes at The phenol and water layers were separated again, and the two water layers were combined. The phenol was eliminated by two extractions with ether; the phenol was re-moved from the walls of the centrifuge tubes by completely filling the tubes with ether after each extraction. The ether dissolved in the water phase was then removed with a stream of nitrogen. The RNA was precipitated with one-tenth volume of potassium acetate (2M)and two volumes of cold absolute ethanol. The samples were kept for 2 hours at  $-20^{\circ}C$ before centrifugation (1 hr, 14,000 rev/min Sorvall SS 34 rotor). The pellets were dried in a vacuum desiccator for 6 to 8 hours at 5°C, and the RNA was then dissolved in 1 ml of buffer (10<sup>-2</sup>M tris, 10<sup>-2</sup>M MgCl<sub>2</sub>, pH 7.5). The radioactivity in the fraction precipitated with TCA was measured on 20  $\mu$ l portions of the final product from which the percentage recovery of synthesized RNA can be determined.

| Time<br>(min) | Specific activity<br>[count min <sup>-1</sup><br>(20 µl) <sup>-1</sup> ] | RNA<br>synthesized<br>(µg/ml) | Total<br>RNA |  |
|---------------|--|-------------------------------|--------------|--|
| 0             | 80   | 0                             | 4            |  |
| 60            | 4,917  | 63                            | 67           |  |
| 150           | 15,641   | 199                           | 203          |  |
| 210           | 20,113   | 256                           | 260          |  |
|               |  |                               |              |  |

tion to the ordinate indicates that the change from autocatalytic to linear synthesis occurred when approximately 1  $\mu$ g of RNA had accumulated. Synthesis continues for approximately 4 hours and then declines slowly. At the end of the experiment, the RNA synthesized corresponded to approximately 25 times the amount put in at the beginning.

The results are consistent with the implication that the product of the reaction can stimulate new enzyme molecules to activity. Thus enzyme recognizes the product as homologous to its own genome. To test this conclusion more directly, a 1-ml reaction mixture was set up as described in Table 2. The synthesis was allowed to proceed for 3.5 hours, by which time the increase of input material was more than 60-fold. The reaction was then terminated, and the RNA was purified by the phenol method, which yielded 55 percent of the synthesized product.

Figure 3 shows the distribution in a linear sucrose gradient of the synthesized product (judged by P32) as compared with  $Q\beta$ -RNA freshly isolated from virus. Much of the product has the 28S size characteristics of Q $\beta$ -RNA. There is evidence of some smaller fragments which may represent either abortive or incomplete syntheses.

Since the product was labeled with P<sup>32</sup>, it was necessary to examine its template function with another identifying isotope (H<sup>3</sup>-UTP). This examination was carried out as described in the legend for Fig. 4, which illustrates the response of the "replicase" to various input levels of the product (triangles) as compared with the original viral RNA (circles). The RNA syn-

thesized in the course of the 60-fold increase is as effective in serving as a template as the original viral RNA.

The data support the assertions that the reaction generated a polynucleotide of the same molecular weight (1 imes 10<sup>6</sup>) as viral RNA and that the "replicase" cannot distinguish this polynucleotide from its homologous genome. It is clear that the enzyme is faithfully copying the recognition sequence employed by the "replicase" to distinguish one RNA molecule from another.

The questions of whether identical replicas have been in fact produced, and whether the product contains all the information required to program the synthesis of complete virus particles are considered in a separate communication (9).

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   Abbreviations used: sRNA—soluble RNA; ATP, CTP, GTP, and UTP—adenosine cytidine, guanosine, and uridine triphosphate; UTP<sup>22</sup>—UTP labeled with P<sup>32</sup>; TCA—trichloreneetic acid roacetic acid.
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## Potassium, Rubidium, Strontium, Thorium, Uranium, and the Ratio of Strontium-87 to Strontium-86 in Oceanic Tholeiitic Basalt

Abstract. The average concentrations of potassium, rubidium, strontium, thorium, and uranium in oceanic tholeiitic basalt are (in parts per million) K. 1400; Rb. 1.2; Sr, 120; Th, 0.2; and U, 0.1. The ratio Sr<sup>87</sup> to Sr<sup>86</sup> is about 0.702, that of K to U is  $1.4 \times 10^4$ , and of Th to U is 1.8. These amounts of K, Th, U, and radiogenic Sr<sup>87</sup> are less than in other common igneous rocks. The ratios of Th to U and Sr<sup>87</sup> to Sr<sup>86</sup> suggest that the source region of the oceanic tholeiites was differentiated from the original mantle material some time in the geologic past.

Studies of submarine volcanics by Engel and Engel (1) suggest that low potassium tholeiites are a predominant primary magma erupted from the mantle and that alkalic basalts are magmatic differentiation products of

the tholeiites. The chemical properties of the tholeiites are fundamental to all hypotheses of terrestrial differentiation, heat flow of the earth, and crustal evolution.

Questions have arisen as to the

Table 1. Replicate analyses of uranium and thorium in the oceanic tholeiitic basalts.

| Sample               | U           | Th           | τь /π |  |  |  |  |
|----------------------|-------------|--------------|-------|--|--|--|--|
| (g)                  | (ppm) (ppm) |              | In/U  |  |  |  |  |
| Atlantic Ocean AD2-1 |             |              |       |  |  |  |  |
| 1                    | 0.16        | 0.13         | 0.8   |  |  |  |  |
| 1                    | .16         | .17          | 1.0   |  |  |  |  |
| 3                    | .16         | .15          | 0.9   |  |  |  |  |
| Mean                 | .16         | .15          | .9    |  |  |  |  |
|                      | Atlantic    | Ocean AD3-2  |       |  |  |  |  |
| 1                    | 0.10        | 0.29         | 2.9   |  |  |  |  |
| 1                    | .09         | .27          | 3.0   |  |  |  |  |
| Mean                 | .10         | . 29         | 2.9   |  |  |  |  |
|                      | Atlantic (  | Ocean AD5-11 |       |  |  |  |  |
| 1                    | 0.09        | 0.13         | 1.4   |  |  |  |  |
| 1                    | .09         | .13          | 1.4   |  |  |  |  |
| Mean                 | .09         | .13          | 1.4   |  |  |  |  |
|                      | Pacific     | Ocean PD3*   |       |  |  |  |  |
| 1                    | 0.08        | 0.19         | 2.4   |  |  |  |  |
| 1                    | .08         | .25          | 3.2   |  |  |  |  |
| 1†                   | .07         | .13          | 1.9   |  |  |  |  |
| 3                    | .10         | . 30         | 3.0   |  |  |  |  |
| 3                    | .09         | . 20         | 2.2   |  |  |  |  |
| 3                    | .10         | .21          | 2.1   |  |  |  |  |
| Mean                 | .09         | .21          | 2.5   |  |  |  |  |
|                      | Pacific (   | Ocean PD4-G  |       |  |  |  |  |
| 1                    | 0.08        | 0.14         | 1.7   |  |  |  |  |
| 1                    | .05         | .11          | 2.2   |  |  |  |  |
| 1†                   | .06         | .14          | 2.3   |  |  |  |  |
| 3                    | .07         | .12          | 1.7   |  |  |  |  |
| Mean                 | .07         | .13          | 2.0   |  |  |  |  |
| Average              | 0.10        | 0.18         | 1.8   |  |  |  |  |

\* Reproducibility of the isotope dilution for uranium and thorium at these levels of concentration is estimated about  $\pm$  10 percent. The large fractionation in the determined values of PD3 seems due to the sample heterogeneity.  $\ddagger$  The samples were decomposed by Na<sub>2</sub>CO<sub>5</sub> fusion. All others were decomposed by HF-HC10<sub>4</sub> followed by Na<sub>2</sub>O<sub>2</sub> fusion.

validity of the chondritic-earth model from the studies of strontium isotopes and the ratio of K to Rb (2), and from abundance ratios of uranium, thorium, and potassium (3, 4).

Accordingly, we determined the concentrations of potassium (1), rubidium, strontium, uranium, and thorium and the ratio,  $Sr^{87}/Sr^{86}$ , in samples of oceanic tholeiitic basalts. Some of the concentrations of potassium and rubidium have already been reported (2). The samples are from the Mid-Atlantic Ridge and from the flanks of the East Pacific Rise. Their petrography has been discussed earlier (1).

The concentrations of rubidium, strontium, uranium, and thorium were determined by isotope dilution techniques (Tables 1 and 2).

The concentrations of uranium and thorium in the oceanic tholeiites are very low compared to the average value of basalts (4) and are about half of that in the tholeiites from Japan and Hawaii (5). However, the Th/U ratio, averaging about 2, is similar to that of the Japanese tholeiites but lower than that of Hawaiian tholeiite. The lower Th/U ratio is in marked contrast with Th/U ratios of approximately 4 as-12 NOVEMBER 1965 Table 2. Amounts of potassium, rubidium, and strontium (in parts per million) and  $Sr^{87}/Sr^{86}$  in the oceanic tholeiitic basalts. Reproducibility of the isotope dilution for rubidium and strontium is about  $\pm 10$  and  $\pm 5$  percent, respectively. We estimate the internal precision of  $Sr^{87}/Sr^{86}$  to be  $\pm 0.0004$ .

| Sample No. | K*              | Rb             | Sr     | Sr <sup>87</sup> /Sr <sup>86</sup> | K/Rb           | Rb/Sr  | 10 <sup>-4</sup> (K/U) |
|------------|-----------------|----------------|--------|------------------------------------|----------------|--------|------------------------|
|            |                 |                | Atlant | ic Ocean                           |                |        |                        |
| AD2-1      | 1160<br>(1345)† | 1.14<br>(1.42) | 134    | 0.7024                             | 1020<br>(950)  | 0.0085 | 0.7                    |
| AD3-2      | 1990            | 2.63           | 109    | .7027                              | 760            | .024   | 2.0                    |
| AD5-11     | 1100            | 0.66           | 150    | .7021                              | 1760           | .0044  | 1.2                    |
|            |                 |                | Pacifi | c Ocean                            |                |        |                        |
| PD3        | 1990<br>(1560)  | 1.06<br>(1.14) | 86     | 0.7016                             | 1890<br>(1370) | 0.012  | 2.2                    |
| PD4-G      | 920<br>(641)    | 0.45<br>(0.35) | 98     | .7017                              | 2022<br>(1830) | .0045  | 1.3                    |
| Average    | 1414            | 1.19           | - 115  | .702                               | 1490           | 0.011  | 1.4                    |

\* Concentrations of potassium are from (1).  $\dagger$  The values in parentheses are by Gast (2).

Table 3. Concentrations (in parts per million) and ratios of potassium, rubidium, strontium, uranium, and thorium in terrestrial material and meteorites.

| K                                      | Rb       | Sr            | U               | Th           | 10 <sup>-4</sup> (K/U) | K/Rb  | Rb/Sr |
|--|----------|---------------|-----------------|--------------|------------------------|-------|-------|
| •••••••••••••••••••••••••••••••••••••• |          |               | Granite         |              |                        |       |       |
| 37,900 (4)*                            | 130 (12) | 400 (12)      | 4.75 (4)        | 18.5 (4)     | 0.8                    | 290   | 0.2   |
|  |          |               | Intermedia      | ate          |                        |       |       |
| 18,000 (3)                             | 60 (12)  | 600 (12)      | 2.0 (3)         | 7.4 (3)      | 0.9                    | 300   | .1    |
|  |          |               | Alkalic bas     | salt         |                        |       |       |
| 12,000 (12)                            | 30 (12)  | 600 (12)      | 0.7 (5)†        | 3.0 (5)      | 1.7                    | 400   | .05   |
|  |          | Tholeiitic ba | salt other than | submarine th | holeiite               |       |       |
| 5,000 (12)                             | 10 (12)  | 230 (12)      | 0.2 (5)         | 0.4 (5)      | 2.5                    | 500   | 0.04  |
|  |          | Su            | bmarine oceani  | c tholeiite  |                        |       |       |
| 1,400                                  | 1.2      | 115           | 0.1             | 0.18         | 1.4                    | 1,400 | 0.01  |
|  |          | A             | chondrite, high | calcium      |                        |       |       |
| 330 (2)                                | 0.20(2)  | 80 (2)        | .081 (4)        | . 51 (4)     | 0.4                    | 1,650 | 0.003 |
|  |          | A             | Ichondrite, low | calcium      |                        |       |       |
| 9 (4)                                  |          |               | .0021 (4)       | .0059 (4)    | 0.4                    |       |       |
|  |          |               | Chondrite       | es           |                        |       |       |
| 845 (4)                                | 2.8 (2)  | 11 (2)        | 0.012 (4)       | 0.040 (4)    | 7.0                    | 300   | 0.25  |

\* Numbers in parentheses indicate the references.  $\dagger$  The average only from Hawaii and Japan. The high K/U ratios of alkalic and tholeiitic basalts might be due to the limited samples.

signed for the terrestrial average and ordinary chondrites (3, 4, 6) and even smaller than the carbonaceous chondrite, Orgeuil (7). These differences may be explained if differentiation has occurred in the source region of the tholeiites under the ridges, rise, and island arcs (5, 8). The lead isotope studies of the tholeiites which are currently in progress (9) indeed suggest that the source region of the tholeiite under the ridge and the rise has been differentiated from an original material some time in the geologic past and the Th/U ratio changed from 4 to 2 at the source region from which the tholeiites were generated, even though the isotopic composition of lead in the tholeiites is of very primitive (less radiogenic) character.

The Sr<sup>87</sup>/Sr<sup>86</sup> ratios of the tholeiites are lower than in most other basalts (10-12). The ratios of the tholeiites from the Mid-Atlantic Ridge show a similar value to those from St. Helena and Ascension Island, which were reported as the lowest found to date; however the ratios from the East Pacific Rise appear to be even somewhat lower.

The concentrations of the reported elements in the tholeiites are further compared to those in terrestrial material and meteorites in Table 3. The data point up the extremely primitive nature of the oceanic tholeiites, that is, amounts potassium, rubidium, strontium, of uranium, and thorium are generally lower, Rb/Sr ratios are lower, and K/Rb ratios are higher than other basalts and approach the values in basaltic achondrites, as pointed out by Gast (2). However, the K/U ratio of the tholeiites is distinct from either that of basaltic achondrites or chondrites but similar to that of 10<sup>4</sup> assigned for the terrestrial average (3, 4). The ratio is 3.5 times larger than that of basaltic achondrites and 5 times smaller than that of chondrites. If the tholeiites are produced by partial melting of an upper mantle material and enriched in uranium relative to alkali metals, the K/U ratio of the upper mantle material should be higher than the observed value or the terrestrial average.

Except for one Mid-Atlantic sample (AD3-2), the Rb/Sr ratios of these samples are actually too low to generate the observed Sr<sup>87</sup>/Sr<sup>86</sup> values, if the earth began with a  $Sr^{87}/Sr^{86}$  ratio of 0.6984 as suggested by Hedge and Walthall (11) and Gast (13, as rectified by Hedge and Walthall). This relation is additional evidence for some sort of differentiation in the past in the source material of these magmas. Either the source had a slightly larger value of Rb/Sr than observed for a long period of time (minimum 0.02 for 4.5 billion years), with the differentiation being recent or a much larger value of Rb/Sr early in earth history, for a short period of time. An example of the first case is a magma not representative of its source, perhaps because it was generated by partial melting of the source material. One example of second case could be a Rb/Sr of about 0.024 for the first 3.5 billion years of earth history and 0.011 for the last 1.0 billion years. In this second example, the magma could be representative of its source but the source has changed during geologic time. We prefer the second case because studies with lead isotopes suggest a rather long differentiation age (9).

The crust-mantle differentiation is most pronounced within a few hundred kilometers of the outer layers of the earth, and most radioactive elements accumulated in these upper layers (8, 14). Tatsumoto (5) suggested that tholeiites may be generated from the basic residual layer in the upper mantle, from which most radioactive elements were previously transported to the crust. If this mode of derivation is the case, we are only comparing extremely differentiated products with achondrites, chondrites, or carbonaceous chondrites, and the contents of potassium, uranium, and thorium in most of the mantle may be different from that observed in oceanic tholeiites. However, the mantle-crust differentiation in oceanic regions is not pronounced compared to that of the continents (14); therefore the chemical character of the oceanic tholeiites is of primary importance as a guide to estimate the chemical composition of the upper mantle. M. TATSUMOTO

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## Thunderstorm Electrification and Raindrop **Collisions and Disjection in an Electric Field**

Abstract. Raindrop collisions in an electric field selectively transfer charges of one sign to the larger disjected drops. The disjected drops, falling away from the smaller drops, separate free charge to establish electric fields as large as those observed in thunderstorms.

An experimental method for obtaining reproducible collisions between freely falling water drops has been described (1). With this method it has been found that whenever the relative kinetic energy between two falling drops approaches that observed in typical natural rainclouds, the collisions result in forceful tearing apart of the drops and scattering of the drop remnants. This mechanical action is called "disjection."

With the use of two "synchrodroppers" which are located at different levels and whose relative phase is regulated by electric circuits, it is possible to secure serial collisions between pairs of drops whose sizes and relative kinetic energy are controlled. Drop collisions were arranged to take place between two insulated horizontal parallel plates excited by a direct current of 5000 volts; an electric field intensity of 600 volt/cm was thereby established between the plates. Holes 3 cm in diameter were drilled in the centers of the plates, permitting the freely falling drop pairs to fall into the region of high electric field and to collide there; the secondary dropsthat is, those formed by collisionthen continued on through the bottom hole and were caught in a highly insulated Faraday cage. This cage was connected to an electrometer and recorder to measure the convected free drop charges.

Direct collisions between the drop pairs produced a neutral mixture of numerous highly charged positive and negative secondary drops. On the other hand, primary drops encountering an off-center (eccentric) collision usually develop into a linear distribution in which the drops at the bottom are considerably larger than those at the top. An intermediate step in the development of such a drop distribution is shown in Fig. 11 of reference 1, which clearly shows the excess water accumulating on the bottom side of the disjected drop pair. Eccentric collisions are far more probable in nature than direct collisions. Accordingly, my coworkers and I considered it important to determine experimentally whether the selective production of larger secondary drops at the bottom of the generated distribution had important electrical effects.

In order to evaluate the free electrical charges produced by disjection in an electric field, we arranged for a continuous series of drops having radii of 0.2 cm to overtake and collide serially with drops having radii of 0.15 cm, at a relative velocity of 170 cm/sec. The smaller drops were arranged so as to have a small horizontal component of velocity; the larg-