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# Neutron Activation Analysis of Japanese Standard Rock Samples II*

By

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**Abstract**

Non-destructive instrumental neutron activation analyses were carried out for determining four major and fourteen trace elements in Japanese standards rock samples, JA-1, JB-1a, JG-1a, JF-1, JGb-1, JR-1, JLk-1, JLs-1 and JDo-1, which have been issued from the Geological Survey of Japan (GSJ). Redetermination of major and trace elements in JA-2, JB-2, JG-2 and JP-1, which have been reported in the previous report, was also performed together with the above samples. Revised results were presented.

## 1. Introduction

In the previous paper (WAKABAYASHI, 1987), analytical results of Japanese geological standard rocks JG-2, JG-3, JR-2, JA-2, JA-3, JP-1, JB-2, JB-3 and JF-2 issued from the Geological Survey of Japan (GSJ) by non-destructive instrumental neutron activation analyses (INAA) were reported. Following to the above work, results of six igneous reock samples JA-1, JB-1a, JG-1a, JF-1, JGb-1 and JR-1, and of recently issued three sedimentary rock samples JLk-1, JLs-1, and JDo-1 are presented here. As for igneous rocks, these results are compared with "1986 consensus values" which have been compiled by GSJ (ANDO, MITA and TERASHIMA, 1987).

In addition, remeasurement of JA-2, JB-2, JG-2 and JP-1 was performed. These results and previous ones are compiled and presented in the appendix section of this report.

## 2. Experimental

The experimental procedures are similar to those used before (WAKABAYASHI, 1987) except for determination of long-lived nuclides of JLs-1 and JDo-1. The procedure is described briefly below.

Each sample was weighed accurately (about 10 mg, except 30 mg of JF-1 and 20 mg of JGb-1) and irradiated for 10 minutes in JRR-4 reactor at a flux of $8\times10^{13}$ neutrons/cm$^2$·sec in the Japan Atomic Energy Research Institute (JAERI) at Tokai.

* The previous report by F. WAKABAYASHI, *Bull. Natn. Sci. Mus., Tokyo, Ser. E*, **10**, 13–19 (1987) should be titled as "Neutron Activation Analysis of Japanese Standard Rock Samples I".

Irradiated samples were cooled about 20 hours, and then produced $\gamma$-ray activities were measured by a Ge(Li) $\gamma$-ray spectrometer without any chemical treatment. For long-lived nuclides, measurements of $\gamma$-activities were carried out for several times at the intervals of 3 or more weeks. JB-1 and W-1 were used as standards. JG-1 (granodiorite, issued from GSJ in 1976) was adopted for the standards of Cs., Rb and Ta.

For determination of JLs-1 and JDo-1, another method was employed because these rocks contain elements which hardly activated by only 10 minutes' neutorn irradiation. About 10 mg of each rock was accurately weighed, sealed in a small quartz tube, then irradiated for about 270 hours in JRR-2 reactor of JAERI at a flux of $3\times10^{13}$ neutrons/cm$^2$·sec. After cooling for about 2 months, produced $\gamma$-activities were measured. Measurements were carried out for three times at two months' intervals. Two rock samples DTS-1 (dunite, issued from the U. S. Geological Survey) and JG-1 were used as standards, and treated in the same way as JLs-1 and JDo-1.

Published best values used as standards, as well as nuclear data for target and produced nuclides used for determination of respecitve elements in this work are summarized in Tables 1, 2 and 3 respectively, except the data listed in the previous paper (WAKABAYASHI, 1987).

Table 1. Published best values used as standards for the analyses.*

| For samples irradiated in JRR-4 reactor | | | | For samples irradiated in JRR-2 reactor | | |
|---|---|---|---|---|---|---|
| Element | W-1** | JB-1*** | JG-1*** | Element | DTS-1** | JG-1*** |
| *Major elements* (%) | | | | | | |
| | | | | Fe | 6.04 | — |
| *Trace elements* (ppm) | | | | | | |
| Cs | — | — | 10.2 | Ce | — | 46.6 |
| Hf | 2.67 | 3.4 | — | Co | 133 | — |
| Rb | — | — | 181 | Cs | — | 10.2 |
| Ta | 0.50 | 3.6 | 1.7 | Eu | — | 0.76 |
| | | | | Sc | 3.6 | — |
| | | | | Sr | — | 184 |
| | | | | Tb | — | 0.84 |
| | | | | Th | — | 13.5 |
| | | | | Yb | — | 2.7 |
| | | | | Zn | 45 | — |
| | | | | Zr | — | 108 |

* Other values used as standards in this work were listed in the previous report; F. WAKABAYASHI, *Bull. Natn. Sci. Mus., Tokyo, Ser. E*, **10**, 13–19 (1987).

** F. J. FLANAGAN, *Geochim. Cosmochim. Acta.*, **37**, 1189 (1973).

*** A. ANDO *et al.*, *Geostand. Newslett.*, **11**, 159 (1987).

Table 2. Nuclear Data used in this work-1—Target nuclides.*

| Element | Nuclide | Isotopic abundance** (%) | Cross section*** (barns) | Produced nuclide (decay scheme & half life****) |
|---|---|---|---|---|
| Cs | $^{133}$Cs | 100 | 27 | $^{134}$Cs |
| | | | 2.5 | $^{134m}$Cs (IT, 2.91h) |
| Hf | $^{180}$Hf | 35.100 | 14 | $^{181}$Hf |
| Rb | $^{85}$Rb | 72.165 | 0.40 | $^{86}$Rb |
| | | | 0.047 | $^{86m}$Rb (IT, 1.017m) |
| Sr | $^{84}$Sr | 0.56 | 0.3 | $^{85}$Sr |
| | | | 0.59 | $^{85m}$Sr (IT, e$^-$, 1.1258h) |
| Yb | $^{168}$Yb | 0.13 | $3.5\times10^3$ | $^{169}$Yb |
| | | | | $^{169m}$Yb (IT, 46s) |
| Zr | $^{96}$Zr | 17.38 | 0.055 | $^{95}$Zr |
| | | | | $^{95}$Nb |

* Data for other nuclides used in this work were listed in the previous report; F. WAKABAYASHI, *Bull. Natn. Sci. Mus., Tokyo, Ser. E*, **10**, 13–19 (1987).

** After IUPAC-CAWIA report, *Pure Appl. Chem.*, **56**, 675 (1984).

*** C. W. LEDERER and V. S. SHIRLEY ed., "Tables of Isotopes" 7th ed., Wiley & Sons (1978).

**** Nuclides which were measured through their decay product only.

Table 3. Nuclear data** used in this work-2—Measured nuclides*.

| Element | Nuclide | Half life | $\gamma$-ray energy used (keV) | $\gamma$-ray intensity (%) |
|---|---|---|---|---|
| Cs | $^{134}$Cs | 2.062y | 604.710 | 97.6 |
| | | | 795.867 | 85.4 |
| Hf | $^{181}$Hf | 42.39d | 132.94 | 35.9 |
| | | | 482.00 | 80.6 |
| Rb | $^{86}$Rb | 18.66d | 1076.69 | 8.78 |
| Sr | $^{85}$Sr | 64.84d | 513.996 | 99.27 |
| Yb | $^{169}$Yb | 32.022d | 177.2144 | 21.5 |
| | | | 197.9581 | 34.9 |
| Zr | $^{95}$Zr | 64.02d | 756.729 | 54.5 |
| | $^{95}$Nb | 34.97d | 765.789 | 99.79 |

* Data for other nuclides used in this work were listed in the previous report; F. WAKABAYASHI, *Bull. Natn. Sci. Mus., Tokyo, Ser. E*, **10**, 13–19 (1987).

** E. BROWNE, R. B. FIRESTONE and V. S. SHIRLEY, "Tables of Radioactive Isotopes" Wiley & Sons (1986).

## 3. Results

Results are tabulated in Tables 4, 5 and 6. Each value in the tables was obtained through averaging at least three data which had been measured from time to time. The quoted errors in the tables are the standard deviations of the reproducibilities.

In Tables 4 and 5, 1986 consensus values for igneous rock series reported by ANDO, MITA and TERASHIMA (1987) are listed together with the results of this work. The results for igneous rocks are generally in good agreement with 1986 values as shown in the tables, which could certify the reliability of the data.

The main components of JLs-1 are CaO (55.02%) and $CO_2$ (43.86%), and those of JDo-1 are MgO (18.40%), CaO (34.12%) and $CO_2$ (46.87%) (ANDO, TERASHIMA, *et al.*, 1987). Among these elements, only $^{47}$Ca's peaks were observed in this work.

Table 4. Results of neutron activation analyses of standard rock samples JA-1, JB-1a and JG-1a.

| Element | JA-1 | | JB-1a | | JG-1a | |
|---|---|---|---|---|---|---|
| | '86 value* | This work | '86 value* | This work | '86 value* | This work |
| *Major elements* (%) | | | | | | |
| Fe | 4.86 | 5.01±0.08 | 6.36 | 6.5±0.1 | 1.43 | 1.45±0.03 |
| Na | 2.86 | 2.84±0.07 | 2.03 | 2.16±0.04 | 2.53 | 2.53±0.04 |
| K | 0.68 | 0.66±0.03 | 1.21 | 1.24±0.03 | 3.35 | 3.37±0.08 |
| Mn | 0.12 | 0.121±0.005 | 0.12 | 0.112±0.004 | 0.05 | 0.044±0.001 |
| *Trace elements* (ppm) | | | | | | |
| Ce | 13.2 | 15.6±0.9 | 67 | 59±2 | 47.1 | 37±1 |
| Co | 11.8 | 12.0±0.5 | 39.5 | 39.1±0.8 | 5.7 | 6.0±0.2 |
| Cs | 0.64 | — | 1.2 | — | 11.4 | 10.0±0.3 |
| Eu | 1.2 | 1.13±0.05 | 1.5 | 1.60±0.04 | 0.72 | 0.74±0.03 |
| Ga | 17.3 | 14±2 | 18 | 17±2 | 17 | 15±3 |
| Hf | 2.4 | 2.6±0.2 | 3.4 | 3.0±0.2 | 3.7 | 3.7±0.1 |
| La | 5.5 | 5.9±0.6 | 38 | 39±2 | 23 | 18±1 |
| Lu | 0.46 | 0.41±0.06 | 0.33 | 0.24±0.04 | 0.53 | 0.33±0.03 |
| Rb | 11.8 | — | 41 | 37±11 | 180 | 152±12 |
| Sc | 28.4 | 28.6±0.3 | 29 | 27.6±0.3 | 6.6 | 6.04±0.08 |
| Sm | 3.6 | 3.6±0.2 | 5.2 | 5.4±0.2 | 4.5 | 4.2±0.2 |
| Ta | 0.1 | — | 2.0 | 1.66±0.09 | 1.7 | 1.8±0.1 |
| Th | 0.82 | 0.97±0.08 | 8.8 | 7.9±0.3 | 12.1 | 10.4±0.2 |
| Zn | 90.6 | 95±14 | 82 | 66±13 | 38.8 | 35±6 |

JA-1 Andesite. Hakone volcano, Old Somma lava (Augite-hypersthene andesite) Quaternary, Manazuru-machi, Kanagawa Prefecture. split 6, position 45.

JB-1a Basalt. Replacement sample of JB-1**. split 8, position 68.

JG-1a Granodiorite. Replacement sample of JG-1***. split 8, position 100.

* A. ANDO *et al.*, *Geostand. Newslett.*, **11**, 159 (1987).

** JB-1 Basalt. Kitamatsuura basalt (Alkali basalt, Titanaugite-olivine basalt) 7.6 Ma, Myokanji Toge, Sasebo, Nagasaki Prefecture.

*** JG-1 Granodiorite. Sori granodiorite (Biotite granodiorite) 85 Ma, Azuma-mura, Gunma Prefecture.

Table 5. Results of neutron activation analyses of standard rock samples JF-1, JGb-1 and JR-1.

| Element | JF-1 | | JGb-1 | | JR-1 | |
|---|---|---|---|---|---|---|
| | '86 value* | This work | '86 value* | This work | '86 value* | This work |
| *Major elements* (%) | | | | | | |
| Fe | 0.056 | 0.055±0.001 | 10.60 | 11.2±0.2 | 0.67 | 0.68±0.03 |
| Na | 2.63 | 2.19±0.05 | 0.913 | 0.95±0.01 | 3.04 | 3.08±0.05 |
| K | 8.343 | 8.3±0.4 | 0.22 | 0.21±0.01 | 3.69 | 4.2±0.2 |
| Mn | 0.0008 | — | 0.13 | 0.162±0.005 | 0.077 | 0.083±0.004 |
| *Trace elements* (ppm) | | | | | | |
| Ce | 4.3 | 3.7±0.1 | 8 | 7.6±0.4 | 49 | 52±2 |
| Co | 0.2 | 0.112±0.006 | 61.6 | 68.6±1.0 | 0.65 | 0.85±0.10 |
| Cs | 2.2 | 1.89±0.05 | 0.27 | — | 20.2 | 21.1±0.7 |
| Eu | 0.85 | 0.71±0.02 | 0.61 | 0.67±0.03 | 0.31 | 0.28±0.02 |
| Ga | 18.1 | 15±2 | 18.9 | 20±4 | 17.6 | 32±3 |
| Hf | 1.3 | 0.90±0.03 | 0.84 | 1.4±0.1 | 4.7 | 4.9±0.3 |
| La | 2.6 | 4.2±0.4 | 3.95 | 4.0±0.3 | 21 | 18.5±0.9 |
| Lu | 0.06 | 0.040±0.002 | 0.16 | 0.132±0.007 | 0.68 | 0.67±0.04 |
| Rb | 264 | 265±14 | 4 | — | 257 | 305±30 |
| Sc | 0.22 | 0.211±0.002 | 35 | 36.1±0.3 | 5.2 | 5.62±0.09 |
| Sm | 0.38 | 0.74±0.04 | 1.5 | 1.7±0.1 | 6.2 | 6.5±0.3 |
| Ta | 0.4 | 0.133±0.006 | 0.17 | — | 1.9 | 1.92±0.05 |
| Th | 1.3 | 1.14±0.02 | 0.53 | 0.56±0.08 | 26.5 | 32.2±0.7 |
| Zn | 3.2 | 1.8±0.2 | 111 | 91±13 | 30 | 30±3 |

JF-1 Feldspar. O-hira feldspar (Mixture of orthoclase and albite) Nagiso-machi, Nagano Prefecture. split 8, position 38.

JGb-1 Gabbro. Utsushigatake (Augite-hypersthene hornblende gabbro) 86 Ma, Funehiki-machi, Fukushima Prefecture. split 2, position 66.

JR-1 Rhyolite. Wada Toge obsidian, 0.8 Ma, north of Wada Toge, Wada-mura, Nagano Prefecture. split 6, position 45.

* A. ANDO *et al.*, *Geostand. Newslett.*, **11**, 159 (1987).

But lacking in adequate standard for Ca in irradiated samples, the contents of Ca in these rocks could not be determined. Because of the very low contents of other elements in these rocks, preliminary results which are composed of twelve elements are reported. Each value for Cs and Sr in the table is not yet subtracted by the contribution of the thermal neutron fission products of uranium in samples. The calculation for other elements is now in progress; the results will be published elsewhere.

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Table 6. Results of neutron activation analyses of standard rock samples JLk-1, JLs-1 and JDo-1.

| Element | JLk-1 | JLs-1 | JDo-1 |
|---|---|---|---|
| *Major elements* (%) | | | |
| Fe | 4.92 ±0.06 | 0.0114±0.0002 | 0.0173±0.0002 |
| Na | 0.82 ±0.02 | — | — |
| K | 2.42 ±0.05 | — | — |
| Mn | 0.195±0.004 | — | 0.0046±0.0003 |
| *Trace elements* (ppm) | | | |
| Ce | 79 ±3 | 1.55 ±0.03 | 4.98 ±0.07 |
| Co | 19.1 ±0.5 | 0.0703±0.0007 | 0.166 ±0.001 |
| Cs | 11.8 ±0.3 | 0.0253±0.0006 | — |
| Eu | 1.28 ±0.03 | 0.0072±0.0001 | 0.131 ±0.001 |
| Ga | 18 ±1 | — | — |
| Hf | 4.1 ±0.2 | — | — |
| La | 41.7 ±0.8 | — | — |
| Lu | 0.52 ±0.05 | — | — |
| Sc | 15.7 ±0.1 | 0.0292±0.0002 | 0.149 ±0.001 |
| Sm | 7.5 ±0.3 | — | — |
| Sr | — | 188 ±9 | 85 ±4 |
| Ta | 1.16 ±0.08 | — | — |
| Tb | — | 0.0045±0.003 | 0.159 ±0.002 |
| Th | 18.0 ±0.5 | 0.043 ±0.006 | 0.104 ±0.003 |
| Yb | — | 0.021 ±0.003 | 0.326 ±0.008 |
| Zn | 105 ±18 | 2.02 ±0.03 | 23.5 ±0.3 |
| Zr | — | 12.0 ±0.4 | 11.0 ±0.5 |

JLk-1 Lake sediment Biwa-ko, Shiga Prefecture. split 3, position 17.
JLs-1 Limestone, Garou, Kamiiso-machi, Hokkaido. split 4, position 6.
JDo-1 Dolomite, Kuzuu-machi, Tochigi Prefecture. split 7, position 19.

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## References

ANDO, A., N. MITA, and S. TERASHIMA, 1987. 1986 values for fifteen GSJ rock reference samples, "Igneous rock series". *Geostandards Newslett.*, **11**: 159–166.

ANDO, A., S. TERASHIMA, T. OKAI, Y. IUCHI and T. IGARASHI, 1987. GSJ rock reference samples, "Sedimentary rock series", JLk-1, JLs-1 and JDo-1. The 1987 annual meeting of the Geochemical Society of Japan, 2D03 (in Japanese).

BROWNE, E. and R. B. FIRESTONE, and V. S. SHIRLEY, 1986. Table of radioactive isotopes. John Wiley & Sons, Inc., New York.

FLANAGAN, F. J., 1973. 1972 values for international geochemical reference samples. *Geochim. Cosmochim. Acta*, **37**: 1189–1200.

HOLDEN, N. E., R. L. MARTIN, and I. L. BARNES, 1984. Isotopic composition of the elements 1983. *Pure Appl. Chem.*, **56**: 675–694.

LEDERER, C. M. and V. S. SHIRLEY, 1978. Table of isotopes, 7th ed., John Wiley & Sons, Inc., New York.

WAKABAYASHI, F., 1987. Determination of major and trace elements in nine Japanese geochemical standard rock samples by instrumental neutron activation analysis. *Bull. Natn. Sci. Mus., Tokyo, Ser. E*, **10**: 13–19.

## Appendix

Redetermination of major and trace elements in JA-2, JB-2, JG-2 and JP-1 was

Table 7. Results of neutron activation analyses of standard rock samples JA-2 and JB-2.

| Element | JA-2 | | JB-2 | |
|---|---|---|---|---|
| | '86 value* | This work | '86 value* | This work |
| *Major elements* (%) | | | | |
| Fe | 4.30 | 4.35 ±0.07 | 10.03 | 11.6 ±0.1 |
| Na | 2.29 | 2.35 ±0.02 | 1.51 | 1.550±0.009 |
| K | 1.48 | 1.45 ±0.02 | 0.36 | 0.371±0.006 |
| Mn | 0.085 | 0.088±0.003 | 0.15 | 0.175±0.004 |
| *Trace elements* (ppm) | | | | |
| Ce | 29 | 32 ±2 | 6.5 | 9 ±1 |
| Co | 30 | 30.0 ±0.5 | 39.8 | 38.1 ±0.6 |
| Cs | 4.2 | 5.1 ±0.2 | 0.90 | 1.1 ±0.2 |
| Eu | 0.91 | 0.97 ±0.02 | 0.85 | 0.95 ±0.04 |
| Ga | 16.4 | 14.7 ±0.6 | 17.0 | 16 ±1 |
| Hf | 2.8 | 2.6 ±0.1 | 1.4 | 1.25 ±0.10 |
| La | 16 | 16.5 ±0.5 | 2.4 | 2.5 ±0.4 |
| Lu | 0.27 | 0.21 ±0.03 | 0.40 | 0.37 ±0.03 |
| Rb | 68 | 111 ±6 | 6.2 | — |
| Sc | 19 | 18.2 ±0.2 | 54 | 54.5 ±0.5 |
| Sm | 3.1 | 3.27 ±0.07 | 2.3 | 2.3 ±0.2 |
| Ta | 0.61 | 0.88 ±0.05 | 0.2 | — |
| Th | 4.7 | 5.6 ±0.3 | 0.33 | 0.870±0.006 |
| Zn | 62.7 | 60 ±6 | 110 | 123 ±9 |

JA-2 Andesite. Goshikidai sanukitoid (Olivine andesite) 13 Ma, Sakaide, Kagawa Prefecture. split 10, position 14.

JB-2 Basalt. O-shima volcano (Tholeiitic basalt, Augite-bronzite basalt) erupted in 1905–1951, northern rim of Mihara crater, O-shima, Tokyo. split 6, position 45.

* A. ANDO *et al.*, *Geostand. Newslett.*, 11, 159 (1987).

Table 8. Results of neutron activation analyses of standard rock samples JG-2 and JP-1.

| Element | JG-2 | | JP-1 | |
|---|---|---|---|---|
| | '86 value* | This work | '86 value* | This work |
| *Major elements* (%) | | | | |
| Fe | 0.64 | 0.74 ±0.02 | 5.83 | 5.69 ±0.09 |
| Na | 2.63 | 2.59 ±0.03 | 0.016 | — |
| K | 3.95 | 3.78 ±0.05 | 0.003 | 0.004±0.002 |
| Mn | 0.012 | 0.020±0.003 | 0.093 | 0.095±0.002 |
| *Trace elements* (ppm) | | | | |
| Ce | 46 | 52 ±1 | 13 | 11.6 ±0.9 |
| Co | 4.5 | 4.3 ±0.1 | 116 | 121 ±1 |
| Cs | 7.5 | 7.6 ±0.3 | <0.1 | 0.24 ±0.02 |
| Eu | — | 0.063±0.004 | — | 0.036±0.005 |
| Ga | 19 | 19 ±1 | 0.5 | 0.59 ±0.08 |
| Hf | 1.8 | 8.7 ±0.4 | 0.2 | 0.26 ±0.04 |
| La | 18 | 19.0 ±0.5 | 3.6 | 6.1 ±0.1 |
| Lu | — | 1.20 ±0.06 | — | 0.031±0.002 |
| Rb | 297 | 329 ±20 | <1 | — |
| Sc | 2.0 | 2.62 ±0.03 | 7.7 | 7.01 ±0.06 |
| Sm | 7.1 | 8.6 ±0.2 | — | 0.035±0.004 |
| Ta | 1.9 | 2.96 ±0.07 | <1 | — |
| Th | 29.7 | 29.8 ±0.6 | 0.18 | 0.29 ±0.03 |
| Zn | 12.7 | — | 29.5 | 36 ±4 |

JG-2 Granite. Naegi granite (Biotite granite) Cretaceous, Hirukawa-mura, Gifu Prefecture. split 5, position 7.

JP-1 Peridotite. Horoman peridotite (Dunite) Horoman, Hokkaido. split 2, position 65.

* A. ANDO *et al.*, *Geostand. Newslett.*, **11**, 159 (1987).

carrird out, becauses some elements' data of these rock samples have been lacked in the previous report. Experimental procedures were almost the same as those described in this report except that irradiated weight of JP-1 was about 35 mg. The results were averaged with previous data. The revised results are tabulated in Tables 7 and 8.