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Variation in the Content of Uranium and Thorium across Igneous Contacts

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Abstract

Recently, variations in the content of uranium and thorium were traced across igneous contacts of the two intrusions of Tanakami and Koya with practically continuous samples by the use of a combined technique involving an alpha-scintillation counter and a radioscope. The results of this determination show that the types of the distribution of alpha-radioactivity across igneous contacts found by a radioscope depend mostly on the content of uranium. On the other hand, the thorium content varies markedly from sample to sample, but the running averages of it near the contact are nearly constant and equal to that of the inner part of the intrusive body.

Introduction

As reported before,[1) 2) 3) 4)] the profile curves showing the alpha-radioactivity distribution across igneous contacts were found to be classifiable into five types[4)] which are presumably corresponding to the conditions under which the contact phenomena took place. It has been a matter of great concern to determine to which elements these radioactivity distributions are due.

HATUDA and the writer offered a method for determining the content of uranium and thorium in rocks by means of a combined measurement with an alpha-scintillation counter and a radioscope with a Lauritsen element.[5)] By this method uranium and thorium content across igneous contacts have been obtained.

Analytical procedure

The procedure of these measurements using the alpha-scintillation counter and the radioscope with a Lauritsen element was fully described in the other paper.[5] Hence, in this paper it will be sufficient to give the formulae with a brief explanation. The uranium and thorium content, U and Th in ppm are given as

$$Th = 40.7\ S - 1.21\ n \quad \cdots\cdots (1)$$

$$U = 29.9\ S - 1.08\ Th \quad \cdots\cdots (2)$$

where S is the shifting rate of the radioscope and n the counting rate of alpha-scintillation counter.

Samples

Along two of the typical traverses across igneous contacts which had been proved experimentally to be remarkably different in radioactivity distribution,[3) 4)] sampling was made. So far as conditions of exposure permit, the samples were taken as nearly as possible at a regular interval of 10 cm over the whole range of the sampling. The locations of these traverses are Koya, Kyoto Pref. and Shishitobi (Tanakami), Shiga Pref. In order to clarify the distribution of elements semi-quantitative spectrographical analyses had been tried.[4)] The variation of alpha-radioactivity observed by the radioscope and beta-radioactivity by the low-background gas-flow counter were also shown in the previous papers.[3) 4)]

The variations in the grain-size of the essential minerals, in the quantities of quartz, orthoclase and plagioclase by semi-modal analyses and in the content of the halo included in biotite are shown in Fig. 1.

Results

The variations in the alpha-radioactivity obtained by the above-mentioned two different methods with practically continuous samples from Shishitobi and Koya were shown in Figs. 2-1 and 2-2, respectively.

Using the values of the alpha-radioactivity thus obtained, the distributions of uranium and thorium have found by the Eqs. (1) and (2), and are shown in Fig. 3-1 and Fig. 3-2.

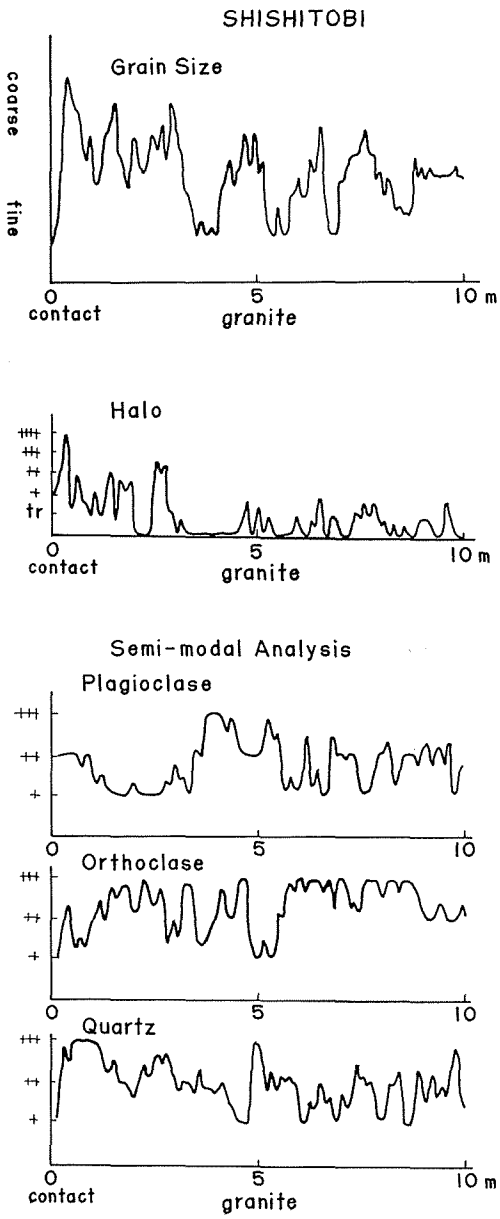

Fig. 1 The variations in the grain-size, the relative amount of quartz, orthoclase and plagioclase, and in the abundance of halo.

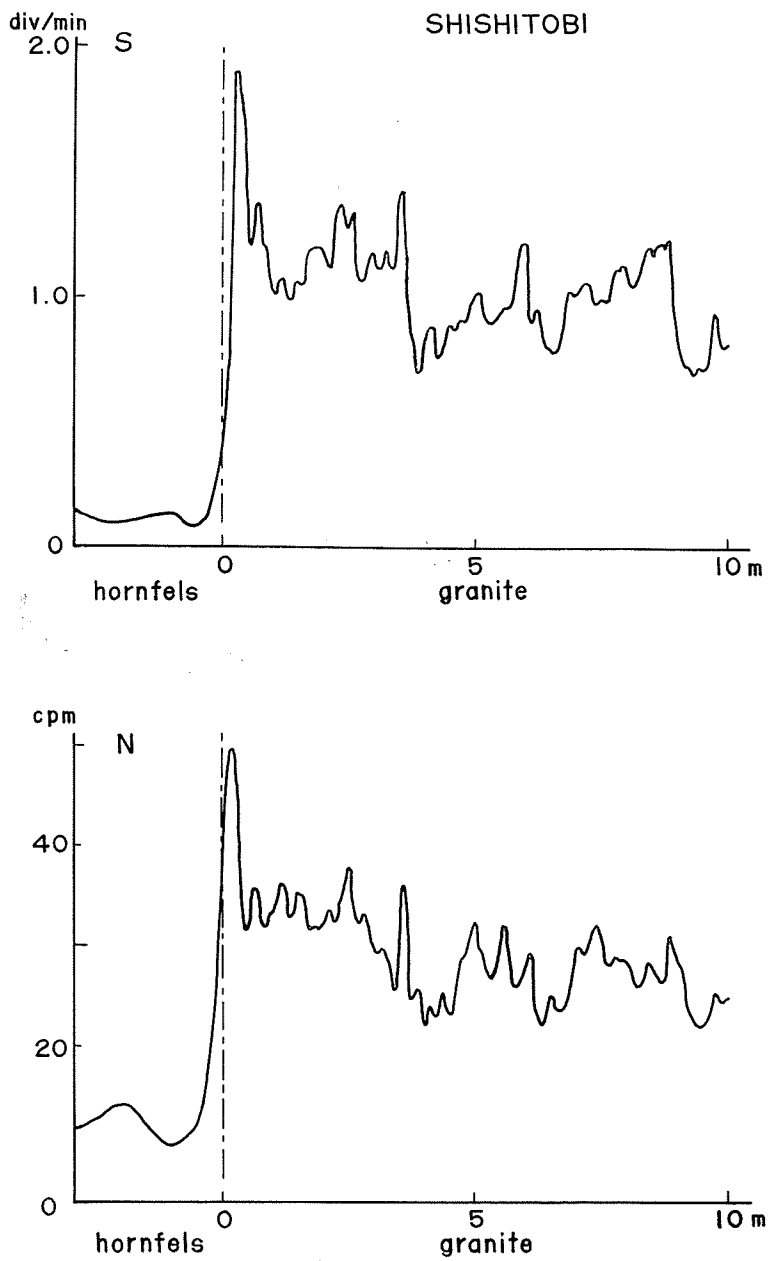

Fig. 2-1 The variation in the alpha-radioactivity obtained by the radioscope (the upper figure) and by the scintillation counter (the lower figure). (Shishitobi)

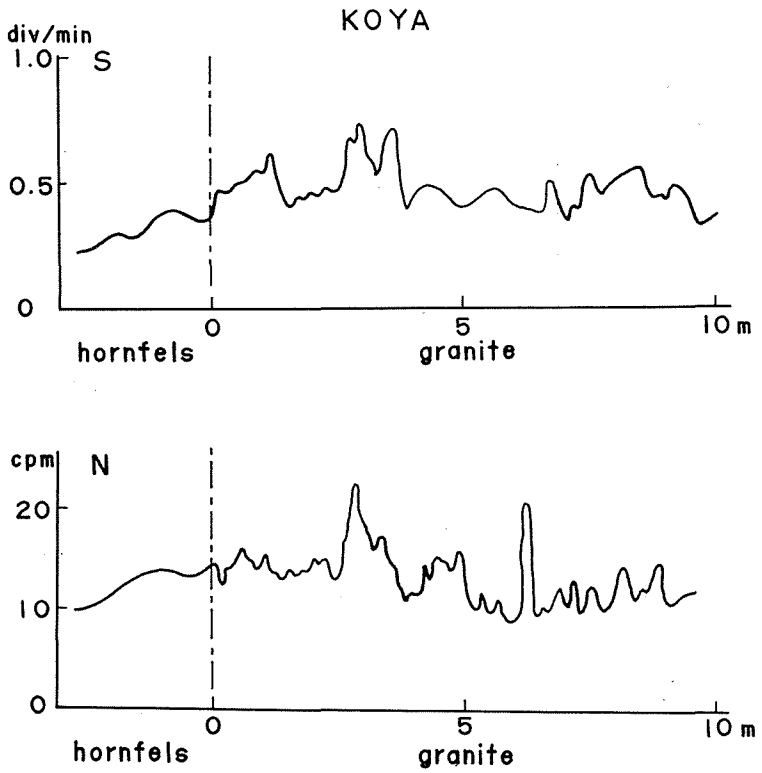

Fig. 2-2 The variation in the alpha-radioactivity obtained by the radioscope (the upper figure) and by the scintillation counter (the lower figure). (Koya)

Discussion of the results

The variation of the alpha-radioactivity across Shishitobi contact observed by the radioscope was classified as the first type.[1) 4)] The difference between the alpha-radioactivity by the radioscope and that by the alpha-scintillation counter is accounted for by the different sensitivities to uranium and thorium of the two apparatuses.

As shown in the Fig. 2-1, the variation of alpha-radioactivity by the alpha-scintillation counter (lower) has many points of likeness to that by the radioscope (upper). So it is also to the variation of uranium content obtained by the combined measurement (Fig. 3-1). As a general tendency, the concentration of uranium decreases as the distance from the contact boundary

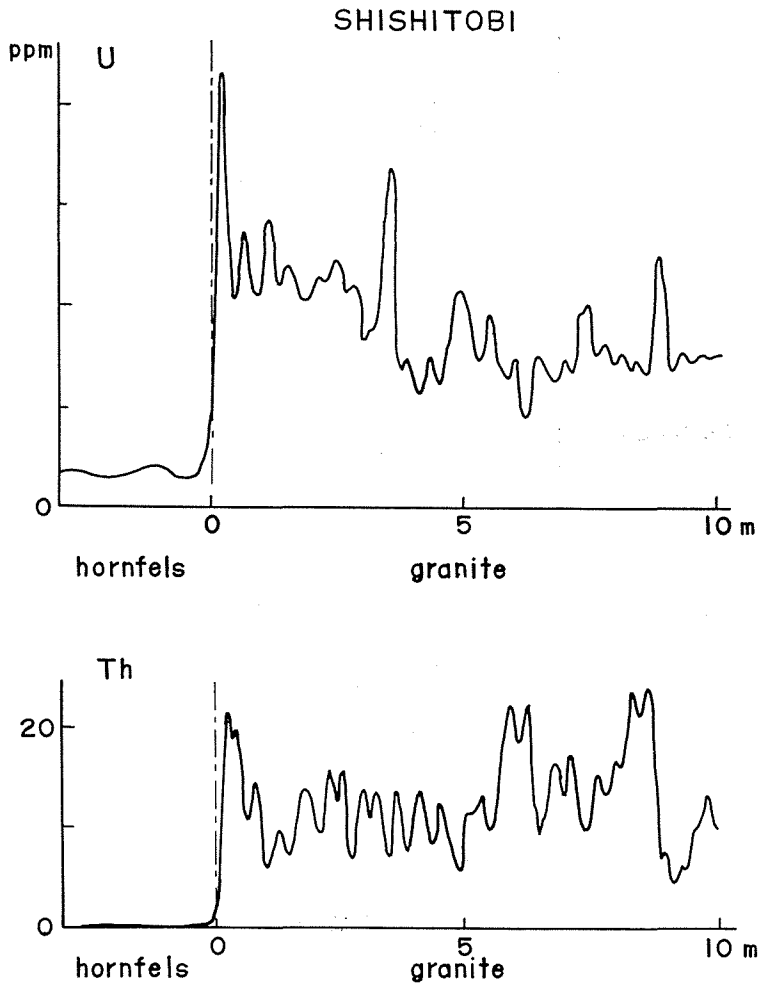

Fig. 3-1 The distributions of uranium (the upper figure) and thorium (the lower figure). (Shishitobi)

increases, while thorium behaves in the reverse manner, showing a slight increase towards the center of the intrusive mass.

The variation of the alpha-radioactivity across Koya contact observed by the radioscope was classified as the third type.[1) 4)] The similarity in the profile curves of the alpha-activity across Koya by the radioscope and by the scintillation counter is less than that in the case of Shishitobi. The tolerable

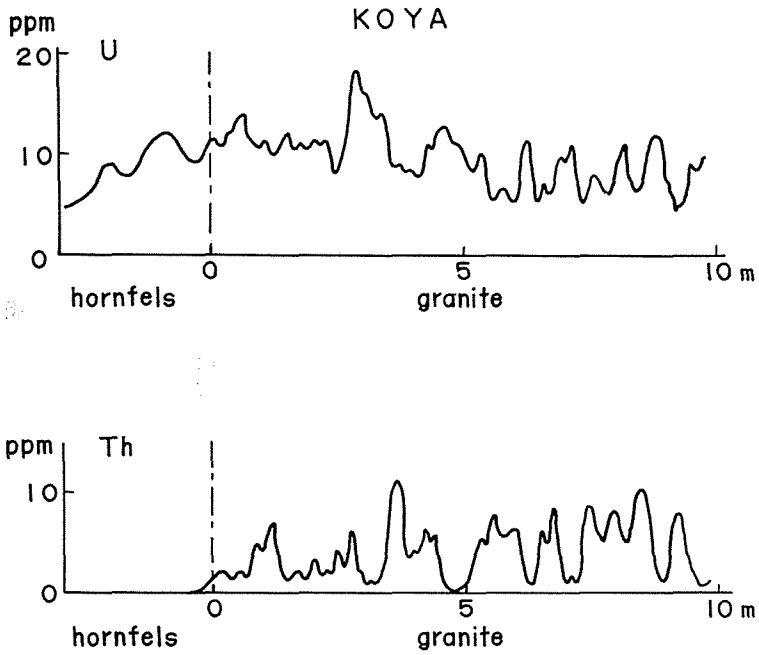

Fig. 3-2 The distributions of uranium (the upper figure) and thorium (the lower figure). (Koya)

quantities of uranium were found in hornfels, but thorium scarcely was found there. The thorium content varies more markedly than the uranium one, showing a tendency of increasing against the contact plane. This difference can be attributed to the different geochemical character of the uranium and thorium.

The uranium is an easily leachable element, namely, during the magmatic cycle, uranium as well as thorium were in the tetra-valent state, and the crystallization of the both elements goes parallel owing to the close similarities in ionic radius, but at a very late magmatic stage, a change took place which caused the uranium to be oxidized into a hexa-valent one, easily movable with the hydrothermal solution, leaving the thorium behind to crystallize. The observed results may be well explained by this relation.

The variations of Th/U across these contacts revealed the similar tendency of an increase with increasing distance from the country rock (Fig. 4).

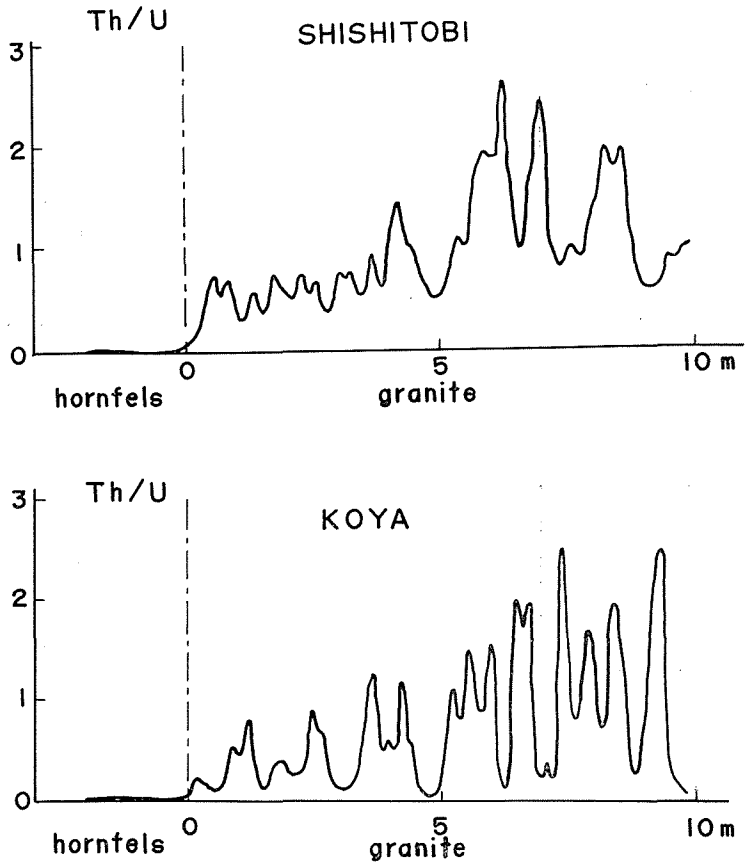

Fig. 4 The variations of *Th/U* across Shishitobi contact (the upper figure) and Koya contact (the lower figure)

The factors controlling the distribution of uranium and thorium in granitic bodies

In the previous paper it was inferred from the viewpoint of radioactivity that the distribution of volatile matter in an intruded body may be associated with the difference in temperature and pressure between granitic melt (or partial melt) and country rocks.

It was reported [4] that there was no remakable difference in the distribution of K^{40} content in intrusives of the different types classified by the alpha-radioactivity. This fact suggests that the conditions during the crystallization of potash-feldspar have little difference in any granitic bodies. Besides, as the

uranium moves as a volatile matter under large differences in temperature and pressure between granitic body and country rocks, it can be deposited in quantities within thin layer of the contact, but this tendency is less remarkable with thorium, as has been mentioned.

Near the contact in shallower depth, the temperature of the intrusive body falls abruptly, giving rise a sharp gradient of temperature. Accordingly, the vapour tension is so decreased that the diffusion of volatile matter from inner part towards the contact zone takes place briskly. On the contrary, with a small difference of temperature and pressure which is conceivable near the contact of great depth, uranium may be deposited equally over the intrusive body. This concept is supported by the variation of grain-size across the igneous contact of a granitic body.

Diffusion of uranium may be compared with the diffusion of pollution in air through the grid. The pollution in air at a high velocity diffuses in a state of turbulence, but in air at a low velocity, it does so in a steady manner. In the case of type Ⅰ,[4] the uranium diffuses through the interstices of the crystals of quartz and feldspar and so on in a condition of turbulence, but in the case of types Ⅱ, Ⅲ and Ⅳ,[4] it does so in a steady manner.

A two dimensional case may be assumed, i. e., the part of $x \geq o$ was intrusive body and that of $x \leq o$ country rock. The cooling history and conditions of an intrusive body were described in the previous paper.[4] Hence in this paper, the diffusion problem of uranium only was treated.

The diffusion of uranium along the x-axis will be described as the equation,

$$\frac{\partial c}{\partial x} = \frac{\partial}{\partial x}\left\{\kappa(x)\frac{\partial c}{\partial x}\right\} \quad \cdots\cdots (5)$$

where c is the concentration of the uranium, t the time, x the horizontal co-ordinate, and $\kappa(x)$ the horizontal diffusion coefficient. Firstly, we suppose the surface of intrusive body $x=o$ to be impenetrable or almost impenetrable to the diffusion of radioactive elements.

We shall take the chemical gradient $\frac{\partial c}{\partial x}$ to be equal to the temperature gradient at time, t.

In the steady state, $\kappa(x)$ may be constant, then we get from Eq. (5),

$$\frac{\partial c}{\partial t} = \kappa(x)\frac{\partial^2 c}{\partial x^2} \quad \cdots\cdots (6)$$

In the turbulent diffusion, $\kappa(x)$ also may depend on the temperature gradient. In this case, we shall take the following boundary conditions:

$$\kappa(x)\frac{\partial c}{\partial x} \underset{x\to o}{\longrightarrow} 0 \quad ; \quad c \underset{x\to h}{\longrightarrow} c_o \quad \cdots\cdots (7)$$

where h is the width of the turbulent region; the zone of $o \leq x \leq h$ is the turbulent part and the part of $x \geq h$ is in a steady state.

Now, we shall find the basic solution of the equation (5). When an instantaneous uniform distribution in a granitic body is observed at the moment $t = o$, this solution satisfies the initial condition:

$$c(x, t) \underset{t \to o}{\longrightarrow} c_0 \quad \cdots\cdots (8)$$

In accordance with the similarity theory for the turbulence through the parallel screen developed by LAUFER (1950), the coefficient of $\kappa(x)$ has the form,

$$\kappa(x) = \Theta_{ij}(x) \quad \cdots\cdots (9)$$

where Θ_{ij} is called the unidimensional energy spectrum tensor and shown in the next figure.

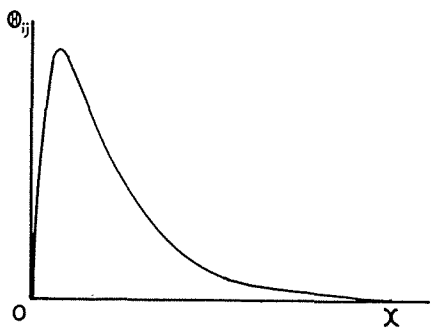

Fig. 5 The variation of Θ_{ij} towards x-direction

From this conception, the variations in the content of radioactive elements in the homogeneous medium, in the case of the steady diffusion and in the case of the turbulent diffusion, may roughly be shown in the next figure.

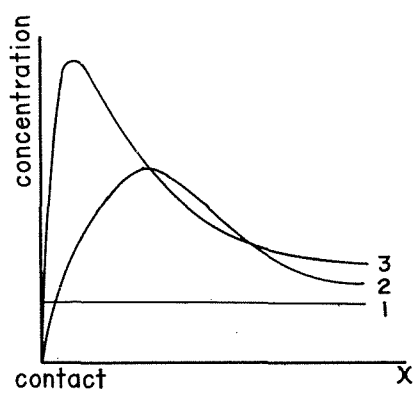

Fig. 6 The variation in radioactive element in the homogeneous medium
1: Steady diffusion
2: Steady diffusion with a moderate temperature gradient
3: Turbulent diffusion with a severe temperature gradient

The radioactive elements in rocks were found generally as the impurities in or around crystals, so that these heterogeneous areas are apt to cause irregularity in distribution, especially in the latter case. This description of the diffusion of uranium may be linked very closely with the result of the measurement, as the first approximation.

A part of the variations of the uranium content across igneous contacts may be explained in this way. On the other hand, the variation of thorium content seemd to suggest the absence of such diffusion as considered in the case of uranium in hexa-valent form.

Conclusion

1) The types of the alpha-radioactivity distribution across contact found by a radioscope depend mostly on the content of uranium, showing less relation with that of thorium.

2) The variation of thorium content is more conspicuous than that of uranium across igneous contacts.

3) The variation of Th/U across the two contacts showed a similar tendency of increase with the increasing distance from the country rock.

4) Uranium is more leachable than thorium.

5) At a very late magmatic stage, uranium is oxidized from tetra- to hexa-valent compound and carried about with the hydrothermal solution, leaving the thorium behind to crystallize.

6) The variation observed in alpha-radioactivity by the radioscope is well explained by the diffusion of uranium in a steady state, but in the case of type I, the turbulent diffusion must be taken into consideration.

Acknowledgements

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References

1) HATUDA, Z. and S. NISHIMURA, Mem. Coll. Sci., Univ. Kyoto, B. **23**, pp. 285-295 (1956)

2) HATUDA, Z. and S. NISHIMURA, Mem. Coll. Sci., Univ. Kyoto, B. **25**, pp. 115-123

(1958)
3) NISHIMURA, S., Mem. Coll. Sci., Univ. Kyoto, B. **28**, pp. 255-264 (1961)
4) NISHIMURA, S., Mem. Coll. Sci., Univ. Kyoto, B. **28**, pp. 265-284 (1961)
5) HATUDA, Z. and S. NISHIMURA, in this volume, pp. 215-226 (1964)
6) LAUFER, J., Jour. Aero. Sci., **17**, p. 277 (1950)