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On Performance Experience and Measurements with Ningyo Waste Assay System (NWAS) -Ⅲ

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A uranium mass assay system, NWAS (Ningyo Waste Assay System), for 200-litter wastes drums applied by NDA method was developed and accumulated the data of the actual uranium bearing wastes drums. The system consists of the 16 pieces of Helium-3 proportional counters for neutron detection generated from U-234(α,n) reaction or U-238 spontaneous fissions with polyethylene moderation and a Germanium solid state detector (Ge-SSD) for gamma ray detection as to determine uranium enrichment. In previous report, some measurement experiences had been introduced briefly. After that the measurements campaigns against the actual wastes drums stored in URCP had been carried out successfully, the uranium determination data of 850 drums had been accumulated approximately. Those characteristics were rich in variety including various kinds of matrices, uranium chemical compositions and range of uranium mass and so on. These works have contributed the decrease of the MUF in URCP, for which was the first purpose of introduction of NWAS. On the other hand several considerable problems on the system or methodology had been revealed technically or analytically through the measurements experiences. Such experiences are to be described precisely, in addition newly gained knowledge will be marshaled. Furthermore as the next improvement plans, the active neutrons assay for uranium bearing wastes drums are now progressing. The results of complications will lead us to the progressive next steps.

Keywords : Uranium Mass Assay, Neutron and Gamma Measurement, NDA, Helium-3 Proportional Counters, U-234(α,n)F Reaction, Spontaneous Fission of U-238, Germanium Solid-State Detector(Ge-SSD), 200-litter Actual Uranium Bearing Wastes Drums, Add-A-Source Method

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＊1 Ningyo-toge Nuclear Industries Co., Ltd.

NWAS の導入と測定経験について-Ⅲ

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200 リットルドラム缶収納の廃棄物中全ウランを定量する非破壊分析（NDA）装置 NWAS を開発、基礎的な試験を経て実ウラン廃棄物の測定に適用した。測定原理は廃棄物中ウランα線と共存するフッ素等との U-234(α,n)反応で生じる中性子と U-238 自発核分裂中性子を、ポリエチレン減速材により熱中性子化し、16 本の He-3 比例計数管を用い測定、また、Ge 半導体検出器によりウラン濃縮度を測定するというものである。前報ではその成果の一端を紹介したが、その後人形峠環境技術センター製錬転換施設に保管されている実ウラン廃棄物の測定作業が順調に進行し、約 850 体の測定を実施した。その間多種多様なマトリックス、ウラン線源物質、広範囲のウラン質量を含有する廃棄物に対する測定経験を積み重ね、適用範囲を拡大させてきた。また、当初の導入目的である製錬転換施設における MUF 低減に貢献を果たした。その反面、測定技術・解析手法における種々の問題点も明らかになった。それらの経験を詳細に報告するとともに、新たに得られた知見を整理した。さらにパッシブ測定方式をアクティブ測定方式へと転換する高度化計画を推進し、既に装置構築を完了した。これまでの経験の意義と課題を集約し次のステップへの糧とする。

＊1　人形峠原子力産業（株）

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1. Introduction

In couple of previous reports the developments process around the Ningyo Waste Assay System (NWAS) at the Ningyo-Toge Environmental Engineering Center of JAEA were described in detail. [Ref.-1] [Ref.-2] For three years our vigorous works had been carried out in order to validate the potency, effectiveness and applicability. Numerous tests were strenuously carried out to identify the characteristics of NWAS. And after confirming the parameters that had been necessary to determine uranium mass, the measurement campaign for actual wastes drums stored in Uranium Refining and Conversion Plant (URCP) had been carried out successfully. During two years the uranium mass of the actual wastes drums up to 850 numerically had been determined and the total uranium mass had been reached up to 23 tons in metal equivalent uranium.

In addition the various kind of wastes drums matrices had been applicable and the chemical composition of uranium had not limited uranium fluorides but adaptable to uranium oxides. At the same time several inherent problems caused by the passive assay had been revealed.

In this report the far deeper considerations and discussions compared with previous two reports have been added. The experiences of the measurements actual wastes drums have given us the confidence and assurance of the neutron assay for uranium. Based on this captured achievement, the further developments of the neutron assay system will be to be purchased which overcome the problems noted above. The answer for the next challenge is the “active neutron assay” as an alternative to “passive neutron assay”.

2. The Significance of Neutron Assay

Recently several reports have been issued which suggested the possibilities of the methodologies of the assay system for uranium bearing wastes. Those methods are roughly characterized gamma assay and neutron assay. The gamma assay is limited to the passive assay in principle, whose methodology is used gamma rays emitted from uranium and its progenies. On the other hand the neutron assay is rich in variation, so as to the passive neutron assay whose methodology is applied neutrons generated by (α,n) neutron reaction or spontaneous fission neutron reaction, and the active neutron assay whose methodology is applied by outer neutron bombardment basically.

In the beginning we had tried the coupling methodologies which were applied simultaneous measurement both by passive neutron and passive gamma ray. Those concepts were presumably intended to take a meritorious point among both, however the poor energy resolution and high background counting of 5 inch NaI(Tl) scintillation detector had prevented from advancing passive gamma assay. Afterwards we had exchanged the gamma ray detector from NaI(Tl) scintillation detector to Ge-SSD with

the primary objective of uranium enrichment determination only.

As we discuss after this, the final conclusion on uranium assay had nothing left except neutron assay from the view of good detector response and simple calibration.

Furthermore we have decided that the methodologies of neutron measurement are to be changed from passive neutron assay to active neutron assay based on good detector response as mentioned later.

3. System Configurations and Measurements Methodology

3.1 Neutron Measurement

As was reported in [Ref.-1] [Ref.-2], the basic conceptual design and of NWAS system had not changed shown in **Figure-1.** In this report the explanations about the structure of the slab box embedded the neutron detectors and polyethylene moderator are to be proved good detector response, so it will be useful information to design neutron detection.

The neutrons emitted from a particular wastes drum are the spontaneous fission neutron from U-238 and generated by the (α,n) reaction between particles from U-234 and low-Z elements especially fluorine atoms with particularly large cross section.

The heart of NWAS consists of two large sized polyethylene-moderated Helium-3 proportional counters in aluminum detector slab boxes. Each Helium-3 detector slab box contains eight of 4atm, 25.4mm diameter and 914mm active length of Helium-3 proportional counters on a 3.8cm pitch, supplied by GE Reuter Stocks (model number : RS-P4-0836-201). Each box has two pre-amplifiers which are attached to four Helium-3 proportional counters. [Ref.-3]

The Helium-3 detector slab boxes contain 12 cm depth of polyethylene moderator and a 2.54cm thick of borated polyethylene (5% of boron by weight) back shield to reduce background neutron detection. **Figure-2** shows the outer structure and the cross section structure, these dimensions had been determined by MCNP calculation which simulated approximately 2MeV neutrons behavior emitted from (α,n) emission and spontaneous fission in the wastes matrix materials down to thermalized. [Ref.-4]

The eight of Helium-3 proportional counters are placed 2.54cm from front surface (nearest drum) of the polyethylene arranged in a straight rows respectively. Those Helium-3 detector slab boxes are set up on 90 degree clockwise and are to be faced with 200-litter wastes drums. Respect to above configurations the bird view are shown in **Figure-3.**

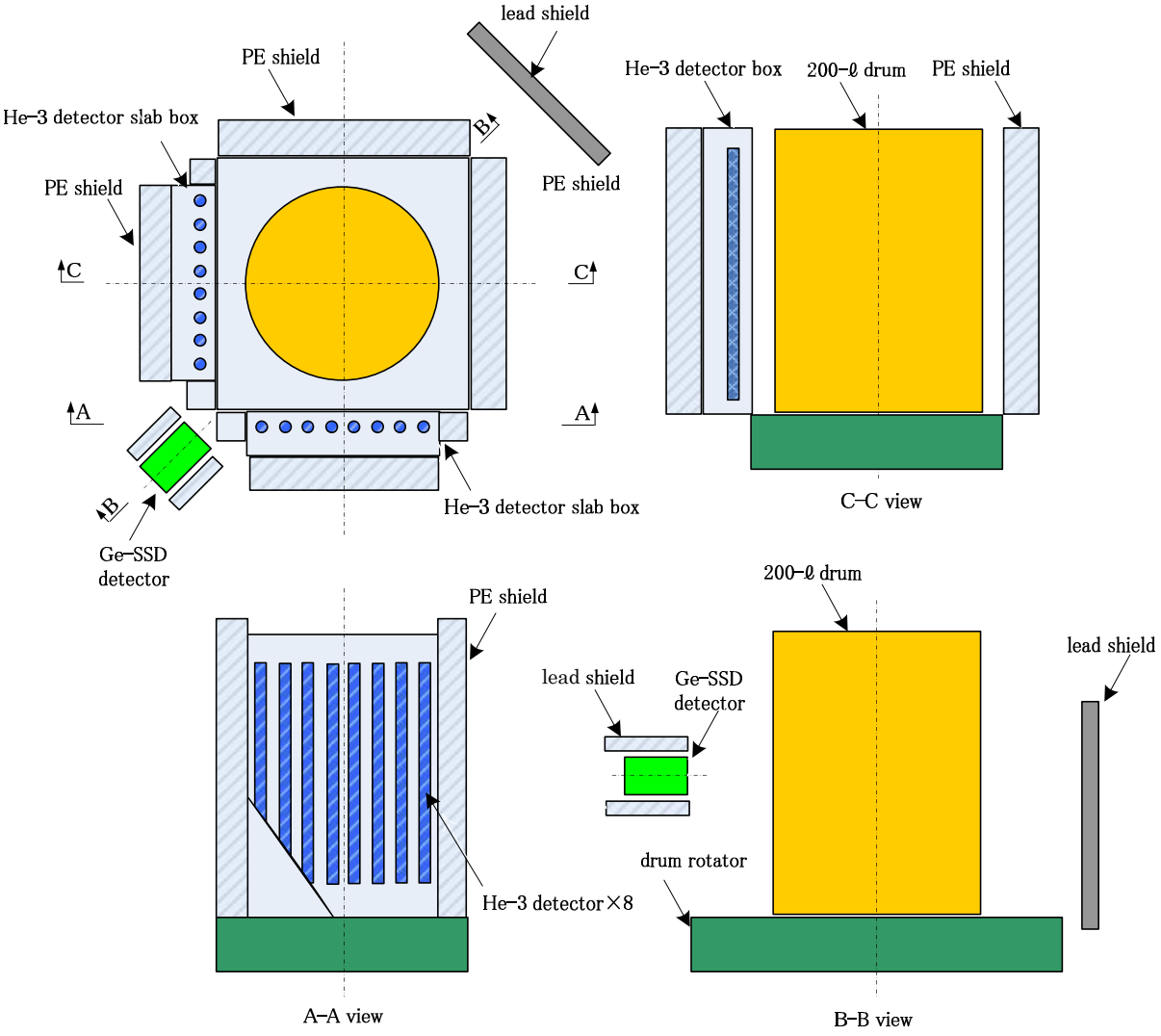


Figure-1　Conceptual Design of NWAS System

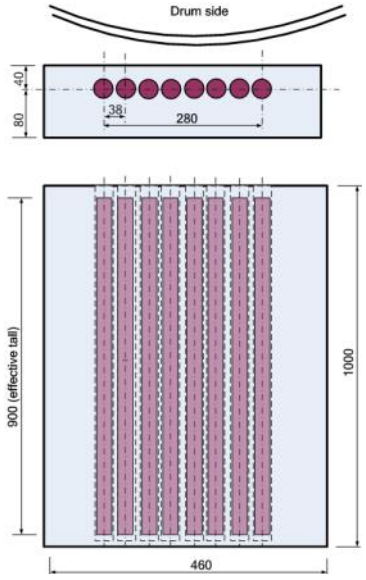


Figure-2　Polyethylene Slab Structure

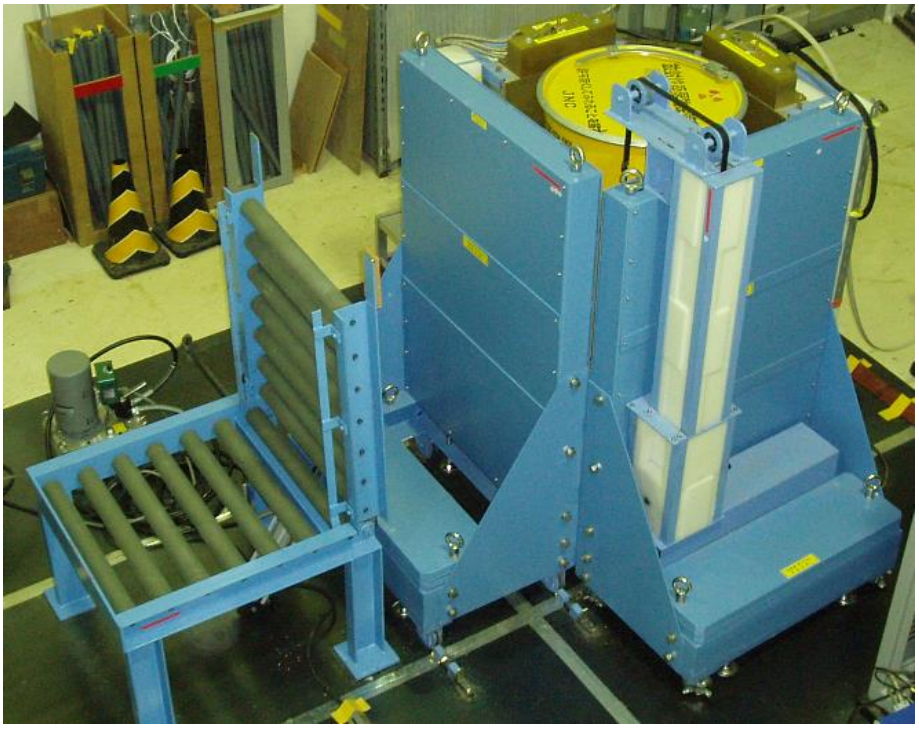


Figure-3　Bird View of NWAS System

The amplifier and control system (Shift Register JSR-12/12N) shown in **Figure-4**, were supplied by Canberra Corporations. Additional shield slabs composed of 100mm thickness of polyethylene for four sides in order to decrease ambient neutron background. One of the four slabs are movable for drum loading and un-loading. The 0.01mm of cadmium foil was wrapped outside of aluminum slab boxes including Helium-3 proportional counters as an additional neutron absorber, in order to decrease ambient neutron background.

3.2　Gamma Spectroscopy

A 77 mm diameter of high purity Ge-SSD with 40mm thickness of lead shield is embedded between two neutron detectors slab boxes at the central level of 200-litter wastes drum. The gamma ray detection aims for determining uranium enrichments by measurement both 186keV/1001keV energy peaks. The detector, its amplifier, and the control system are supplied by Canberra Corporations, shown in **Figure-5.** The energy resolutions of the Ge-SSD as an index of FWHM was limited within 2.1keV (at 1333keV), therefore the satisfactory analysis had been achieved unlike in the case of NaI(Tl) detector.

Figure-4　Control System

Figure-5　Gamma Spectroscopy

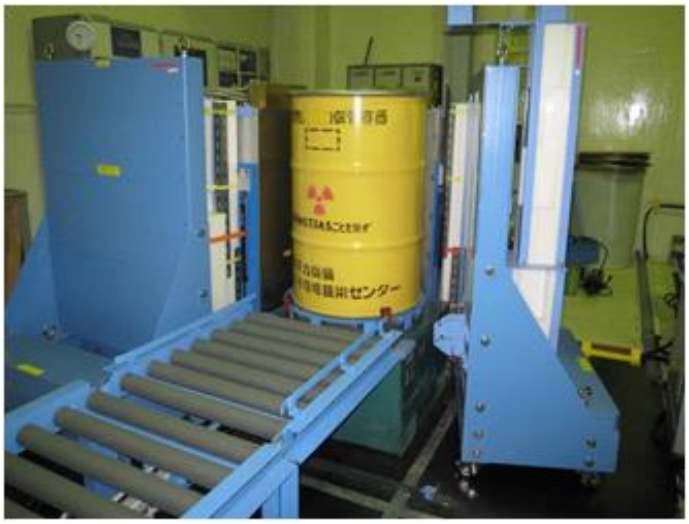


Figure-6　Drum Rotation and Transport System

3.3 Utilities

(1) Drum transport system

In principle, the measurements are carried out rotating the drum at 2.5 rpm of speed. The drum rotation system withstands up to 450 kg of drum weight. Originally it was not so easy and not so convenient that we moved to the actual drums on the drum rotator by using drum handling porter. For the purpose of easy and safe drum handling, a drum transport system connecting with the drum rotator with rubber shown in **Figure-6,** had been introduced.

The drum transport system contains horizontal drum transport rollers and flip-up style transport rollers that moved by hydraulically-powered driving, both are connected directly and flatly when loading a drum. The system enabled us to move drums easily and conveniently.

(2) Personal computer

NEC personal computer (Versa Pro installed Microsoft Windows XP) is currently installed for the control the shift register and data acquisition.

(3) Software

Supreme “INCC” for neutron data acquisition program is used, which was provided by LANL [Ref-5]. It has been fulfilled not only to acquire counting but also to exclude the sudden burst pulses, which are mainly derived from cosmic rays.

INCC’s characteristics are that the procedures which enable to reduce the relative counting errors by comparing series cycle counting rates in shortly. Implemental explanation says the following up stochastic check (so called QC check) enable to exclude some counting values as was over standard deviation. The main policy of QC check is described in the followings.

Measurements normally consist of repeat cycles, for our example, 300 cycles of 12 seconds each for low uranium mass drums. The accidentals/singles test compares the singles rate with the accidental coincidence rate at the end of each cycle. If the neutron source rate is constant during the cycle, then, within statistical errors, the accidentals rate should equal the square of the singles rate times the gate length. If the rates do not agree within statistical errors and the quality control tests are turned on, then the cycle is rejected and another is made automatically. The limit of acceptance is set by the parameter "Accidentals/singles test outlier limit (sigma)", which is the limit expressed in standard deviations. The usual limit is 3 standard deviations.

Convenient and commercially provided “Gennie 2000” for gamma ray acquisition program is used. “Gennie 2000” has the multi functions which include of analyzing gamma rays spectroscopy and evaluate net peak counts and its errors.

4. Measuring and Analysis Approach

4.1 Background Measurements

Since NWAS aim at the well-established passive measurements of very low level of thermal neutron and gamma ray emanating from uranium bearing wastes drums, it is important to determine the backgrounds, how to account then, and to calculate appropriately.

The analysis of the neutron background rate is important in the point of view of "signal to background ratio" for NWAS. Furthermore the measured background rate, although fairly stable throughout the measurement periods, is still affected by the presence of the wastes drum being measured.

Long time background measurements (usually 72,000 seconds) were carried out periodically so that background rate variance was identified and confirmed. Until now, by aggressive efforts the average of neutron background was improved down to 4.2 cps compared with the average that was previously reported. However during the test of NWAS, considerable difficulty was found in assessing the correct (and appropriate) background count rate, which changed depending on the presence of matrices. (In this report various substances packing in drum are defined to be called "matrices")

When drums with metals (mainly steel) were placed on the rotation platform, the effective background rate appeared to increase (by about 10% on average) in the neutron detector. This was due to the increase in cosmic ray spallation neutrons created by the presence of the high-Z materials in the drum. Conversely, when drums with combustibles or empty were placed on the rotation platform, the effective background rate appeared to decrease (by about 10% on average) in the background neutrons by the moderating low-Z matrix materials. Therefore the prior series of the test by using the different matrix materials, the background rate was certainly measured correspondingly so that a correction will be applied to the measured background and net neutron count rate that is based on the matrix material (low-Z or high-Z materials) within the drum itself.

However, the above problem is not so important. Because the background level often vary more than a little, and they are considered by the influence of humidity in the detector Helium-3 detector slab boxes. In order to address the humidity problem, the maintenance procedure was improved, including frequent exchanging the silica gels are needed based on the recommendation by LANL [Ref.-7].

For practical purposes in actual use, we came to conclusion that the just previous value would be adopted the measured value of every matrix materials as the background. The typical background data obtained by the neutron detector is shown in **Table-1.** There is no great distinction among them, but steel drums show slightly higher backgrounds than others.

Table-1 Typical Background Data of Neutron Detector (example)

Kind of Matrix materials	time (sec)	counts (cps)	error (cps)	relative error
None	72,000	4.391	0.008	0.0018
Empty	72,000	4.444	0.008	0.0018
NaF pellets	72,000	4.104	0.008	0.0019
alumina pellets	72,000	4.283	0.152	0.0355
steel pieces(0.5g/cc)	72,000	4.561	0.008	0.0018
steel pieces(0.9g/cc)	72,000	4.570	0.010	0.0022

4.2 Detector Calibration

For neutron detectors, 117,000 neutrons/sec traceable source intensity of Cf-252 checking source (approximately 1MBq) is used for calibration, and the counting efficiency is checked periodically. The checking source is settled at the center position of empty drum. A Cf-252 source holder is installed in NWAS, so that it can be placed in the center hole of the drum rotation platform. This source holder and Cf-252 source were mounted on the drum rotation system.

The measured counting efficiency is 5.434 % in average. This condition remains unchanged after our previous report. The latest precise data is shown in **Table-2**. The errors are derived from the counting uncertainties and the source intensity definition error. (Approximate 5% uncertainty according to the certificate of the source)

Table-2 Calibrated Neutron Counting Efficiency (example)

	Date	Elapsed Time (days)	Average count rate (cps)	Counting efficiency (%)
1	2011/8/30	561	4.184E+3	5.4
2	2011/9/8	570	4.182E+3	5.4
3	2011/10/6	598	4.166E+3	5.4
4	2011/11/1	624	4.128E+3	5.5
5	2011/12/5	658	4.047E+3	5.5
average				5.4

[note] Cf-252 source was certificated on 15/Feb/2010.

4.3 Neutron Response

The neutron measurements were carried out by “rate only” mode of INCC, which are capable to acquire neutron counts from 16 Helium-3 proportional counters by time

division method, normally used in 12 seconds.

Predictable outcomes suggest that there arise the results that a neutron response is difference from the kind of uranium sources and/or matrices.

The relationships of the declared uranium mass versus neutron count rate per second (=cps) was obtained. It expresses the features of the neutron emission rate based on uranium source items such as chemical composition or uranium enrichment, also expresses the characteristics of each matrix materials reflected neutron penetration. Those neutron response performances are summarized and shown in **Figure-7.**

The data showed a good linearity with small counting errors, so the evaluations by extrapolations are expected surely. Furthermore the neutron response data between fluorides and oxides suggested few conclusive differences.

The variances of these factors reflect the differences depending on the chemical composition of uranium and/or matrices. For one example, in case of NaF as matrix materials, the factor "F_w" values were estimated 0.169/0.123 in regard to UF_4/U_3O_8 standard source. The results suggest that the neutron emission rate of uranium fluorides are greater than uranium oxides. For another example, in case of UF_4, a standard source, the factor "Fw" values were estimated 0.169 / 0.138 / 0.218 / 0.191 / 0.082 in regard to NaF(d=1.0g/cc) / alumina(d=0.8g/cc) / steel(d=0.5g/cc) / steel(d=0.9g/cc) / CaF_2(d=1.5g/cc) as matrices respectively. The results suggest that the neutron penetration rate through the matrices was depending on chemical components and/or averaged bulk density.

It was expected that the weighing factor "F_w" of fluorides is greater than that of uranium oxides. However, there were few conclusive differences between fluorides and oxides as source. This fact means that NWAS are expected to utilize for not only uranium-fluorides measurements but also uranium-oxides measurements.

By using the weighing factor "F_w", the uranium mass was calculated according to Equation-1. Those mutual relations between the calculated uranium mass versus the declared uranium mass are summarized graphically in **Figure-8.** The data also showed a good linearity if considering 5% error of the calibrated source, so the uranium mass determinations are possible by this method.

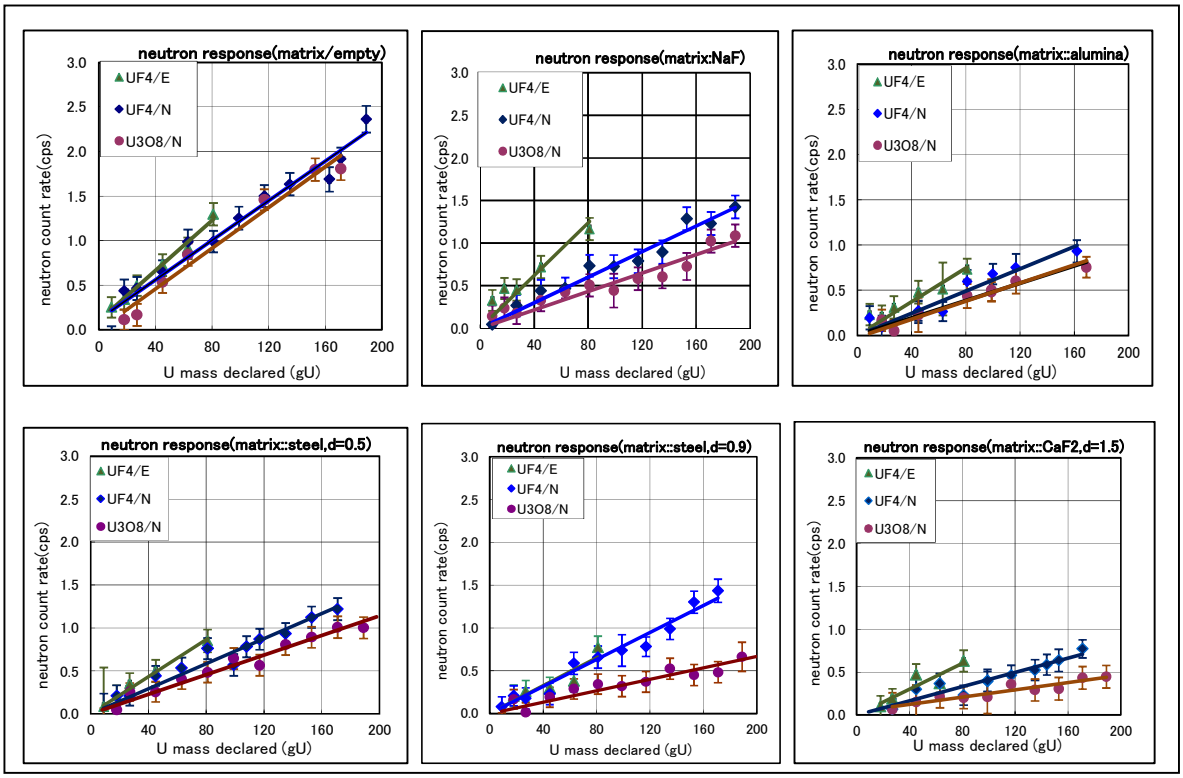


Figure-7　Neutron Response Performance

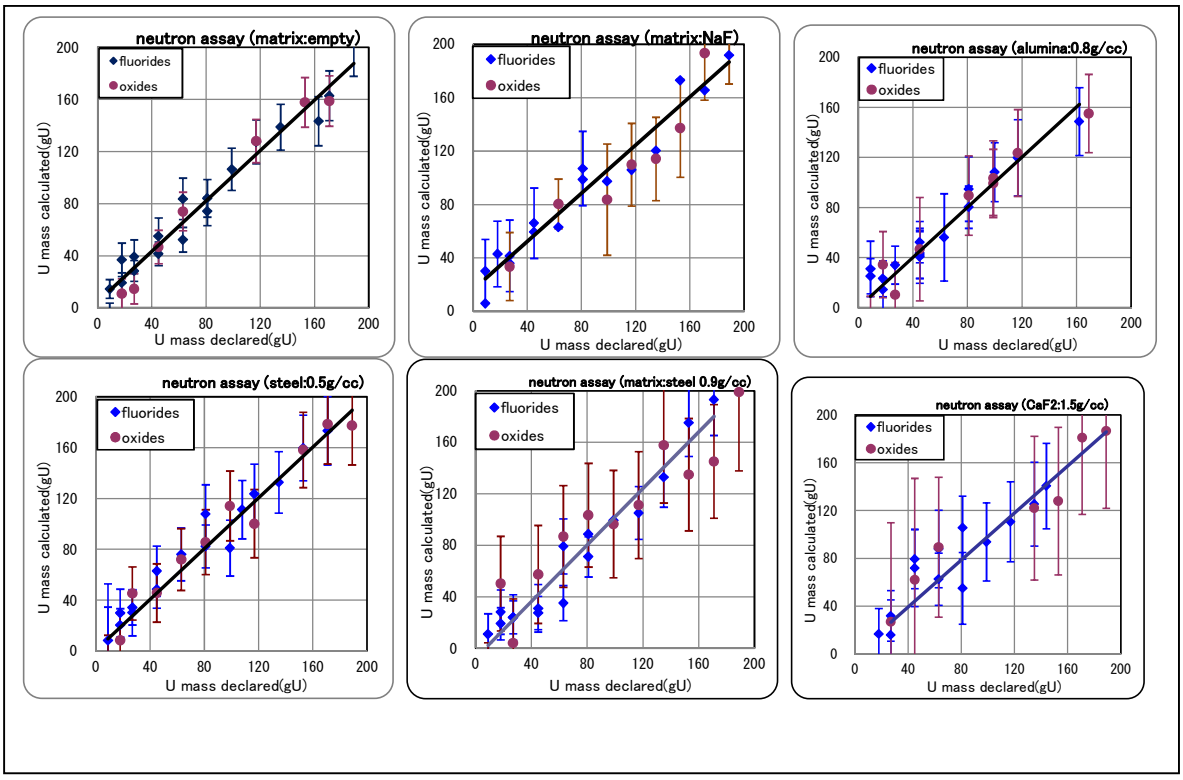


Figure-8　Relations between Calculated/Declared Uranium Mass

4.4 Analysis Methods

Uranium mass is determined by the following formula based on the thermal neutron counting, i.e. obviously uranium enrichment and weighing factors of "Fw" which are key factors in uranium mass determination. The important point is that the neutron emission rate which differ from the chemical composition of uranium and/or the enrichment of uranium. The formula for estimation of uranium mass are as follows.

$$M = \frac{n_S - n_B}{\varepsilon \cdot Y(E)} \qquad \text{---- (Equation-1)}$$

$$Y(E) = 0.0136 + F_W \cdot IE$$

where M : calculated uranium mass (gU)

n_S : measured background-subtracted single rate for sample (cps)

n_B : measured background single rate for empty condition (cps)

ε : detection efficiency (-)

Y(E) : neutron emission yield depending on uranium enrichments (n/s/gU)

IE : uranium enrichment (U-235 enrichment %)

0.0136 : neutron emission rates of spontaneous fission (n/s/gU) [Ref.-3]

Fw : weighing factor of neutron emission rates of (α,n) reaction depending on chemical composition or matrix materials (n/s/gU)

The error estimation of uranium mass is necessary to include the counting error and the certification error of Cf-252 source, evaluated 5% approximately.

$$\sigma M = M \times \left(\sqrt{\left(\frac{\sigma_{ns}}{n_s - n_b} \right)^2 + \left(\frac{\sigma n_B}{n_b} \right)^2} + 0.05 \right) \qquad \text{---- (Equation-2)}$$

In the previous report, we have brought the samples of weighing factors of "Fw" derived from (α,n) reaction and neutron penetration through the matrices. But some mistakes were found afterwards, that was in the obtained experimental re-estimation of weighing factors of "Fw", and it was followed up.

The weighing factor "Fw" is obtained by the fitting calculation between the declared uranium mass and the calculated uranium mass on each matrix materials, i.e. it is the stochastic calculation to minimize relative errors between the experimental values and the calculated values.

The estimated weighing factor "Fw" values are shown in **Table-3**.

Table-3 Overview of Weighing Factor "F_W"

chemical composition	empty	NaF (d=1.0)	alumina (d=0.8)	steel (d=0.5)	steel (d=0.9)	CaF_2 (d=1.5)
UF_4	0.288	0.169	0.138	0.218	0.191	0.082
U_3O_8	0.278	0.123	0.107	0.194	0.111	0.039

4.5 Error Evaluations and Detection Limit

Generally the detection limits of the radiation measurement system are defined from the background rates, the counting time, and the parameters regarding statistical uncertainty. For the purpose of estimation of the detection limit for this method, the 3 σ method was applied, which is popular in Japan. The analytical formula is described in Equation-3. The second formula which calculates part of obtaining uranium mass, are equal to Equation-1.

$$MDM = \frac{n_D}{\varepsilon \cdot Y(E)} \qquad \text{---- (Equation-3)}$$

$$Y(E) = 0.0136 + F_W \cdot IE$$

$$n_D = \left(\frac{k}{2}\right) \times \left[\frac{k}{t_s} + \sqrt{\left(\frac{k}{t_s}\right)^2 + 4n_B\left(\frac{1}{t_b} + \frac{1}{t_s}\right)}\right]$$

where MDM : minimum detectable mass (gU)

n_D : minimum net detectable counting rate (cps)

ε : Cf-252 counting efficiency (-)

Y(E) : neutron emission yield depended on uranium enrichments (n/s/gU)

k : multiple factor against standard deviation (k=3)

n_B : background rates (cps)

t_s : counting time for sample (cps)

t_b : counting time for background (cps)

The evaluation trials for the typical detection limit according to described above formulae had been performed, its summary corresponding to chemical form, matrix materials, counting time and uranium enrichment are shown in **Table-4.** The evaluated values are almost within 10-20gU except CaF_2 powder.

Table-4 Typical Detection Limit Data by Neutron Measurements

chemical composition	uranium enrich	Meas. time(min)	empty	NaF (1.0g/cc)	alumina (0.8g/cc)	steel (0.5g/cc)	steel (0.9g/cc)	CaF_2 (1.5g/cc)
UF_4	1.3%	20	9.4	13	19	12	14	30
UF_4	1.3%	40	6.7	9.4	13	8.7	10	21
UF_4	1.3%	60	5.5	7.7	11	7.2	8.1	18
UF_4	1.08%	20	11	16	23	15	17	36
UF_4	1.08%	40	8.0	11	16	10	12	25
UF_4	1.08%	60	6.6	9.2	13	8.5	9.7	21
UF_4	0.71%	20	17	23	33	22	24	51
UF_4	0.71%	40	12	17	23	15	17	36
UF_4	0.71%	60	9.8	14	19	13	14	29
U_3O_8	0.71%	20	9.8	16	24	14	23	74
U_3O_8	0.71%	40	6.9	11	17	9.7	16	53
U_3O_8	0.71%	60	5.7	9.1	14	8.0	13	43

4.6 Determination of Uranium Enrichment

In order to estimate the total uranium mass, the weighing factors "F_W" introduced above, and the uranium enrichments were used according to [Equation-1]. Previously, the each enrichment data was supplied from the past archival record with respect to each actual drum units.

However, such uranium enrichment data was so vague and exhaustive that the precise enrichment value could not be assured.

Therefore we applied supplementary method by gamma spectroscopy data analyzed from 186keV peak of U-235 and 1001keV peak of Pa234m, progeny of U-238 used Ge-SSD. This method provides the analysis of individual identification of the uranium enrichment data. The analytical formula is shown in below. The counting efficiency including shielding effect of each kind of matrix materials had been calculated by MCNP5 code.

$$IE = \frac{M(U\ 235)}{M(U\ 235) + M(U\ 238)} \times 100 \qquad \text{---- (Equation-4)}$$

$$M(U\ 235) = \frac{PA(186\ keV)}{t \times \varepsilon(186\ keV) \times \eta(186\ keV) \times SA(U\ 235)}$$

$$M\ (U\ 238\ ) = \frac{PA\ (1001\ \ keV\ )}{t \times \varepsilon\ (1001\ \ keV\ ) \times \eta\ (1001\ \ keV\ ) \times SA\ (U\ 238\ )}$$

where IE:uranium enrichment(%)
M:uranium mass(gU)
PA:peak count rate(cts)
t:counting time(sec)
ε :counting efficiency(-)
η :gamma ray emission branch(-)
SA:specific activity(Bq/gU)

4.7 Mal-distribution of Uranium Sources

It is widely known that some corrections are required against the source mal-distribution in NDA measurements used gamma rays. In fact, gamma rays are generally inclined to be attenuated in matrices, especially through relatively high averaged bulk density matrices.

Contrary to this fact, such problems are relatively eliminated in NDA.used neutron methodology. In our NWAS high sensitivity and long (or tall) detectors are available. As a matter of fact, NDA.used neutron methodology has the supreme merits which suggest high penetration through matrices especially high averaged bulk density like steels, therefore less needs are required for corrections against the source mal-distribution.

In order to verify the flat response toward both horizontally and vertically, the impacts of the source mal-distribution was examined by changing the deposition of the Cf-252 point source on the arbitrary position horizontally and vertically. This geometry for in source mal-distribution is shown in **Figure-9,** and the trend analysis is shown in **Figure-10**.

Toward horizontal direction, there also found a flat distribution within 25 cm range from the center of drum. Toward vertical direction, there found a flat distribution within 50 cm range from the center of drum.

These results confirmed that there were less impacts based on source mal-distribution. In fact, it was obvious that there appeared no response deviation toward vertical direction because of the tall neutron detectors.

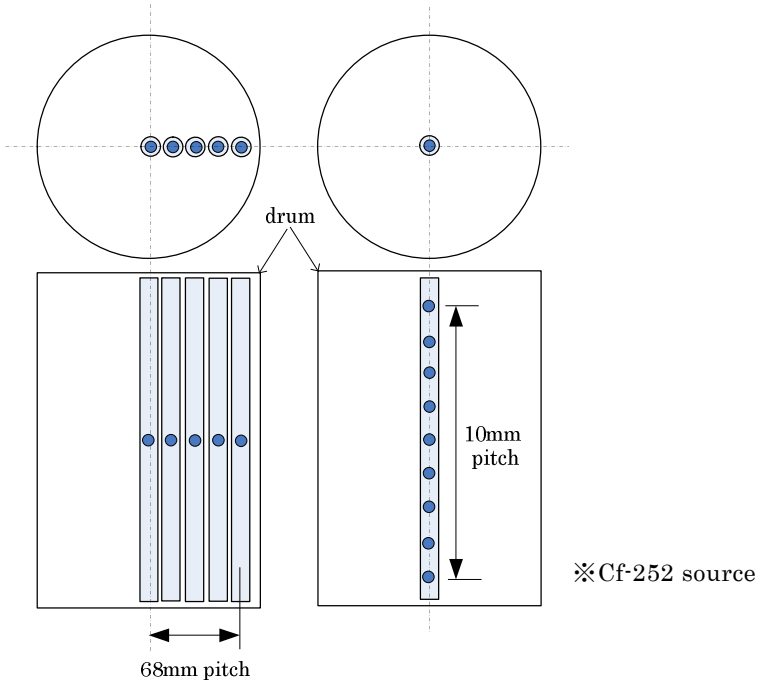
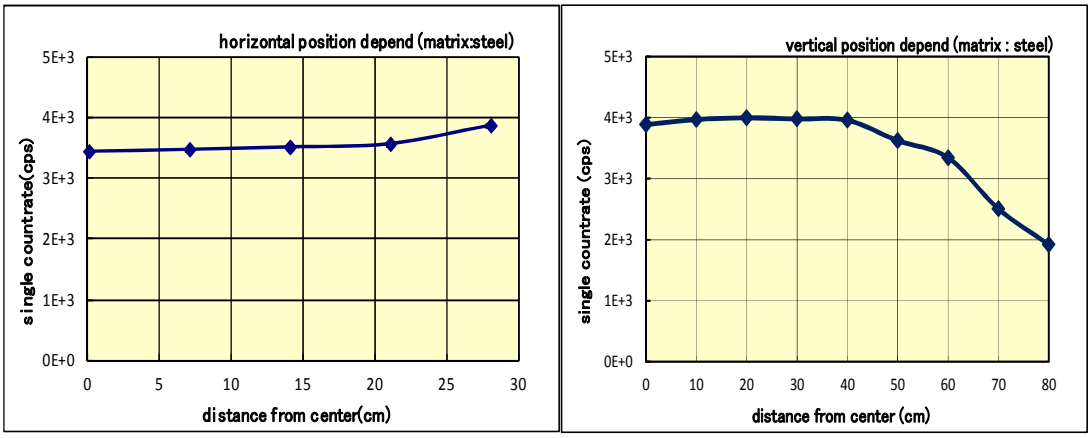


Figure-9　Mal-distribution Geometry



(a) horizontal direction　　(b) vertical direction

Figure-10　Trend Analysis for Mal-distribution

5. Summary of the Determination Uranium Mass of Actual Wastes Drums

5.1 Characteristics of the Actual Wastes Drums

As was described in previous reports the actual uranium bearing wastes drums stored in URCP are the great variety from the view of uranium chemical form, matrices in drum or uranium mass, so multifaceted approach have been required.

i) wastes drums filled with uniform matrix materials and averaged bulk density, such as sodium fluorides pellets, magnesium fluorides pellets or alumina (aluminum oxides) pellets, those had been generated as uranium conversion process wastes

ii) wastes drums filled with uniform matrix materials but non-uniform averaged bulk density, such as steel fragments and pipes generated from dismantling chemical plants

iii) wastes drums filled with almost all non-uniform matrix materials nor averaged bulk density, such as complex scrap pieces (steels, vinyl chloride material and so on) generated from dismantling

iv) wastes drums filled with indeterminate forms, such as calcium fluorides precipitates or magnesium fluorides with neutralization sediment mainly generated from waste solution treatment.

v) wastes drums filled with nuclear fuel materials itself, mainly tetra-fluoride analogue which had been left without shipping from the conversion plant whose amounts are now larger than others.

The typical examples are described below and upper views of typical actual wastes drums are shown in **Figure-11.**

And the distributions of bulk density and surface dose-rate of the uranium bearing wastes drums are separately investigated below.

i) Bulk density of the drum showed the range from 0.03 to 1.7 grams per cubic centimeter

ii) Surface dose-rate of the drum showed the range from ND to 50 μ Sv/h (ND means below 0.2 μ Sv/h)

iii) Total weight drum weight had ranged from 5kg to 350kg each in principle, normally limited up to 200kg.

iv) The nuclides compositions are characterized to separate two groups roughly, one is the uranium-235/238 and their progenies, the other is further added the uranium-232 and their progenies (thorium series progenies down to tharium-208) as reprocessed uranium.

v) Surface contamination on drum was strictly controlled below or not detected.

Scrap Uranium : UF_4 (wet and dry) as v)

Uranium absorbents : Alumina pellets as i)

Sludge : A neutralization sediment by treatment of waste solution as iv)

Metallic Fragments : metal pipes, valves, and frames dismantled as ii)

Others : VC pipes fragments as iii)

Others : FRP fragments as iii)

Figure-11 Upper Views of the Typical Actual Wastes Drums Matrix

5.2 Summary Math for Measurements of the Actual Wastes Drums

From September of 2010 to the end of 2012, over 850 measurement trials had been carried out, and numerous useful data have been obtained successfully. In advance of the neutron assay, the total weight, the surface dose-rate and neutron penetration data (applied AAS method) by Cf-252 source had been measured individually in parallel with checking those matrices via packing condition data toward each waste drum.

The summary math for measurements of the actual wastes drums are marshaled in **Table-5** in which the kind of matrix materials, number of data, region of uranium mass as gram uranium metal equivalent, its relative error (%), the accumulated uranium mass as gram uranium metal equivalent and counting time have been listed.

Among all the data the 20 groups had been listed according to order of working, which was numbered below as "tag". Moreover the 9 groups had been classified from the 20 according to the same matrix materials, whose divisions are used in **Figure-12**.

The "anal. method" means uranium assay analytical method "A", "B", "C" described below. Especially the uranium determination as a mass of equivalent metal uranium had been reached over 23 tons. It is indisputable that those measurements campaign had contributed to decrease MUF which had been initial purposes of NWAS.

The statistically-characterized summary math from the view of distribution of uranium mass by each matrix materials are verified. **Figure-12** shows the evaluated average uranium mass per waste drum sorted by matrix materials and the evaluated relative ratio of uranium mass by matrix materials. The matrix materials with highest uranium concentration per drum and major part of uranium mass evaluated was the (8)UF_4 powder which had been left as the reject product in URCP. The matrix materials with next highest was the (2)active alumina pellets as fluidization media which was the most important chemical process wastes in URCP. Contrary that the matrix materials with least uranium concentration per drum was (7)scrap analogue in which assumed little contaminated by uranium.

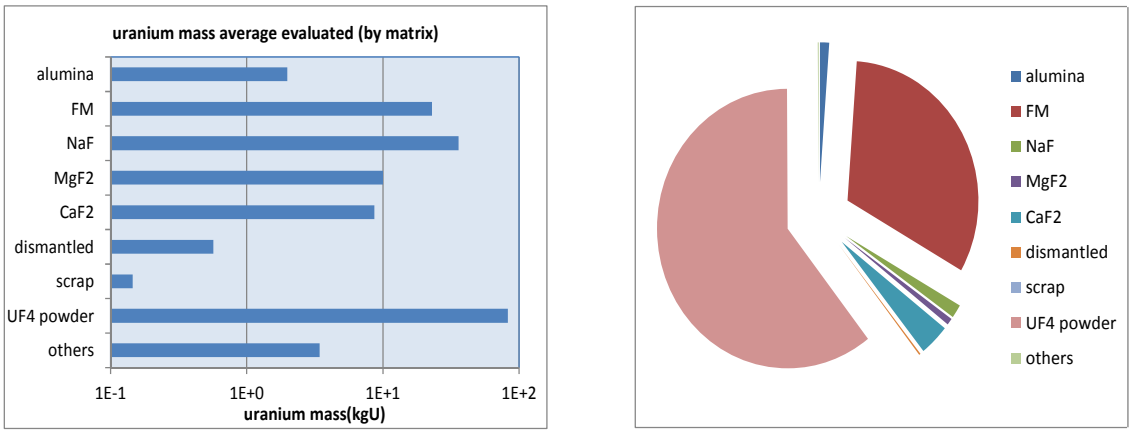


Figure-12 Evaluated Uranium Mass Sorted by Matrices

Table-5 Summary Math for Measurements of the Actual Wastes Drum by NWAS

No	tag	kind of matrix materials	data No.	region of U mass (gU)	Rel. err(%)	anal meth	mass (kgU)	time (min)
1	$1	alumina-1(NU)	22	600-3400	6.3-9.2	A	3.6E+1	60min
2	$10	alumina-2(RU)	80	243-14000	5.7-12	A	2.0E+2	60min
3	$15	alumina-3(RU)	30	51-5800	5.9-34	C	2.6E+1	60min
4	$16	alumina-FM-4(NU/RU)	11	176-47200	5.6-15	C	1.9E+2	60min
5	$2	Alumina-FM-1(RU)	71	5000-48000	5.6-6.5	A	2.1E+3	60min
6	$12	Alumina-FM-2(RU)	14	15000-33000	5.6-5.7	A	3.6E+2	60min
7	$14	Alumina-FM-3(NU)	228	1800-58000	5.6-7.5	C	4.8E+3	30min
8	$3	NaF pellets	10	11000-63000	5.7-6.1	A	3.6E+2	60min
9	$6	MgF_2 precipitates	18	770-34000	5.9-11	C	1.7E+2	60min
10	$5	CaF_2 precipitates-1	21	48-31000	6.0-110	C	1.5E+2	60min
11	$7	CaF_2 precipitates-2	6	<50	---	C	---	60min
12	$11	CaF_2 precipitates-3	56	<50-35000	5.7-90	C	5.0E+2	60min
13	$13	CaF_2 precipitates-4	15	6800-15000	6.3-7.0	C	1.7E+2	30min
14	$4	Metals-1(dismantled)	10	130-2700	7.0-14	A	1.3E+1	60min
15	$9	Metals-2(dismantled)	23	<10-1560	8.3-53	B	9.3E+0	60min
16	$18	Metals-3(dismantled)	40	70-2400	6.6-32	B	1.9E+1	60min
17	$8	Scrap materials	20	<12-770	9.2-120	C	2.9E+0	60min
18	$17	Scrap UF_4	169	3200-156000	5.7-8.0	C	1.4E+4	10min
19	$19	others(alumina+α)	4	860-8100	6.5-9.1	C	1.3E+1	60min
20	$20	others(mixture)	3	2900-14000	5.9-6.7	C	2.3E+1	30min
		Total	851				2.3E+4	

[note] i) Relative errors are include 5% of Cf-252 source uncertainty
"Anal.meth" is defined as A) used Fw value method, B) used variable Fw value method, C) used Added-A-Source method

ii) FM means "Fluidization Media" which had been generated from the uranium fluidization process as was the most important one in URCP. The refined uranium tetra-fluorides powder had been suspended and flew with porous alumina pellets, finally uranium with high vapor pressure had been vaporized and thorium progenies with low vapor pressure generated from U-232 had been absorbed with alumina pellets.

iii) "Tag" means the identified number for each group used below

The spread data sheets consolidated whole parameters used for the determination uranium mass are in **Appendix-A** (as $11, $12, $13, $14, $15, $17, $18).

We adopted the following analytical methods to determine uranium mass. The analytical method for uranium determination had been evolved through accumulating the experiences of measurements.

i) As the first step, the "Fw" values which calibrated against the each matrix materials through the mockup tests had been used toward the wastes drums identified the matrix materials and its bulk density. This determination method was easily comprehensible, mainly used to the wastes drums filled with alumina pellets simply, because those exhibited uniform properties and averaged bulk density. We named this as "A" or "Fw method". As was described above, the uranium assay is implemented by neutron count rate of Helium-3 detectors, counting efficiency obtained from Cf-252 calibration, uranium enrichment and the weighing factors (defined as Fw) by Equation-1, which is the same method as mockup tests. In these cases, the weighing factors "Fw" and counting efficiency are used as the constant values as was defined. This method was available to the wastes drums filled with uniform matrix materials and averaged bulk density, such as sodium fluorides pellets or alumina (aluminum oxides) pellets.

ii) As the second step, it was required to prepare the modified method capable to determine uranium mass against different properties especially those bulk density. Because bulk density of wastes matrix materials obviously caused the differences of the neutron penetration rate in waste matrix materials, the "Fw" value described above may change to the accompaniment of it. Therefore the applied method above had been considered that the "Fw" value will be defined as the function of bulk density, especially in the case of the wastes drums contained metal fragments generated from the dismantled. The experimental calibration curve for metal fragments "Fw" value concerning fluorides and oxides against bulk density are shown in **Figure-13**. We named this method as "B" or "interpolated" and had used these data by interpolation against arbitrary averaged bulk density.

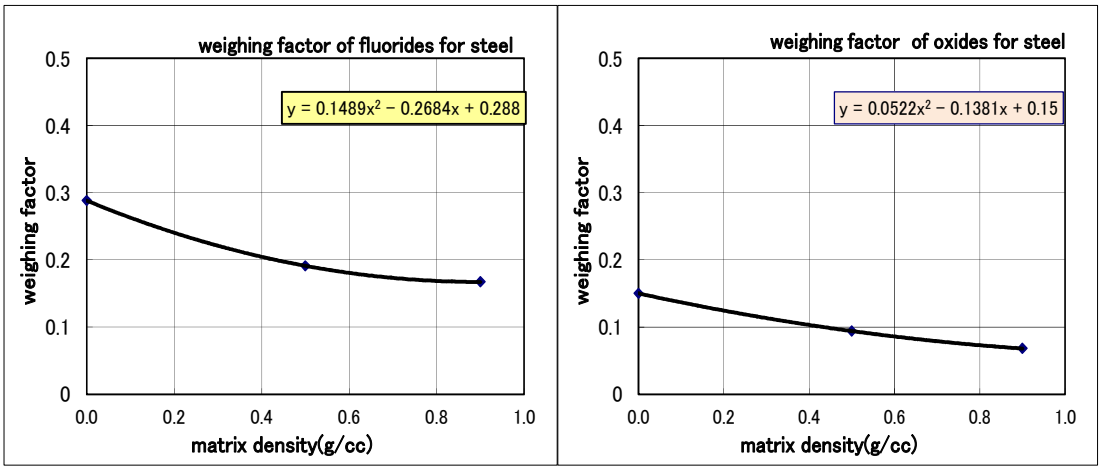


Figure-13 Fw Values Comparison among Uranium Fluoride and Uranium Oxides

iii) However they are not necessarily adaptable to all actual drums, some analytical methods corresponding to the status of actual wastes drums were tried in parallel. As the third step, Add-A-Source method was also adopted. We named this method as "C" or "AAS method". In previous report the principles around AAS method had been fully described. [Ref.-2] In this paper the brief supplemental remarks are to be appended at the front, the neutron penetration data which varied among matrices will be numerated. AAS method is commonly used for the purpose of purchasing the matrix materials information regarding perturbation easily in NDA analysis, in a word, are by measured corrected counting efficiency from penetration data one by one, as a result are capable to analyze "matrix materials unknown wastes drums". The 1MBq of Cf-252 standard sealed radioactive source is installed. The neutron penetration data had ranged from 0.13 to 0.91.among all matrices. The statistical grouping sheet of the penetration data corresponding to matrix materials are shown in **Table-6.**

Table-6 The Penetration Data Corresponding to Matrix

No.	kind of matrix materials	data	neutron penetration ratio			remarks
			min	max	average	
1	alumina pellets	132	0.37	0.63	0.48	
2	fluidization media	324	0.25	0.91	0.43	
3	NaF pellets	10	0.39	0.64	0.47	
4	MgF_2 precipitates	18	0.13	0.90	0.45	
5	CaF_2 precipitates	98	0.14	0.28	0.17	low penetration
6	Metals(=dismantled)	73	0.51	0.89	0.66	high penetration
7	scrap materials	20	0.22	0.74	0.46	
8	scrap UF_4	169	0.16	0.80	0.43	
9	others	7				no data

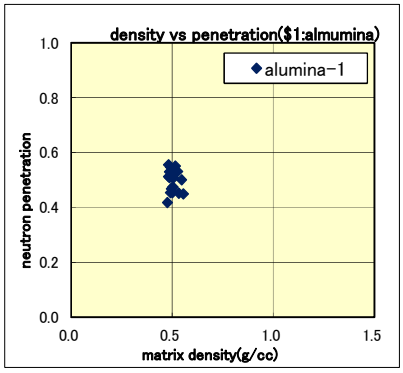
Concerning AAS method it had been important that neutron penetration rate was correlated with the matrix materials density. For all cases the direct relativity of neutron penetration data to the matrix materials density had been verified as is shown in **Figure-14.** The followings are the remarkable features :

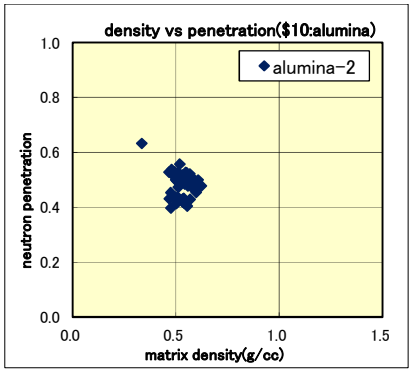
i) The plotted points are condensed in case of rigid homogeneous matrix materials like alumina pellets.

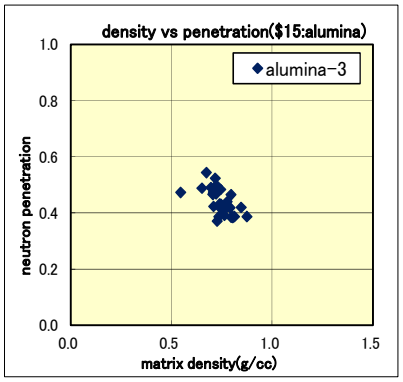
ii) The plotted points are formed hyperbolic function in case of wide distribution of density like NaF pellets, MgF_2 pellets, metals and scrap UF_4.

iii) The plotted points are formed horizontally constant in case of CaF_2 precipitates only. The reason why these peculiar effects had occurred is assumed that the moisture in the matrix materials had caused to fluctuate the penetration. (details are described in **6.3**)

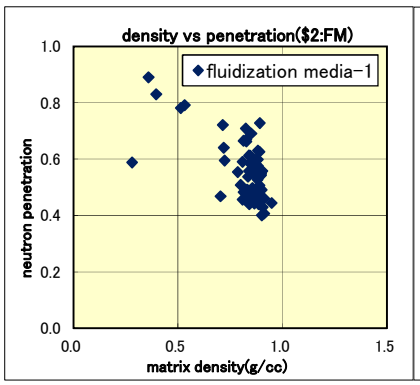
1. Alumina pellets

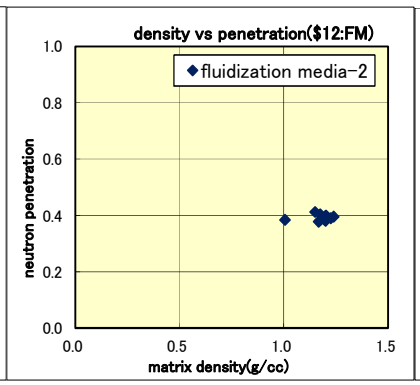


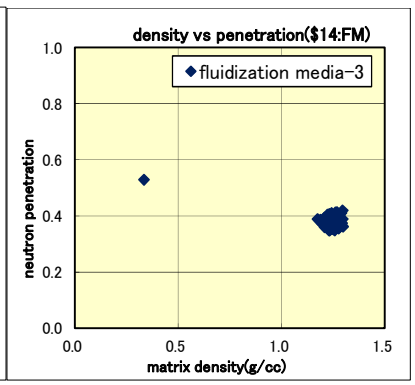




2. fluidization media(=FM)

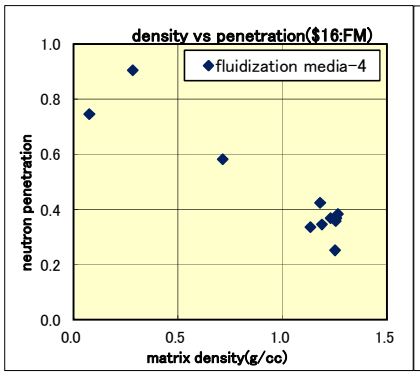


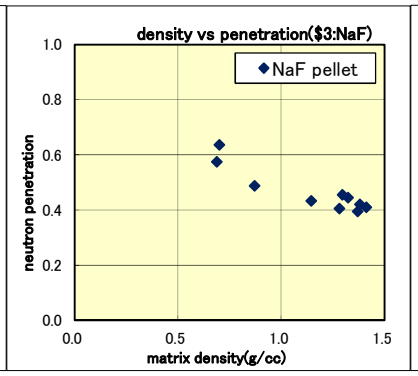


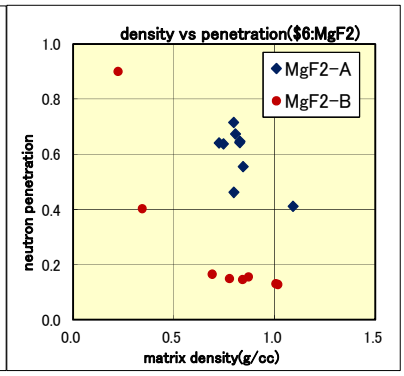


3. NaF pellets

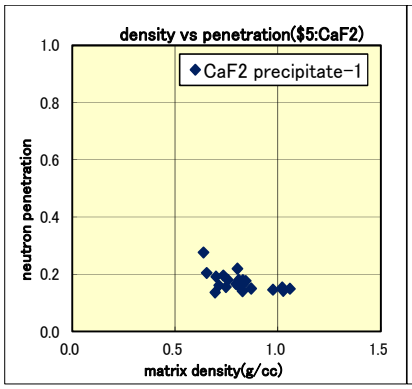
4. MgF2 precipitates

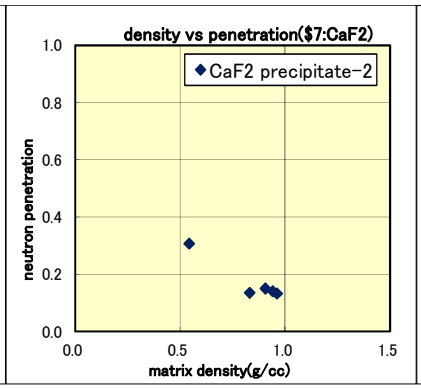






5. CaF2 precipitates





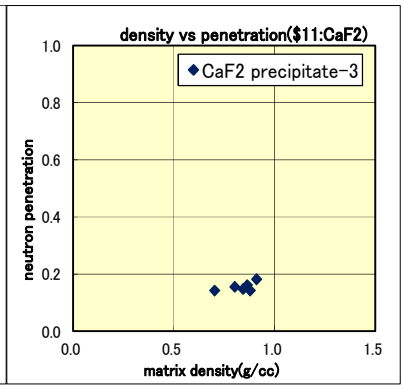


Figure-14　Correlation between Neutron Penetration and Wastes Density(1/2)

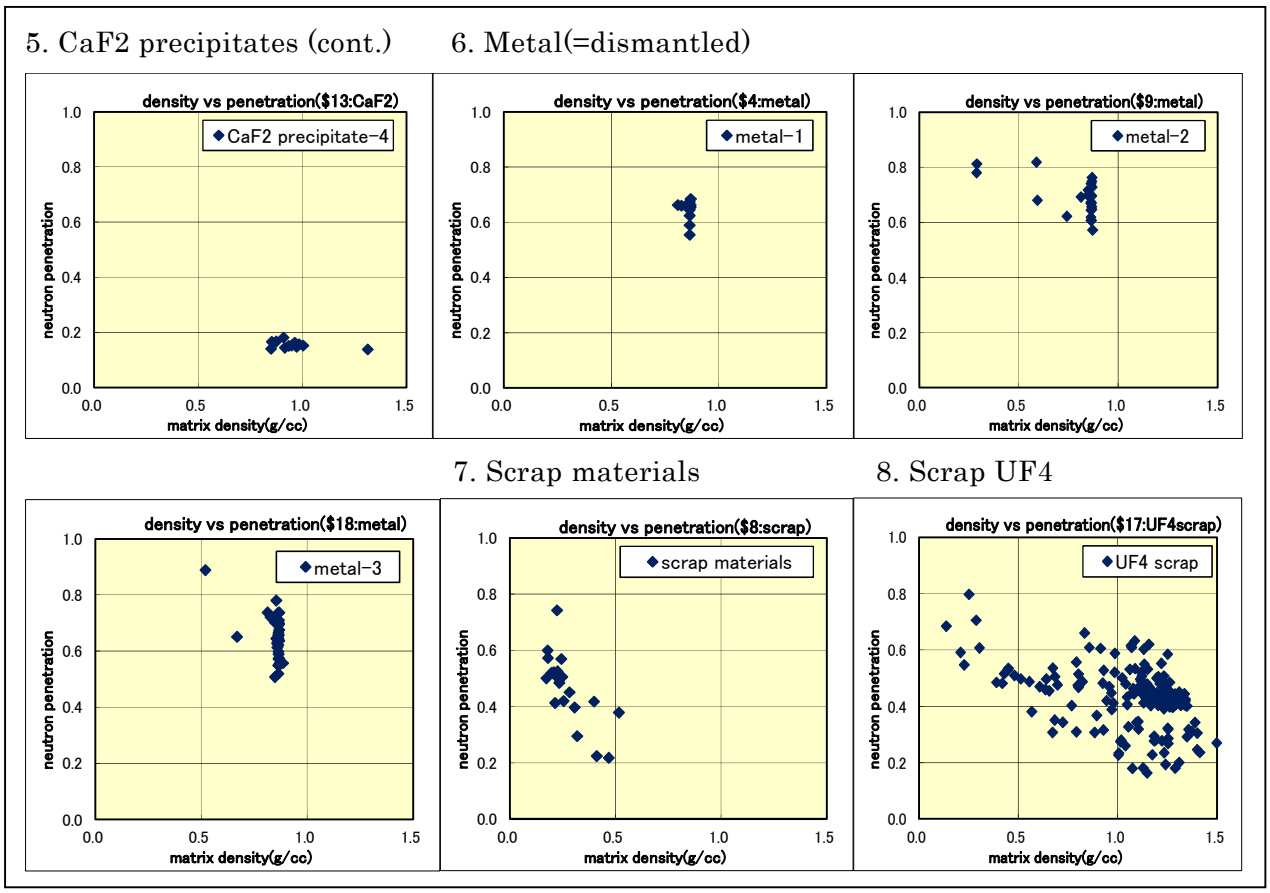


Figure-14　Correlation between Neutron Penetration and Wastes Density(2/2)

5.3　Review of Three Analysis Methods

As was described in **5.2**, three analysis methods have been separated depending on matrix materials conditions.　It is greatly important whether the mutual data are to be of one accord or not.　Apart from the data evaluated in **Table-5**, the validation trials had been performed for several cases.　The compared trends of two or three methods are listed in **Table-7** and **Figure-15**.　Obviously enough they were on good accord in homogeneous matrix materials within 30% approximately.　Otherwise in the case of heterogeneous matrix materials had been on poor accord because of instability of the "Fw" value.

Table-7　The Methods Comparison between Evaluated Uranium Mass

No.	kind of matrix materials	data	difference	remarks
1	alumina	132	2%	accorded well
2	fluidization media	324	18%	accorded well
3	NaF pellets	10	32%	
4	MgF_2 precipitates	18	250%	partially differed much
5	CaF_2 precipitates	98	54%	influenced by vapor as poison
6	metals	73	14%	accorded well
7	scrap materials	20	211%	partially differed much

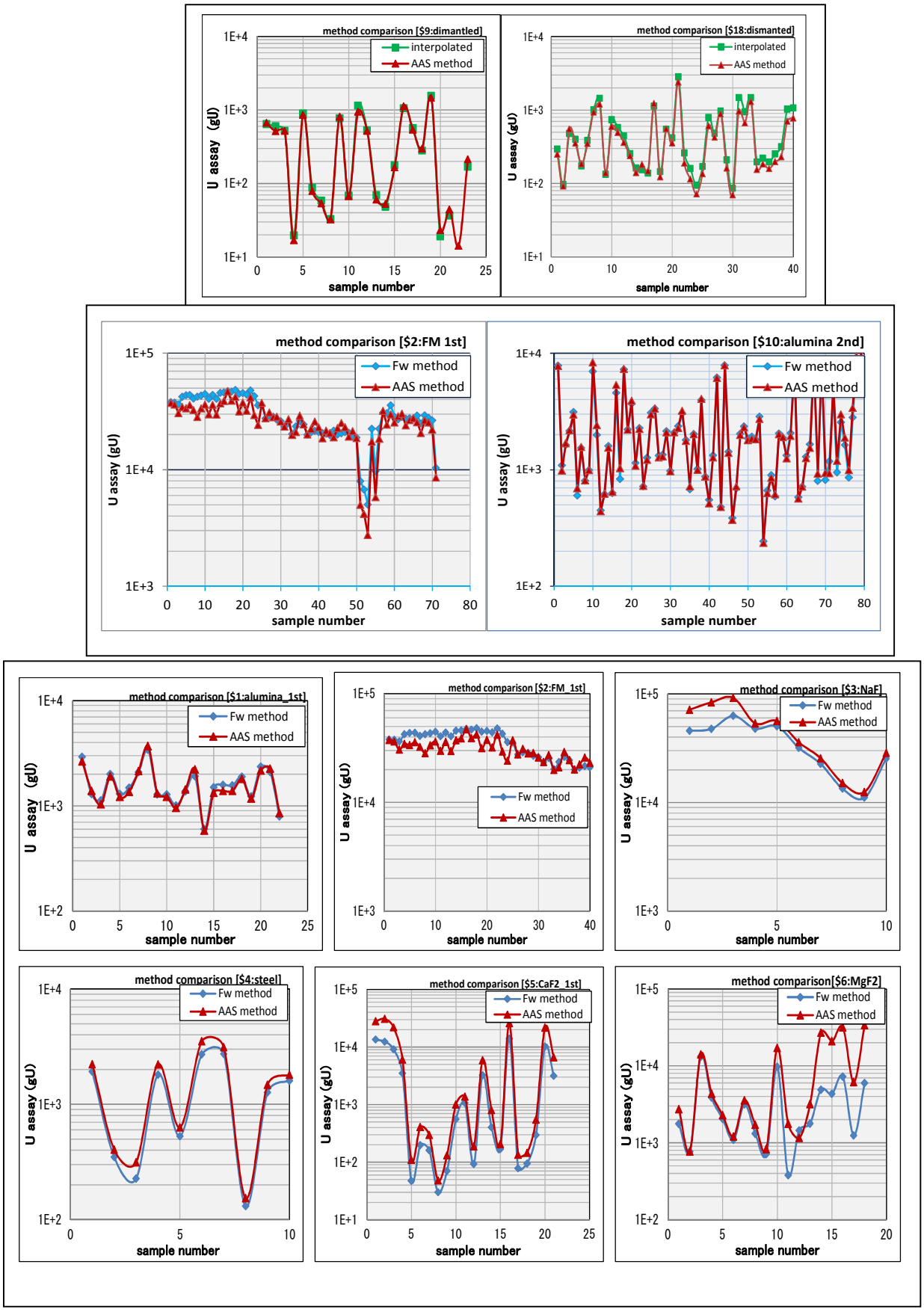


Figure-15　Comparison of Each Evaluated Method(1/2)

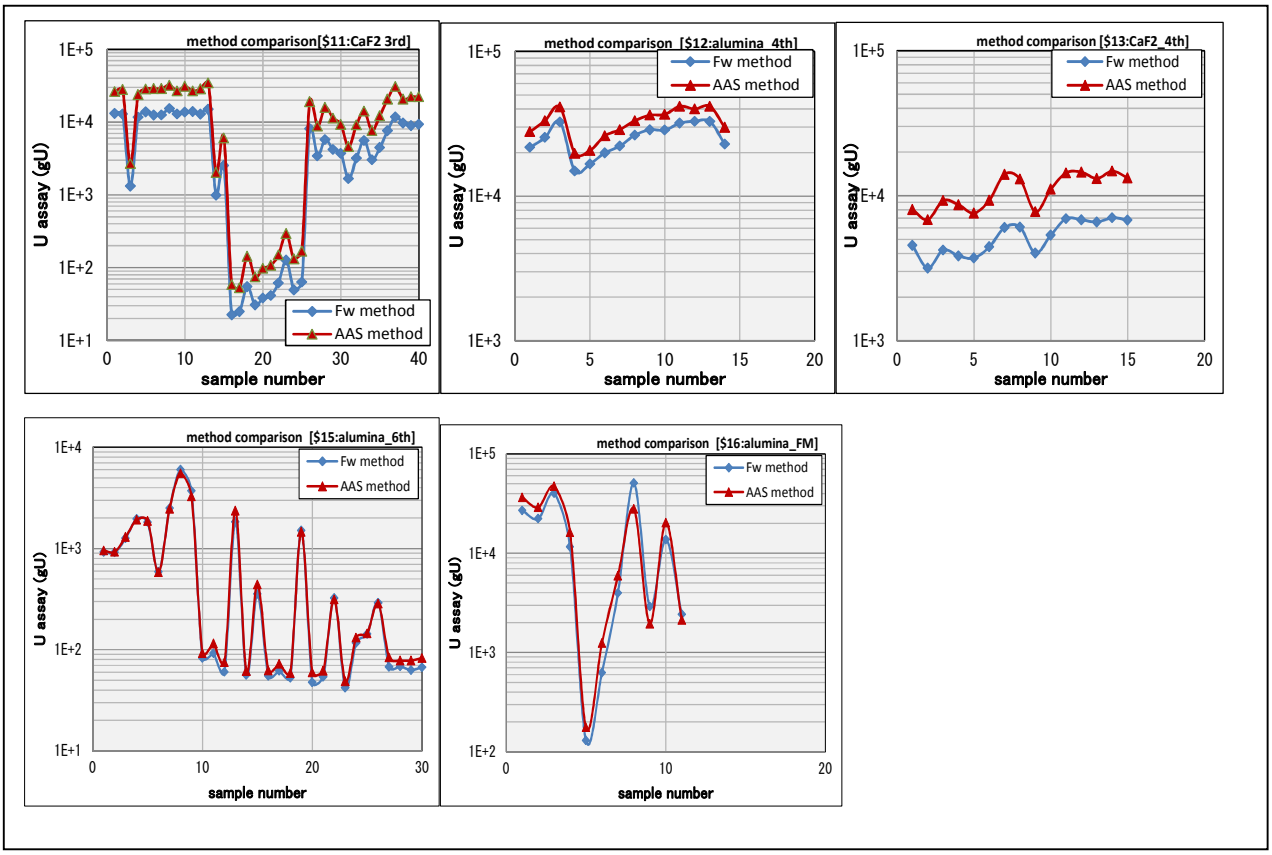


Figure-15　Comparison of Each Evaluated Method(2/2)

5.4　Trials for the Wastes Drums Contained with Uranium Oxides

Almost all wastes drums generated in URCP were contaminated with uranium fluorides except front chemical process which were flown with uranium oxides mainly. At the beginning the measurement works for the wastes drums contaminated with uranium fluorides have been carried out and achieve satisfactory results described above, however, it had not meant that no so much possibility but some potency to apply to the wastes for contaminated with uranium oxides. The mockup tests had proved half neutron response approximately described in **4.3** and shown in **Figure-13,** therefore it will be proved to be less dependency on chemical form in our passive neutron assay method. Such results had not dovetailed with the suggestion that the cross section of (α,n) reaction with oxygen as target atom indicates tenth part lower than with fluorine as target atom. [Ref.-6] Hence this variance will be remained as to be solved sooner or later.

The trial had been carried out to measure 20 actual drums with contaminated uranium oxides (recorded as UO_2, UO_3) in parallel with 20 actual drums with contaminated uranium fluorides, whose matrices were the steel fragments respectively. The “Fw” values were used the interpolated value corresponding the each density shown in **Figure-13**.

The spread data sheets consolidated whole parameters used for the determination uranium mass are shown in **Appendix-A($18).**

Fully satisfied determination uranium mass in case of uranium oxides as chemical form have been proven. In this regard, the detection limit value (estimated in **4.5**) may be multipled twice.

5.5 Verifications of Time Sequence

Regarding the counting time selection the most remarkable points of attention is the shortening relative error as less as possible with due considerations to suppress relative errors. At the beginning the counting time had been defined 60 minutes uniformly on the grounds that we had been informed less information about wastes drums. With the accumulation of measurements experiences, we had got the usual so-called “pay rate” for relative error. Therefore the new trial for counting time choice had been studied that the shortening time had been available in the case of high signal counts. Those target were aspired the followings :

i) more than 20 cps in net count rate permit 10 minutes measurement
ii) more than 10 cps in net count rate permit 20 minutes measurement
iii) more than 5 cps in net count rate permit 30 minutes measurement
iv) less than 5 cps in net count rate keep 60 minutes measurement

In these cases every data had kept less than 10 % error. Several examples which evaluated the relative error depending on the counting time are shown in **Figure-16.**

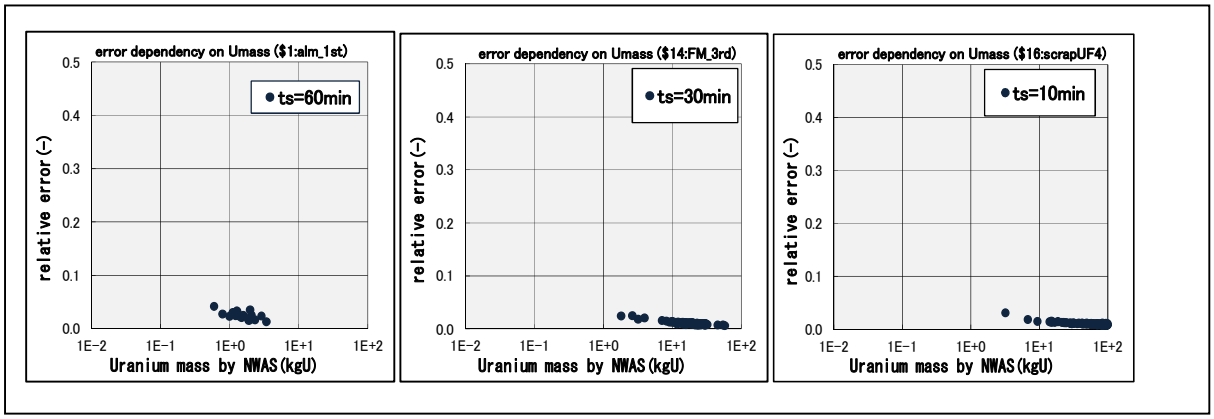


Figure-16 Relative Error Evaluations Depending on Uranium Mass

5.6 Validation of Uranium Enrichment Determination

For the purpose of the determination of uranium enrichment the gamma-ray energy analysis used Ge-SSD as was described in **4.6** had been carried out by to every wastes drum besides neutron detection. In previous report appointed “so much error”, the improvements of the shielding conditions had contributed low background condition. It

was successfully analyzed the uranium enrichment within 20% error by counting ratio of 186keV and 1001keV peaks of gamma ray spectrometry. The example data analyzed uranium enrichment is shown in **Appendix-B**. The good trend examples analyzed uranium enrichment is shown in **Figure-17.** In the case of "$11", the evaluated results had been clearly separated to two patterns, with small errors or with large errors, it was attributed to the amount of uranium mass. In the case of "$12" and, the "$13", the evaluated results had been traced the real uranium enrichment which in response to changes actually. In the case of "$14", the evaluated results had been duplicated the actual uranium enrichment as natural uranium except several points..

As might be expected the uranium enrichment had not identified in the wastes drums with no uranium. However another problem had been occurred that too much counting caused the pile up effect of Ge-SSD, which will be a strong contributor to identification of uranium enrichment error. The typical gamma ray spectrums and pile upped gamma ray spectrums are also shown in **Figure-18.**

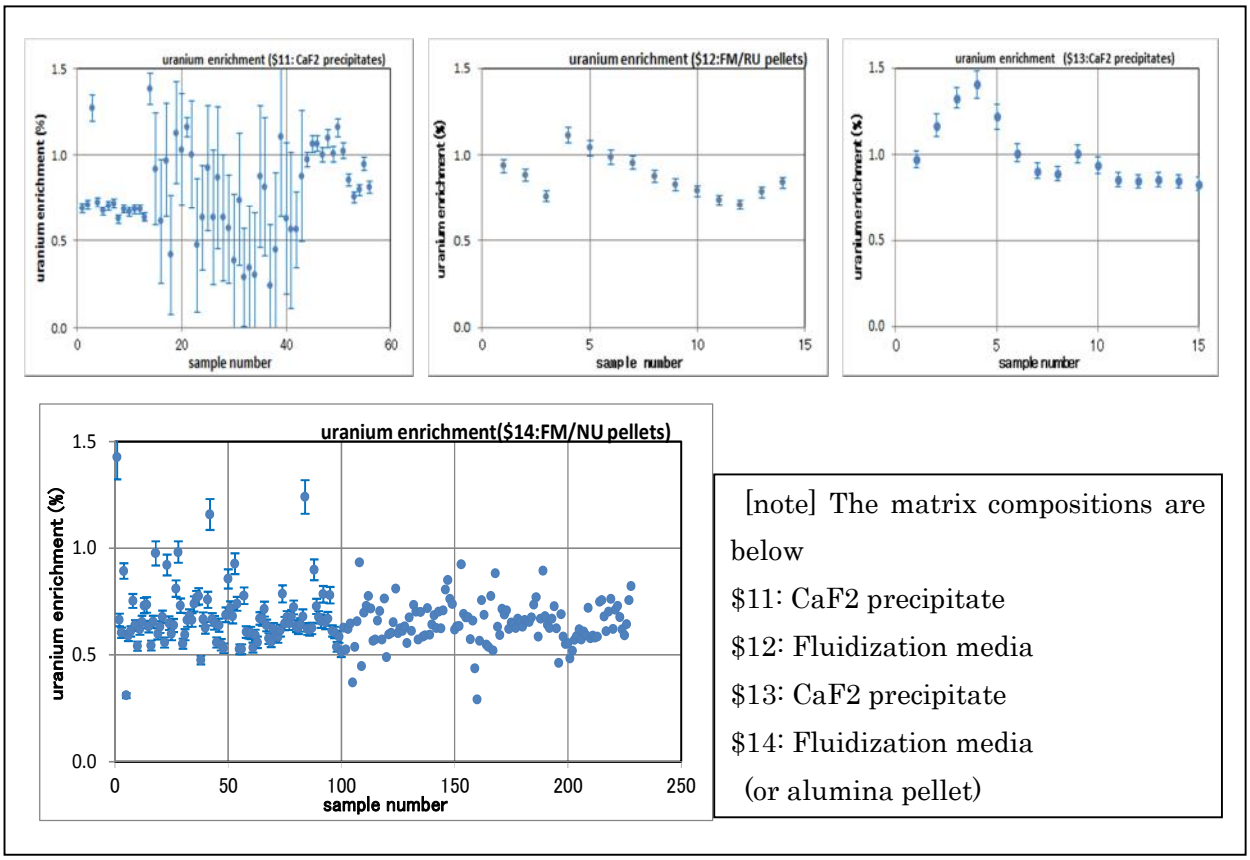
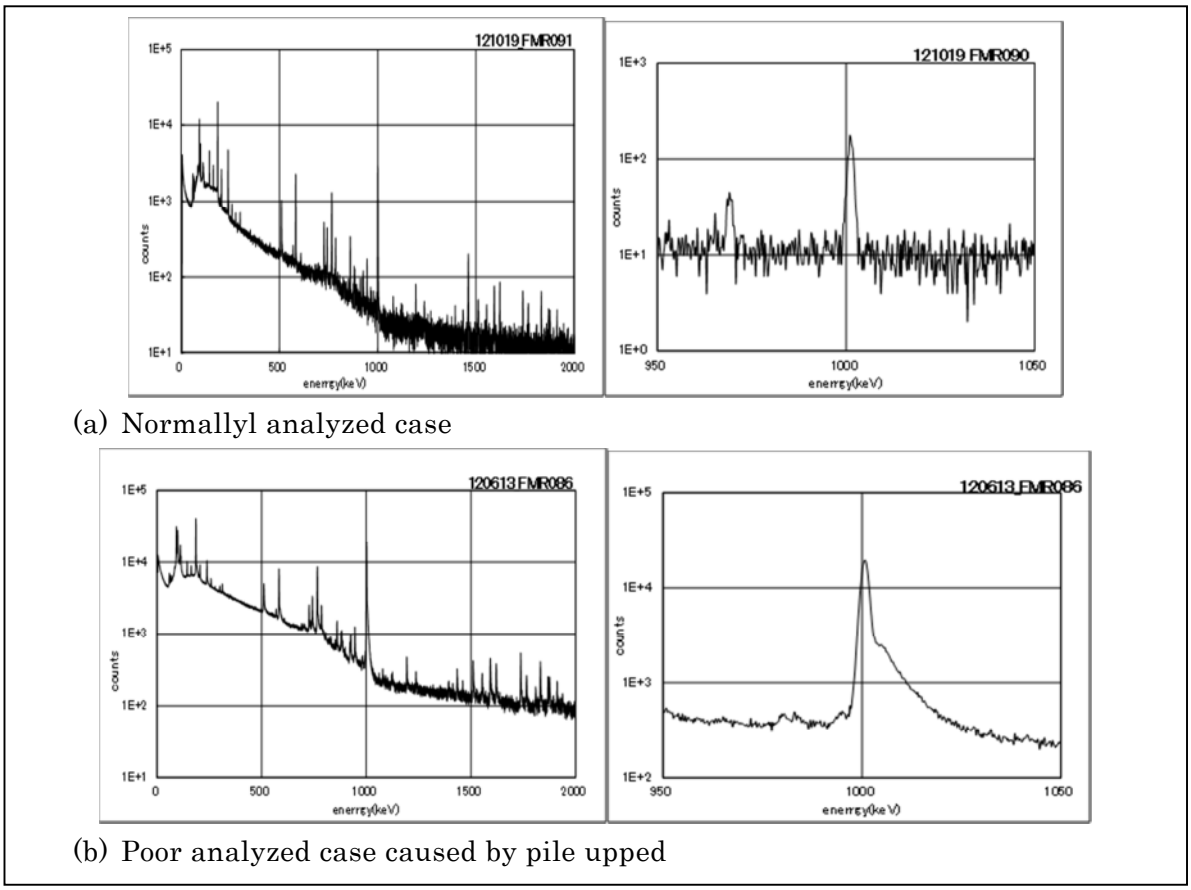


Figure-17 Trend Example of the Uranium Enrichment Determination



(a) Normallyl analyzed case

(b) Poor analyzed case caused by pile upped

Figure-18 Typical Gamma Spectrum of Uranium Bearing Wastes

6. Marshaled Action Assignment

6.1 Matrix Dependence

Described above the methodology for measuring and analyzing uranium mass are now establishing. We are now convincing the applicability to almost all actual wastes drums in URCP. However we don't mean to say that there are no problems around this methodology.

There is no denying it remains some actual drums which are not applicable to this methodology. From the beginning, the differences of neutron response between fluorides and oxides are to so much, one order or more, noted by LANL. [Ref.-3] Contrary to that, experimental neutron response data of oxides found to be 50-60% against that of fluorides approximately. These facts will support the potentiality to measure against uranium oxides wastes.

We do not refer to a decisive basis, however it's an interesting situation to reconsider the neutron emission rate of uranium oxides by (α,n)reaction. That is to say, the neutron emission rate of uranium oxides by (α,n)reaction are comparable to that of fluorides considering the contribution of oxygen-18 whose content is 0.205% with higher cross section of (α,n)reaction than oxygen-16.

6.2 Uncertainty of Uranium Enrichment Validation

To obtain good counting efficiency of the Ge-SSD is to be needed to attach the detector onto the wastes drums. The latest position was 65 mm from surface the drum. Doing so may cause not to put whole drum in perspective. As a result the uranium mass of gamma detection and neutron detection had not been assented.

In order to overcome such problems in the future the two selections are required whether to stand back the detector from drum in spite of decrease counting efficiency or to focus the barrow points of matrix materials and to determine local uranium mass by strong shielding collimator. Anyhow there often arise the situation in which the uranium enrichment had been hard to validate by gamma ray detection, that's the some of residual assignment.

6.3 Influences of Neutron Measurements Poison

To our surprise there are so many wastes drums with containing up to 50% moisture in URCP. Those were generated from chemical neutralization precipitates in major proportions of calcium fluorides. Off course any improvements for dehydrate process are to be expected before disposal, for a long time until now such conditions had been kept. Some actual wastes drums contain significant amount of water content (up to 50%), so hydrogen atoms in water would possibly interfere neutron detection. The methodology for accurate measuring such drums is to be purchased surely.

In general hydrogen atoms are apt to capture neutron, its cross section is far and away from other elements. In case of measurement works against wastes drums containing calcium fluorides these moisture problems had to be always considered any time in the view of decrease of counting efficiency or increase of relative error. Therefore additional tests were carried out how susceptible to those effects by adding water little by little to calcium fluorides powder. The **Figure-19** showed the counting decrease against added water. The influences by adding moisture had been no negligible up to 40% decrease had been observed but saturated on some level.

No matter how noted above, the determination processes for the wastes drums contained calcium fluorides powder are no failure used Add-A-Source method except the increase of detection limit. The decrease of neutron penetration or the decrease counting efficiency had been observed in **Figure-14(1/2).**

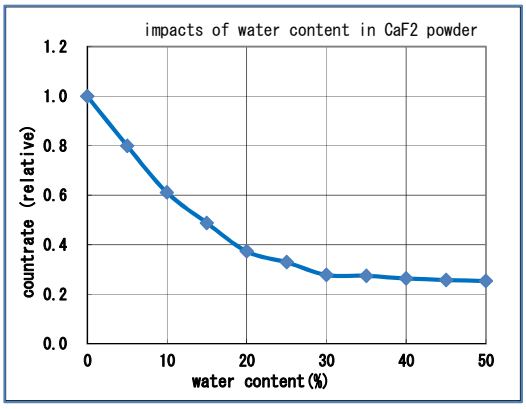


Figure-19 Influence of the Moisture in the Matrices

7. Results and Discussion

7.1 Tidemarks of NWAS Developments

Through three years experiences of NWAS we had learned our lessons so much.

Some artifices had contributed us the fairly good result, especially Added-A-Source method is supreme one. At the beginning it had been confused the handling the existence of "Fw", especially for unknown or mixed matrix materials, however the application of Added-A-Source method had solved the defects as long as uranium compounds are limited uranium fluorides. Therefore the wide range of uranium assay with good accuracy had been achieved and contributed MUF decrease in URCP as was the initial target.

In this regard there remain some problems concerning detection limit or measurement time for low uranium drums. As long as NWAS is based on passive uranium assay used (α,n) reaction, it is not released from chemical compound's influence. The neutron emission rate of uranium oxides are assumed to be approximately half of uranium fluorides experimentally, however those seem to be due to weak evidence considering natural content of oxygen-18, further studies are necessary.

7.2 Comparison against Gamma Uranium Assay

We had reached a conviction that the neutron assay method for uranium is superior to gamma assay from the view of good detector response and low background level. Actually we had originally been using large sized NaI(Tl) scintillation detector for the purpose of determining uranium mass in parallel with neutron detection. [Ref.-1] That system, however, had been revealed the high background and poor energy resolution, so we had abandoned to use it describe previously. On the other hand the experiences of the gamma detection by Ge-SSD had taught us the difficulty precise determination uranium mass because of its low counting efficiency and its characteristics. After all the gamma detections had been used for only the uranium enrichments.

8. Perspective View of Active Assay System

8.1 Viewpoints of the Improvement of NWAS

Full experience made great contributions to the precise nuclear materials control through uranium assay. At the same time it had to be encountered to require the improvement points of NWAS, that is to say, to overcome the problems regarding uranium chemical composition dependency, high sensitive uranium assay and shortening the counting time. The further study of measurement methods is needed. Our trials will surely contribute to improve safe-guards data. Furthermore our trials are to be involved the possibility for uranium clearance validation measurements, whose requirements are to be verified to measure below 1Bq/gram matrix materials in

concentration.

Further improvements, for example far more well detection sensitivity, less dependence on matrices and so on, will be expected so as to far more accuracy or response, the next plans are to be described roughly in **Appendix-D**.

8.2 Utilization of Passive Neutron Assay Experiences

Generally spoken, neutron detection technique is comparatively more difficult than gamma ray detection technique, however, we had validated to be inappropriate. The stable and reliable neutron assay will be proved by significant much know-how for handling and calibrating neutron detection accumulated through four years experiences. In active neutron assay system the same Helium-3 proportional counters and high voltage supplier are to be used. The maintenances of detectors are also the most important, we are able to utilize neutron assay systems from our experiences.

9. Conclusion

The project of passive uranium assay system “NWAS” had been validated its good accuracy, safe handling and effectiveness for the application to the actual wastes drums. Our trials for the passive uranium assay had come to fruition successfully and just finished once, the whole system had been dismantled to each part. Through these works have contributed the decrease the MUF of URCP, for which was the first purpose of introduction of NWAS.

Despite purchasing multilateral knowledge, it had not been fully analyzed against so-called “unknown” wastes drums, that is to say, some pre-information had been needed. To the next active uranium assay project the amount knowledge extracted this project will be transferred.

Acknowledgements

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Appendix-A

Table-A1 Uranium Determination Spread Sheet Extracted Several Matrices

($11 : CaF2 precipitates RU ---by "C" =AAS method)

No.	Sample Description	Number of Cycles	Cycle Count Time	Total Count Time	Singles (cps)	Singles Err	BKG (cps)	BKG Err	Calcul. Enrich a/(a+b)	counting effi ciency	weighing factor	neutron emission (n/s/gU)	**Neutron Assay (gU)**	gU Err
1	CaF2	298	12	3576	50.924	0.127	5.917	0.009	0.71	0.0089	0.288	0.218	**2.62E+4**	1.54E+3
2	CaF2	298	12	3576	49.863	0.126	5.917	0.009	0.71	0.0082	0.288	0.218	**2.79E+4**	1.64E+3
3	CaF2	292	12	3504	5.139	0.057	5.917	0.009	1.27	0.0087	0.288	0.378	**1.55E+3**	1.30E+2
4	CaF2	298	12	3576	45.43	0.121	5.917	0.009	0.73	0.0087	0.288	0.222	**2.34E+4**	1.38E+3
5	CaF2	298	12	3576	53.733	0.130	5.917	0.009	0.71	0.0087	0.288	0.218	**2.85E+4**	1.67E+3
6	CaF2	297	12	3564	48.638	0.125	5.917	0.009	0.71	0.0078	0.288	0.217	**2.90E+4**	1.71E+3
7	CaF2	299	12	3588	49.023	0.125	5.917	0.009	0.72	0.0078	0.288	0.220	**2.86E+4**	1.68E+3
8	CaF2	295	12	3540	59.672	0.137	5.917	0.009	0.71	0.0086	0.288	0.218	**3.20E+4**	1.87E+3
9	CaF2	300	12	3600	50.483	0.126	4.548	0.008	0.71	0.0087	0.288	0.218	**2.68E+4**	1.58E+3
10	CaF2	299	12	3588	53.206	0.129	4.548	0.008	0.71	0.0079	0.288	0.218	**3.09E+4**	1.83E+3

Table-A2 Uranium Determination Spread Sheet Extracted Several Matrices

($12 : alumina-FM RU --- by "A" =Fw value method)

No.	Sample Description	Number of Cycles	Cycle Count Time	Total Count Time	Singles (cps)	Singles Err	BKG (cps)	BKG Err	Calcul. Enrich a/(a+b)	counting effi ciency	weighing factor	neutron emission (n/s/gU)	**Neutron Assay (gU)**	gU Err
1	alumina	300	12	3600	193.283	0.236	5.772	0.009	0.94	0.0540	0.138	0.142	**2.51E+4**	1.41E+3
2	alumina	297	12	3564	226.103	0.257	5.772	0.009	0.89	0.0540	0.138	0.136	**3.09E+4**	1.72E+3
3	alumina	297	12	3564	289.931	0.29	5.772	0.009	0.76	0.0540	0.138	0.118	**4.54E+4**	2.52E+3
4	alumina	300	12	3600	132.769	0.197	5.772	0.009	1.11	0.0540	0.138	0.167	**1.47E+4**	8.32E+2
5	alumina	297	12	3564	148.537	0.209	5.772	0.009	1.04	0.0540	0.138	0.157	**1.76E+4**	9.89E+2
6	alumina	300	12	3600	177.124	0.227	5.772	0.009	0.99	0.0540	0.138	0.149	**2.20E+4**	1.23E+3
7	alumina	297	12	3564	197.83	0.24	5.772	0.009	0.96	0.0540	0.138	0.145	**2.52E+4**	1.41E+3
8	alumina	298	12	3576	235.066	0.261	5.772	0.009	0.88	0.0540	0.138	0.134	**3.25E+4**	1.81E+3
9	alumina	300	12	3600	255.984	0.271	5.772	0.009	0.83	0.0540	0.138	0.128	**3.71E+4**	2.07E+3
10	alumina	300	12	3600	254.705	0.271	5.772	0.009	0.79	0.0540	0.138	0.123	**3.85E+4**	2.14E+3

Table-A3 Uranium Determination Spread Sheet Extracted Several Matrices

($13 : CaF2 precipitates RU --- by "C" =AAS method)

No.	Sample Description	Number of Cycles	Cycle Count Time	Total Count Time	Singles (cps)	Singles Err	BKG (cps)	BKG Err	Calcul. Enrich a/(a+b)	counting effi ciency	weighing factor	neutron emission (n/s/gU)	**Neutron Assay (gU)**	gU Err
1	CaF2	148	12	1776	23.024	0.125	4.487	0.009	0.98	0.010	0.288	0.294	**8.02E+3**	5.41E+2
2	CaF2	149	12	1788	17.857	0.112	4.487	0.009	1.10	0.008	0.288	0.330	**6.83E+3**	4.77E+2
3	CaF2	296	12	3552	23.764	0.092	5.400	0.012	1.10	0.008	0.288	0.330	**9.25E+3**	5.87E+2
4	CaF2	299	12	3588	21.65	0.088	5.400	0.012	1.10	0.008	0.288	0.330	**8.67E+3**	5.54E+2
5	CaF2	148	12	1776	20.901	0.12	4.487	0.009	1.10	0.008	0.288	0.330	**7.58E+3**	5.17E+2
6	CaF2	150	12	1800	23.19	0.124	4.487	0.009	1.01	0.008	0.288	0.304	**9.29E+3**	6.24E+2
7	CaF2	157	12	1884	28.729	0.133	4.487	0.009	0.91	0.007	0.288	0.274	**1.40E+4**	9.14E+2
8	CaF2	144	12	1728	28.519	0.139	4.487	0.009	0.89	0.008	0.288	0.270	**1.30E+4**	8.57E+2
9	CaF2	150	12	1800	20.932	0.119	4.487	0.009	1.01	0.009	0.288	0.303	**7.75E+3**	5.27E+2
10	CaF2	149	12	1788	26.307	0.132	4.487	0.009	0.94	0.008	0.288	0.284	**1.11E+4**	7.35E+2

[notes]

Table-A4 Uranium Determination Spread Sheet Extracted Several Matrices

($14 : alumina-FM NU --- by "C" =AAS method)

No.	Sample Description	Number of Cycles	Cycle Count Time	Total Count Time	Singles (cps)	Singles Err	BKG (cps)	BKG Err	Calcul. Enrich a/(a+b)	counting effi ciency	weighing factor	neutron emission (n/s/gU)	**Neutron Assay (gU)**	gU Err
1	alumina	295	12	3540	115.978	0.187	5.365	0.009	0.62	0.020	0.288	0.218	**2.61E+4**	1.49E+3
2	alumina	150	12	1800	98.034	0.243	5.733	0.009	0.65	0.020	0.288	0.218	**2.19E+4**	1.29E+3
3	alumina	150	12	1800	74.373	0.213	5.733	0.009	0.73	0.020	0.288	0.218	**1.71E+4**	1.02E+3
4	alumina	150	12	1800	72.618	0.211	5.733	0.009	0.73	0.020	0.288	0.218	**1.63E+4**	9.75E+2
5	alumina	148	12	1776	113.877	0.262	5.733	0.009	0.63	0.021	0.288	0.218	**2.47E+4**	1.44E+3
6	alumina	150	12	1800	130.02	0.278	5.733	0.009	0.54	0.021	0.288	0.218	**2.82E+4**	1.64E+3
7	alumina	150	12	1800	103.286	0.249	5.733	0.009	0.66	0.021	0.288	0.218	**2.29E+4**	1.34E+3
8	alumina	150	12	1800	37.182	0.156	5.592	0.013	0.98	0.021	0.288	0.218	**8.29E+3**	5.34E+2
9	alumina	150	12	1800	93.108	0.237	5.592	0.013	0.60	0.021	0.288	0.218	**2.00E+4**	1.21E+3
10	alumina	149	12	1788	103.042	0.249	5.592	0.013	0.71	0.020	0.288	0.218	**2.36E+4**	1.42E+3

Table-A5 Uranium Determination Spread Sheet Extracted Several Matrices

($15 : alumina RU --- by "C" =AAS method)

No.	Sample Description	Number of Cycles	Cycle Count Time	Total Count Time	Singles (cps)	Singles Err	BKG (cps)	BKG Err	Declared Enrich. (%)	counting effi ciency	weighing factor	neutron emission (n/s/gU)	Neutron Assay (gU)	gU Err
1	alumina	293	12	3516	8.241	0.059	3.742	0.007	1.1	0.025	0.288	0.330	**9.86E+2**	7.12E+1
2	alumina	294	12	3528	8.141	0.059	3.742	0.007	1.1	0.026	0.288	0.330	**9.56E+2**	6.93E+1
3	alumina	294	12	3528	11.287	0.066	3.742	0.007	1.1	0.026	0.288	0.330	**1.34E+3**	9.18E+1
4	alumina	297	12	3564	17.476	0.078	3.742	0.007	1.1	0.027	0.288	0.330	**1.98E+3**	1.28E+2
5	alumina	296	12	3552	16.239	0.076	3.742	0.007	1.1	0.025	0.288	0.330	**1.94E+3**	1.26E+2
6	alumina	294	12	3528	5.257	0.051	3.742	0.007	1.1	0.027	0.288	0.330	**5.99E+2**	4.77E+1
7	alumina	300	12	3600	22.217	0.086	3.803	0.007	1.1	0.026	0.288	0.330	**2.56E+3**	1.61E+2
8	alumina	298	12	3576	53.702	0.128	3.803	0.007	1.1	0.029	0.288	0.330	**5.70E+3**	3.37E+2
9	alumina	300	12	3600	32.995	0.102	3.803	0.007	1.1	0.030	0.288	0.330	**3.38E+3**	2.05E+2
10	alumina	295	12	3540	0.741	0.037	3.803	0.007	1.1	0.023	0.288	0.330	**9.56E+1**	1.91E+1

Table-A6 Uranium Determination Spread Sheet Extracted Several Matrices

($17 : scrap UF4 RU --- by "C" =AAS method)

No.	Sample Description	Number of Cycles	Cycle Count Time	Total Count Time	Singles (cps)	Singles Err	BKG (cps)	BKG Err	Calcul. Enrich a/(a+b)	counting effi ciency	weighing factor	neutron emission (n/s/gU)	**Neutron Assay (gU)**	gU Err
1	(scrap UF4)	50	12	600	428.232	0.860	4.399	0.008	1.1	0.026	0.288	0.330	**4.98E+4**	2.89E+3
2	(scrap UF4)	50	12	600	525.255	0.952	4.399	0.008	1.1	0.034	0.288	0.330	**4.66E+4**	2.69E+3
3	(scrap UF4)	50	12	600	481.155	0.912	4.399	0.008	1.1	0.027	0.288	0.330	**5.39E+4**	3.12E+3
4	(scrap UF4)	50	12	600	534.845	0.961	4.399	0.008	1.1	0.030	0.288	0.330	**5.46E+4**	3.15E+3
5	(scrap UF4)	50	12	600	596.777	1.016	4.399	0.008	1.1	0.022	0.288	0.330	**8.30E+4**	4.77E+3
6	(scrap UF4)	50	12	600	563.168	0.988	4.399	0.008	1.1	0.023	0.288	0.330	**7.45E+4**	4.29E+3
7	(scrap UF4)	50	12	600	578.181	1.001	4.399	0.008	1.1	0.026	0.288	0.330	**6.69E+4**	3.85E+3
8	(scrap UF4)	50	12	600	514.622	0.945	4.399	0.008	1.1	0.023	0.288	0.330	**6.85E+4**	3.96E+3
9	(scrap UF4)	50	12	600	550.824	0.975	4.399	0.008	1.1	0.022	0.288	0.330	**7.66E+4**	4.41E+3
10	(scrap UF4)	50	12	600	502.282	0.932	4.399	0.008	1.1	0.024	0.288	0.330	**6.30E+4**	3.64E+3

[note] NU is classified natural uranium, RU is classified reprocessed uranium.

Table-A7 Uranium Determination Spread Sheet Extracted Several Matrices

($18 :dismantled RU --- by "B" =variable Fw value method)

No.	Sample Description	Number of Cycles	Cycle Count Time	Total Count Time	Singles (cps)	Singles Err	BKG (cps)	BKG Err	Calcul. Enrich a/(a+b)	counting effi ciency	weighing factor	neutron emission (n/s/gU)	**Neutron Assay (gU)**	gU Err
1	steel	287	12	3444	3.1	0.047	4.238	0.008	1.1	0.054	0.17	0.198	**2.93E+2**	2.80E+1
2	steel	297	12	3564	1.0	0.039	4.238	0.008	1.1	0.054	0.17	0.198	**9.67E+1**	1.58E+1
3	steel	296	12	3552	5.1	0.052	4.238	0.008	1.1	0.054	0.17	0.198	**4.78E+2**	3.88E+1
4	steel	296	12	3552	4.3	0.05	4.238	0.008	1.1	0.054	0.17	0.198	**4.00E+2**	3.43E+1
5	steel	291	12	3492	1.8	0.043	4.238	0.008	1.1	0.054	0.17	0.197	**1.73E+2**	2.08E+1
6	steel	288	12	3456	4.1	0.05	4.251	0.008	1.1	0.054	0.17	0.198	**3.84E+2**	3.34E+1
7	steel	293	12	3516	10.7	0.066	4.238	0.008	1.1	0.054	0.17	0.198	**1.00E+3**	6.97E+1
8	steel	294	12	3528	15.4	0.075	4.238	0.008	1.1	0.054	0.17	0.198	**1.44E+3**	9.46E+1
9	steel	285	12	3420	1.4	0.041	4.238	0.008	1.1	0.054	0.17	0.198	**1.34E+2**	1.82E+1
10	steel	290	12	3480	7.9	0.06	4.238	0.008	1.1	0.054	0.17	0.198	**7.37E+2**	5.42E+1

No.	Sample Description	Number of Cycles	Cycle Count Time	Total Count Time	Singles (cps)	Singles Err	BKG (cps)	BKG Err	Calcul. Enrich a/(a+b)	counting effi ciency	weighing factor	neutron emission (n/s/gU)	**Neutron Assay (gU)**	gU Err
11	steel	297	12	3564	13.8	0.071	4.005	0.008	1.1	0.054	0.07	0.090	**2.83E+3**	1.89E+2
12	steel	297	12	3564	1.6	0.041	4.005	0.008	1.1	0.054	0.09	0.115	**2.60E+2**	3.29E+1
13	steel	298	12	3576	0.8	0.037	4.005	0.008	1.1	0.054	0.07	0.090	**1.60E+2**	3.08E+1
14	steel	290	12	3480	0.5	0.037	3.968	0.008	1.1	0.054	0.07	0.090	**9.49E+1**	2.76E+1
15	steel	292	12	3504	0.8	0.038	4.005	0.008	1.1	0.054	0.07	0.090	**1.71E+2**	3.20E+1
16	steel	291	12	3492	3.8	0.048	4.005	0.008	1.1	0.054	0.07	0.090	**7.90E+2**	6.94E+1
17	steel	295	12	3540	2.7	0.044	4.005	0.008	1.1	0.054	0.08	0.103	**4.82E+2**	4.81E+1
18	steel	297	12	3564	4.7	0.05	3.968	0.008	1.1	0.054	0.07	0.090	**9.70E+2**	7.98E+1
19	steel	294	12	3528	1.0	0.039	4.005	0.008	1.1	0.054	0.07	0.090	**2.10E+2**	3.46E+1
20	steel	279	12	3348	0.4	0.037	3.968	0.008	1.1	0.054	0.07	0.090	**8.61E+1**	2.71E+1

[note] The data from 1 to 10 are uranium fluorides, from 11 to 20 are uranium oxides.

The weighing factors are different.

RU is classified reprocessed uranium.

Appendix-B

Uranium Enrichment Measurement Spread Sheet ($11 : CaF2 precipitates RU)

No.	meas. time (sec)	a)186keV (cts)	Err (1sigma)	b)1001keV (cts)	Err (1sigma)	a)186keV (cts/sec)	Err (3sigma)	b)1001keV (cts/sec)	Err (3sigma)	a)U235 (gU)	Err (3sigma)	b)U238 (gU)	Err (3sigma)	**Calcul. Enrich (%)**	Calcul. Enrich Err(3%)
1	3600	5.6E+5	1.2E+3	2.8E+5	5.6E+2	1.5E+2	1.0E+0	7.9E+1	4.7E-1	5.4E+1	3.6E-1	7.7E+3	4.6E+1	**6.9E-1**	2.8E-2
2	3600	5.8E+5	8.5E+2	2.9E+5	5.6E+2	1.6E+2	7.1E-1	7.9E+1	4.7E-1	5.6E+1	2.5E-1	7.8E+3	4.6E+1	**7.1E-1**	2.7E-2
3	3600	7.9E+4	4.0E+2	2.2E+4	1.5E+2	2.2E+1	3.4E-1	6.0E+0	1.2E-1	7.6E+0	1.2E-1	5.9E+2	1.2E+1	**1.3E+0**	7.6E-2
4	3600	5.0E+5	7.9E+2	2.4E+5	5.1E+2	1.4E+2	6.6E-1	6.7E+1	4.3E-1	4.8E+1	2.3E-1	6.6E+3	4.2E+1	**7.3E-1**	2.8E-2
5	3600	6.2E+5	8.8E+2	3.2E+5	6.0E+2	1.7E+2	7.3E-1	8.9E+1	5.0E-1	5.9E+1	2.5E-1	8.7E+3	4.9E+1	**6.7E-1**	2.6E-2
6	3600	5.6E+5	8.3E+2	2.8E+5	5.5E+2	1.6E+2	6.9E-1	7.7E+1	4.6E-1	5.4E+1	2.4E-1	7.6E+3	4.5E+1	**7.1E-1**	2.7E-2
7	3600	5.8E+5	8.4E+2	2.8E+5	5.6E+2	1.6E+2	7.0E-1	7.8E+1	4.6E-1	5.6E+1	2.4E-1	7.7E+3	4.6E+1	**7.2E-1**	2.8E-2
8	3600	6.5E+5	9.0E+2	3.7E+5	6.4E+2	1.8E+2	7.5E-1	1.0E+2	5.4E-1	6.3E+1	2.6E-1	1.0E+4	5.3E+1	**6.2E-1**	2.4E-2
9	3600	5.8E+5	8.4E+2	2.9E+5	5.7E+2	1.6E+2	7.0E-1	8.2E+1	4.8E-1	5.5E+1	2.4E-1	8.0E+3	4.7E+1	**6.9E-1**	2.6E-2
10	3600	6.1E+5	8.7E+2	3.2E+5	6.0E+2	1.7E+2	7.3E-1	8.9E+1	5.0E-1	5.9E+1	2.5E-1	8.7E+3	4.9E+1	**6.7E-1**	2.6E-2

Uranium Enrichment Measurement Spread Sheet ($12 : alumina-FM)

No.	sample No.	Filename	Date	Sample Description	Decl. Enrich (%)	meas. time (sec)	a)186keV (cts)	Err (1sigma)	b)1001 keV (cts)	Err (1sigma)	a)186keV (cts/s)	b)1001 keV (cts/s)	a)U235 (gU)	b)U238 (gU)	**Calcul. Enrich (%)**	Calcul. Enrich Err(3%)
1	ALUF4-R-072	120423_FMR072	2012/4/23	alumina	1.1	3600	5.3E+5	9.7E+2	2.2E+5	5.1E+2	1.5E+2	6.0E+1	6.0E+1	6.3E+3	**9.4E-1**	3.8E-2
2	ALUF4-R-073	120423_FMR073	2012/4/23	alumina	1.1	3600	6.4E+5	1.0E+3	2.7E+5	5.7E+2	1.8E+2	7.5E+1	7.1E+1	8.0E+3	**8.9E-1**	3.5E-2
3	ALUF4-R-074	120423_FMR074	2012/4/23	alumina	1.1	3600	6.7E+5	1.1E+3	3.3E+5	6.4E+2	1.9E+2	9.3E+1	7.5E+1	9.8E+3	**7.6E-1**	3.0E-2
4	ALUF4-R-075	120424_FMR075	2012/4/24	alumina	1.1	3600	4.8E+5	7.7E+2	1.6E+5	3.8E+2	1.3E+2	4.5E+1	5.3E+1	4.7E+3	**1.1E+0**	4.4E-2
5	ALUF4-R-076	120424_FMR076	2012/4/24	alumina	1.1	3600	4.8E+5	8.6E+2	1.7E+5	4.5E+2	1.3E+2	4.8E+1	5.3E+1	5.1E+3	**1.0E+0**	4.2E-2
6	ALUF4-R-077	120424_FMR077	2012/4/24	alumina	1.1	3600	5.4E+5	9.5E+2	2.1E+5	5.0E+2	1.5E+2	5.8E+1	6.1E+1	6.1E+3	**9.9E-1**	4.0E-2
7	ALUF4-R-078	120424_FMR078	2012/4/24	alumina	1.1	3600	5.9E+5	1.0E+3	2.4E+5	5.3E+2	1.7E+2	6.5E+1	6.7E+1	6.9E+3	**9.6E-1**	3.8E-2
8	ALUF4-R-079	120425_FMR079	2012/4/25	alumina	1.1	3600	6.5E+5	1.1E+3	2.8E+5	5.8E+2	1.8E+2	7.8E+1	7.3E+1	8.2E+3	**8.8E-1**	3.4E-2
9	ALUF4-R-080	120425_FMR080	2012/4/25	alumina	1.1	3600	6.7E+5	1.1E+3	3.1E+5	6.1E+2	1.9E+2	8.5E+1	7.5E+1	9.0E+3	**8.3E-1**	3.2E-2
10	ALUF4-R-081	120425_FMR081	2012/4/25	alumina	1.1	3600	6.3E+5	1.1E+3	3.0E+5	6.0E+2	1.8E+2	8.4E+1	7.1E+1	8.9E+3	**7.9E-1**	3.1E-2

Uranium Enrichment Measurement Spread Sheet ($13 : CaF2 precipitates NU)

No.	sample No.	Sample Description	Decl. Enrich (%)	meas. time (sec)	a)186keV (cts)	Err (1sigma)	b)1001keV (cts)	Err (1sigma)	a)186keV (cps)	Err (3sigma)	b)1001keV (cps)	Err (3sigma)	a)U235 (gU)	Err (3sigma)	b)U238 (gU)	Err (3sigma)	**Calcul. Enrich (%)**	Calcul. Enrich Err
1	20-R転-1	CaF2	0.711	1800	1.6E+5	5.8E+2	5.6E+4	2.4E+2	8.7E+1	9.7E-1	3.1E+1	4.1E-1	3.0E+1	3.4E-1	3.0E+3	4.0E+1	**9.8E-1**	4.9E-2
2	20-R転-2	CaF2	0.711	1800	9.9E+4	4.9E+2	3.0E+4	1.8E+2	5.5E+1	8.1E-1	1.6E+1	2.9E-1	1.9E+1	2.8E-1	1.6E+3	2.9E+1	**1.2E+0**	6.7E-2
3	20-R転-3	CaF2	0.711	3600	1.7E+5	2.0E+2	4.4E+4	2.2E+2	4.7E+1	1.7E-1	1.2E+1	1.8E-1	1.6E+1	5.7E-2	1.2E+3	1.8E+1	**1.3E+0**	6.1E-2
4	20-R転-4	CaF2	0.711	3600	1.5E+5	7.0E+2	3.7E+4	2.0E+2	4.1E+1	5.8E-1	1.0E+1	1.6E-1	1.4E+1	2.0E-1	1.0E+3	1.6E+1	**1.4E+0**	7.8E-2
5	20-R転-5	CaF2	0.711	1800	1.0E+5	5.7E+2	2.8E+4	1.7E+2	5.5E+1	9.6E-1	1.6E+1	2.9E-1	1.9E+1	3.3E-1	1.6E+3	2.9E+1	**1.2E+0**	7.4E-2
6	20-R転-6	CaF2	0.711	1800	1.6E+5	5.9E+2	5.3E+4	2.4E+2	8.6E+1	9.9E-1	3.0E+1	4.0E-1	3.0E+1	3.4E-1	2.9E+3	3.9E+1	**1.0E+0**	5.2E-2
7	20-R転-7	CaF2	0.711	1800	1.8E+5	7.0E+2	6.8E+4	2.7E+2	9.8E+1	1.2E+0	3.8E+1	4.5E-1	3.4E+1	4.0E-1	3.7E+3	4.5E+1	**9.1E-1**	4.6E-2
8	20-R転-8	CaF2	0.711	1800	1.9E+5	6.9E+2	7.6E+4	2.9E+2	1.1E+2	1.1E+0	4.2E+1	4.8E-1	3.7E+1	4.0E-1	4.2E+3	4.7E+1	**8.9E-1**	4.4E-2
9	20-R転-9	CaF2	0.711	1800	1.4E+5	5.6E+2	4.9E+4	2.3E+2	7.8E+1	9.4E-1	2.7E+1	3.8E-1	2.7E+1	3.3E-1	2.7E+3	3.7E+1	**1.0E+0**	5.2E-2
10	20-R転-10	CaF2	0.711	1800	1.9E+5	6.9E+2	7.1E+4	2.8E+2	1.1E+2	1.2E+0	4.0E+1	4.6E-1	3.7E+1	4.0E-1	3.9E+3	4.5E+1	**9.4E-1**	4.6E-2

Uranium Enrichment Measurement Spread Sheet ($14 : alumina-FM)

No.	sample No.	Sample Description	Decl. Enrich (%)	meas. time (sec)	a)186keV (cts)	Err (1sigma)	b)1001keV (cts)	Err (1sigma)	a)186keV (cps)	Err (3sigma)	b)1001keV (cps)	Err (3sigma)	a)U235 (gU)	Err (3sigma)	b)U238 (gU)	Err (3sigma)	**Calcul. Enrich (%)**	Calcul. Enrich Err
1	ALUF4-N-011	alumina	0.711	3600	4.7E+5	7.8E+2	2.9E+5	5.7E+2	1.3E+2	6.5E-1	8.0E+1	4.7E-1	5.3E+1	2.6E-1	8.5E+3	5.0E+1	**6.2E-1**	2.4E-2
2	ALUF4-N-012	alumina	0.711	1800	2.2E+5	5.2E+2	1.3E+5	3.7E+2	1.2E+2	8.7E-1	7.0E+1	6.2E-1	4.9E+1	3.5E-1	7.4E+3	6.6E+1	**6.5E-1**	2.9E-2
3	ALUF4-N-013	alumina	0.711	1800	1.8E+5	6.7E+2	9.1E+4	3.1E+2	9.8E+1	1.1E+0	5.1E+1	5.2E-1	4.0E+1	4.5E-1	5.4E+3	5.6E+1	**7.3E-1**	3.6E-2
4	ALUF4-N-014	alumina	0.711	1800	1.8E+5	6.7E+2	9.1E+4	3.1E+2	9.8E+1	1.1E+0	5.1E+1	5.2E-1	4.0E+1	4.5E-1	5.4E+3	5.5E+1	**7.3E-1**	3.6E-2
5	ALUF4-N-015	alumina	0.711	1800	2.4E+5	5.6E+2	1.4E+5	4.0E+2	1.3E+2	9.3E-1	7.9E+1	6.7E-1	5.4E+1	3.8E-1	8.4E+3	7.1E+1	**6.3E-1**	2.7E-2
6	ALUF4-N-016	alumina	0.711	1800	2.5E+5	5.8E+2	1.7E+5	4.4E+2	1.4E+2	9.6E-1	9.6E+1	7.4E-1	5.5E+1	3.9E-1	1.0E+4	7.9E+1	**5.4E-1**	2.3E-2
7	ALUF4-N-017	alumina	0.711	1800	2.2E+5	5.3E+2	1.3E+5	3.7E+2	1.2E+2	8.8E-1	7.1E+1	6.2E-1	4.9E+1	3.6E-1	7.5E+3	6.6E+1	**6.6E-1**	2.9E-2
8	ALUF4-N-018	alumina	0.711	1800	9.1E+4	4.8E+2	3.5E+4	1.9E+2	5.1E+1	8.0E-1	2.0E+1	3.2E-1	2.1E+1	3.2E-1	2.1E+3	3.4E+1	**9.8E-1**	5.6E-2
9	ALUF4-N-019	alumina	0.711	1800	2.2E+5	5.3E+2	1.4E+5	3.9E+2	1.2E+2	8.8E-1	7.7E+1	6.6E-1	4.9E+1	3.6E-1	8.2E+3	7.0E+1	**6.0E-1**	2.6E-2
10	ALUF4-N-020	alumina	0.711	1800	2.1E+5	5.3E+2	1.2E+5	3.7E+2	1.1E+2	8.8E-1	6.9E+1	6.2E-1	4.6E+1	3.6E-1	7.3E+3	6.5E+1	**6.3E-1**	2.8E-2

Appendix-C　Correlations between Uranium Mass and Surface Dose-rate

It is commonly important and useful that purchasing the surface dose-rate data of actual waste drums.　Those data may assume roughly the uranium mass of actual waste drums.　We had previously checked the surface dose-rate data before determination uranium mass for almost all actual waste drums.　Those works had verified that the uranium mass and the surface dose-rate data were proportional among the same group, however the proportional coefficient were not uniform but showed the particular values followed by each group.　The features of the dependency on the matrices and source spectrum are described in the followings.

Typical correlations the evaluated uranium mass data and the surface dose-rate data are summarized in **Figure-A1**.　The proportional coefficient of regression line are added shown in each graph, those values range from 0.1 to 1.0 approximately.　It is inferable that those are ascribable to the gamma ray energy spectrum, i.e. to the wastes drums in URCP so much impurity nuclides were adhered, for example, thorium progenies nuclide generated from U-232 which was inevitable as long as reprocessed uranium.　These facts are suggested to be not used for the determination uranium mass from the surface dose-rate data, but used for the discrimination method among natural uranium or reprocessed uranium.

Fortunately the existence of gamma emitter nuclides was largely unaffected in neutron passive assay compared with in gamma passive assay.

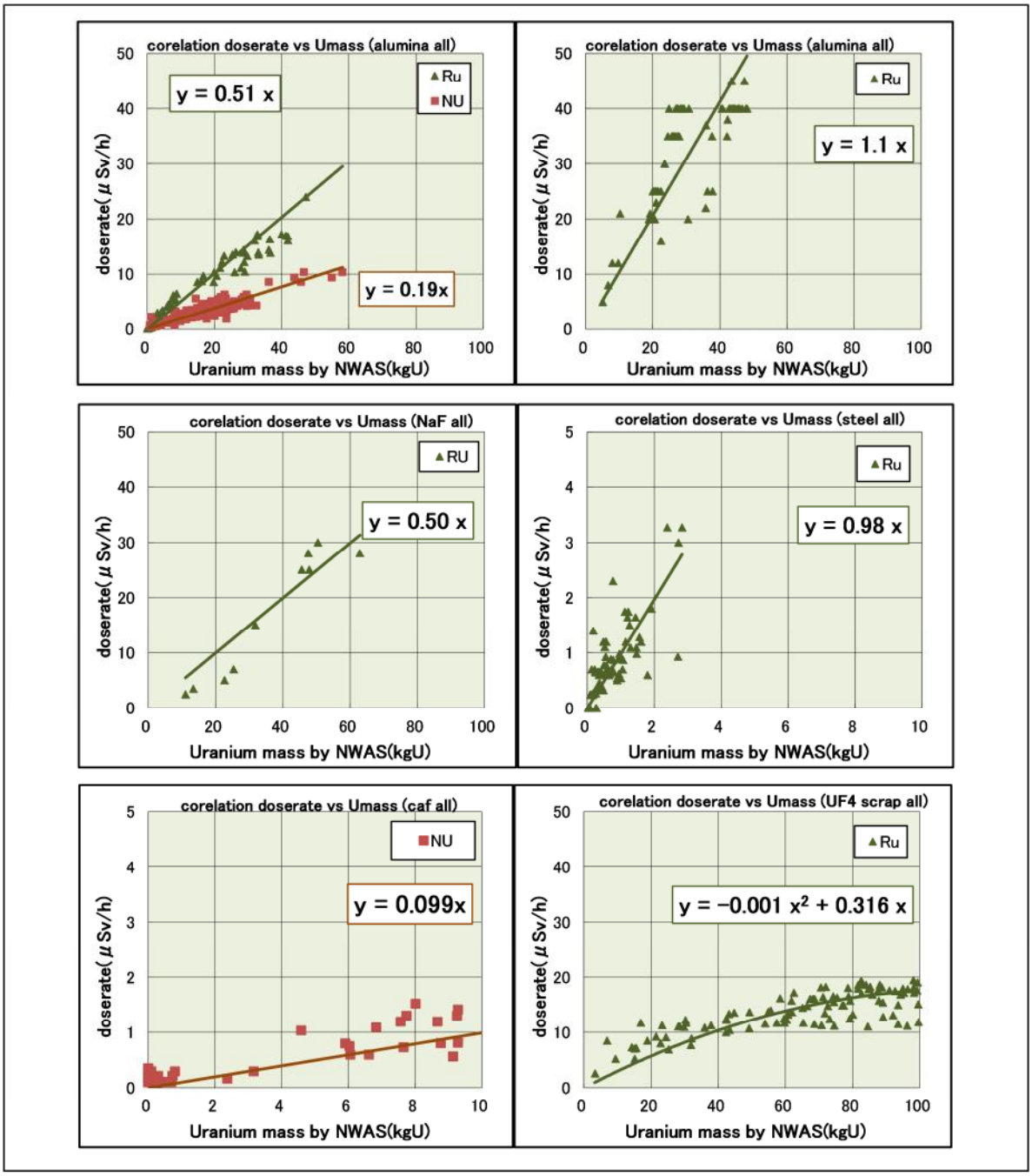

Figure-A1　Dose-rate Dependence on the Evaluated Uranium Mass

Appendix-D The Perspectives for Active Neutron Assay

On the basis of know-how for the passive neutron assay system, the active neutron assay system are now under investigating as the next step. The adopted method are the FNDI (Fast Neutron Direct Interrogation Method) among Active Assay System, which was distinguished from others feature based on different neutron thermalization. [Ref.-8]
Basic technical methodology are :

14 MeV neutron generator used (d,T) reaction (1e+8 neutrons/sec emissions)
Nuclear fission of U-235 induced from thermalized neutrons by matrices itself
Fast neutron detection by 14 pieces of Helium-3 proportional counters embedded in cadmium banks (same as passive assay)
Thermal neutron capture banks designed by cadmium covers
One piece of Helium-3 proportional counter as an additional detector embedded out of cadmium banks as neutron monitor
Effective reflectors inside 50cm of surrounding concrete wall

Advantages of active neutron assay are :

High neutron response, less dependence for uranium chemical composition
Expected flat distributed detection by neutron moderation by waste matrix itself
Capable to high accuracy analysis applied “die-away analysis”

The goal of the FNDI technically are :

Quick and high sensitive neutron detection
Fine evaluation with less dependence on matrix materials or uranium chemical composition difficult

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国際単位系（SI）

表１．SI 基本単位

基本量	SI 基本単位	
	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd

表２．基本単位を用いて表されるSI組立単位の例

組立量	SI 基本単位	
	名称	記号
面積	平方メートル	m^2
体積	立法メートル	m^3
速さ，速度	メートル毎秒	m/s
加速度	メートル毎秒毎秒	m/s^2
波数	毎メートル	m^{-1}
密度，質量密度	キログラム毎立方メートル	kg/m^3
面積密度	キログラム毎平方メートル	kg/m^2
比体積	立方メートル毎キログラム	m^3/kg
電流密度	アンペア毎平方メートル	A/m^2
磁界の強さ	アンペア毎メートル	A/m
量濃度(a)，濃度	モル毎立方メートル	mol/m^3
質量濃度	キログラム毎立法メートル	kg/m^3
輝度	カンデラ毎平方メートル	cd/m^2
屈折率(b)	（数字の） 1	1
比透磁率(b)	（数字の） 1	1

(a) 量濃度（amount concentration）は臨床化学の分野では物質濃度（substance concentration）ともよばれる。
(b) これらは無次元量あるいは次元１をもつ量であるが、そのことを表す単位記号である数字の１は通常は表記しない。

表３．固有の名称と記号で表されるSI組立単位

組立量	SI 組立単位			
	名称	記号	他のSI単位による表し方	SI基本単位による表し方
平面角	ラジアン(b)	rad	1 (b)	m/m
立体角	ステラジアン(b)	sr(c)	1 (b)	m^2/m^2
周波数	ヘルツ(d)	Hz		s^{-1}
力	ニュートン	N		m kg s^{-2}
圧力，応力	パスカル	Pa	N/m^2	m^{-1} kg s^{-2}
エネルギー，仕事，熱量	ジュール	J	N m	m^2 kg s^{-2}
仕事率，工率，放射束	ワット	W	J/s	m^2 kg s^{-3}
電荷，電気量	クーロン	C		s A
電位差（電圧），起電力	ボルト	V	W/A	m^2 kg s^{-3} A^{-1}
静電容量	ファラド	F	C/V	m^{-2} kg^{-1} s^4 A^2
電気抵抗	オーム	Ω	V/A	m^2 kg s^{-3} A^{-2}
コンダクタンス	ジーメンス	S	A/V	m^{-2} kg^{-1} s^3 A^2
磁束	ウエーバ	Wb	Vs	m^2 kg s^{-2} A^{-1}
磁束密度	テスラ	T	Wb/m^2	kg s^{-2} A^{-1}
インダクタンス	ヘンリー	H	Wb/A	m^2 kg s^{-2} A^{-2}
セルシウス温度	セルシウス度(e)	℃		K
光束	ルーメン	lm	cd sr(c)	cd
照度	ルクス	lx	lm/m^2	m^{-2} cd
放射性核種の放射能(f)	ベクレル(d)	Bq		s^{-1}
吸収線量, 比エネルギー分与, カーマ	グレイ	Gy	J/kg	m^2 s^{-2}
線量当量, 周辺線量当量, 方向性線量当量, 個人線量当量	シーベルト(g)	Sv	J/kg	m^2 s^{-2}
酸素活性	カタール	kat		s^{-1} mol

(a)SI接頭語は固有の名称と記号を持つ組立単位と組み合わせても使用できる。しかし接頭語を付した単位はもはやコヒーレントではない。
(b)ラジアンとステラジアンは数字の１に対する単位の特別な名称で、量についての情報をつたえるために使われる。実際には、使用する時には記号radおよびsrが用いられるが、習慣として組立単位としての記号である数字の１は明示されない。
(c)測光学ではステラジアンという名称と記号srを単位の表し方の中に、そのまま維持している。
(d)ヘルツは周期現象についてのみ、ベクレルは放射性核種の統計的過程についてのみ使用される。
(e)セルシウス度はケルビンの特別な名称で、セルシウス温度を表すために使用される。セルシウス度とケルビンの単位の大きさは同一である。したがって、温度差や温度間隔を表す数値はどちらの単位で表しても同じである。
(f)放射性核種の放射能（activity referred to a radionuclide）は、しばしば誤った用語で"radioactivity"と記される。
(g)単位シーベルト（PV,2002,70,205）についてはCIPM勧告2（CI-2002）を参照。

表４．単位の中に固有の名称と記号を含むSI組立単位の例

組立量	SI 組立単位		
	名称	記号	SI 基本単位による表し方
粘度	パスカル秒	Pa s	m^{-1} kg s^{-1}
力のモーメント	ニュートンメートル	N m	m^2 kg s^{-2}
表面張力	ニュートン毎メートル	N/m	kg s^{-2}
角速度	ラジアン毎秒	rad/s	m m^{-1} s^{-1}=s^{-1}
角加速度	ラジアン毎秒毎秒	rad/s^2	m m^{-1} s^{-2}=s^{-2}
熱流密度，放射照度	ワット毎平方メートル	W/m^2	kg s^{-3}
熱容量，エントロピー	ジュール毎ケルビン	J/K	m^2 kg s^{-2} K^{-1}
比熱容量，比エントロピー	ジュール毎キログラム毎ケルビン	J/(kg K)	m^2 s^{-2} K^{-1}
比エネルギー	ジュール毎キログラム	J/kg	m^2 s^{-2}
熱伝導率	ワット毎メートル毎ケルビン	W/(m K)	m kg s^{-3} K^{-1}
体積エネルギー	ジュール毎立方メートル	J/m^3	m^{-1} kg s^{-2}
電界の強さ	ボルト毎メートル	V/m	m kg s^{-3} A^{-1}
電荷密度	クーロン毎立方メートル	C/m^3	m^{-3} sA
表面電荷	クーロン毎平方メートル	C/m^2	m^{-2} sA
電束密度，電気変位	クーロン毎平方メートル	C/m^2	m^{-2} sA
誘電率	ファラド毎メートル	F/m	m^{-3} kg^{-1} s^4 A^2
透磁率	ヘンリー毎メートル	H/m	m kg s^{-2} A^{-2}
モルエネルギー	ジュール毎モル	J/mol	m^2 kg s^{-2} mol^{-1}
モルエントロピー, モル熱容量	ジュール毎モル毎ケルビン	J/(mol K)	m^2 kg s^{-2} K^{-1} mol^{-1}
照射線量（X線及びγ線）	クーロン毎キログラム	C/kg	kg^{-1} sA
吸収線量率	グレイ毎秒	Gy/s	m^2 s^{-3}
放射強度	ワット毎ステラジアン	W/sr	m^4 m^{-2} kg s^{-3}=m^2 kg s^{-3}
放射輝度	ワット毎平方メートル毎ステラジアン	$W/(m^2$ sr)	m^2 m^{-2} kg s^{-3}=kg s^{-3}
酵素活性濃度	カタール毎立方メートル	kat/m^3	m^{-3} s^{-1} mol

表５．SI 接頭語

乗数	接頭語	記号	乗数	接頭語	記号
10^{24}	ヨタ	Y	10^{-1}	デシ	d
10^{21}	ゼタ	Z	10^{-2}	センチ	c
10^{18}	エクサ	E	10^{-3}	ミリ	m
10^{15}	ペタ	P	10^{-6}	マイクロ	μ
10^{12}	テラ	T	10^{-9}	ナノ	n
10^9	ギガ	G	10^{-12}	ピコ	p
10^6	メガ	M	10^{-15}	フェムト	f
10^3	キロ	k	10^{-18}	アト	a
10^2	ヘクト	h	10^{-21}	ゼプト	z
10^1	デカ	da	10^{-24}	ヨクト	y

表６．SIに属さないが、SIと併用される単位

名称	記号	SI 単位による値
分	min	1 min=60s
時	h	1h =60 min=3600 s
日	d	1 d=24 h=86 400 s
度	°	1°=(π/180) rad
分	’	1’=(1/60)°=(π/10800) rad
秒	”	1”=(1/60)’=(π/648000) rad
ヘクタール	ha	$1ha=1hm^2=10^4m^2$
リットル	L, l	$1L=11=1dm^3=10^3cm^3=10^{-3}m^3$
トン	t	$1t=10^3$ kg

表７．SIに属さないが、SIと併用される単位で、SI単位で表される数値が実験的に得られるもの

名称	記号	SI 単位で表される数値
電子ボルト	eV	$1eV=1.602\ 176\ 53(14)\times10^{-19}J$
ダルトン	Da	$1Da=1.660\ 538\ 86(28)\times10^{-27}kg$
統一原子質量単位	u	1u=1 Da
天文単位	ua	$1ua=1.495\ 978\ 706\ 91(6)\times10^{11}m$

表８．SIに属さないが、SIと併用されるその他の単位

名称	記号	SI 単位で表される数値
バール	bar	1 bar=0.1MPa=100kPa=10^5Pa
水銀柱ミリメートル	mmHg	1mmHg=133.322Pa
オングストローム	Å	1 Å=0.1nm=100pm=10^{-10}m
海里	M	1 M=1852m
バーン	b	$1\ b=100fm^2=(10^{-12}cm)2=10^{-28}m^2$
ノット	kn	1 kn=(1852/3600)m/s
ネーパ	Np	SI単位との数値的な関係は、対数量の定義に依存。
ベル	B	
デジベル	dB	

表９．固有の名称をもつCGS組立単位

名称	記号	SI 単位で表される数値
エルグ	erg	$1\ erg=10^{-7}$ J
ダイン	dyn	$1\ dyn=10^{-5}$N
ポアズ	P	1 P=1 dyn s cm^{-2}=0.1Pa s
ストークス	St	1 St =$1cm^2$ s^{-1}=$10^{-4}m^2$ s^{-1}
スチルブ	sb	1 sb =1cd cm^{-2}=10^4cd m^{-2}
フォト	ph	1 ph=1cd sr cm^{-2} 10^4lx
ガル	Gal	1 Gal =1cm s^{-2}=$10^{-2}ms^{-2}$
マクスウェル	Mx	1 Mx = 1G cm^2=10^{-8}Wb
ガウス	G	1 G =1Mx cm^{-2} =10^{-4}T
エルステッド(c)	Oe	1 Oe≙ $(10^3/4\pi)$A m^{-1}

(c) ３元系のCGS単位系とSIでは直接比較できないため、等号「 ≙ 」は対応関係を示すものである。

表10．SIに属さないその他の単位の例

名称	記号	SI 単位で表される数値
キュリー	Ci	$1\ Ci=3.7\times10^{10}Bq$
レントゲン	R	$1\ R = 2.58\times10^{-4}C/kg$
ラド	rad	$1\ rad=1cGy=10^{-2}Gy$
レム	rem	$1\ rem=1\ cSv=10^{-2}Sv$
ガンマ	γ	1γ=1 nT=10-9T
フェルミ		1フェルミ=1 fm=10-15m
メートル系カラット		1メートル系カラット = 200 mg = 2×10-4kg
トル	Torr	1 Torr = (101 325/760) Pa
標準大気圧	atm	1 atm = 101 325 Pa
カロリー	cal	1cal=4.1858J（「15℃」カロリー），4.1868J（「IT」カロリー）4.184J（「熱化学」カロリー）
ミクロン	μ	1 μ =1μm=10^{-6}m

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