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A Spent PWR Fuel Combination Method for Optimizing DUPIC Fuel Composition

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Abstract

A combination method of spent pressurized water reactor (PWR) fuel was proposed, which adjusts the fuel composition for direct use of spent PWR fuel in Canada deuterium uranium (CANDU) reactors (DUPIC). This method reduces the composition heterogeneity (variation) of the DUPIC fuel caused by directly reusing spent PWR fuel as DUPIC fuel feedstock. In this study, a combination method was used to find the optimum mixture composition from the spent PWR fuel database by minimizing the composition variation of major fissile isotopes ^{235}U and ^{239}Pu. The simulation results have shown that the combination method can reduce the composition variation of ^{235}U and ^{239}Pu to 0.11% and 1.40%, respectively, through assembly-wise mixing operation only. It is also believed that the result could be improved further through rod-wise combination technique, if isotopic composition of each spent PWR fuel rod is known by direct measurement during the DUPIC fuel fabrication process.

1. Introduction

The pressurized water reactor (PWR) fuel contains 3.5 4.4 wt% ^{235}U initially. When the PWR fuel is discharged, total fissile content is approximately 1.5 wt% including unburnt ^{235}U and newly created ^{239}Pu and ^{241}Pu. Such a fissile content is twice that of natural uranium and, therefore, more than enough to be burnt again in a Canada deuterium uranium (CANDU) reactor, which was originally designed for natural uranium fuel. However, in order to accommodate high excess reactivity of spent PWR fuel in a CANDU reactor, it is recommended to reduce the number of fuel bundles loaded per refueling operation (e.g., 2-bundle shift refueling scheme) so that the maximum channel and bundle powers are kept lower than license limits.[1] On the other hand, in the direct use of spent PWR fuel in CANDU reactors (DUPIC), the DUPIC fuel composition changes depending on initial enrichment, discharge burnup, and specific power of the PWR fuel. If the DUPIC fuel is loaded in a CANDU reactor without any adjustment on fuel composition, it is expected that the uncertainties of the core performance parameters increase, which eventually reduces the operational margin of a DUPIC-fueled CANDU core.

Therefore two approaches to resolve the fuel composition heterogeneity have been proposed, which are fissile content adjustment and reactivity control method.[2,3] In these composition adjustment methods, the content of important isotopes or reactivity of the fuel lattice is tightly controlled by adding slightly enriched uranium (SEU) and/or depleted uranium (DU) to the spent PWR fuel mixture during DUPIC fuel fabrication process. Therefore it was known that the DUPIC fuel fabrication cost increases compared to the case that does not use SEU for DUPIC fuel fabrication.

This study examines the possibility of mixing only spent PWR fuels without using slightly enriched uranium to achieve the reference DUPIC fuel composition. In fact the diverse isotopic composition of spent PWR fuel can also be used as source material for fuel composition adjustment. In other words, the reference fissile content, for example, can be obtained by mixing spent PWR fuel assemblies of low and high fissile content. In this study, we have generated database for spent PWR fuels in Korea and defined the reference DUPIC fuel composition. Then a combination method, which is one of the optimization techniques, was developed and applied to find the optimum combination of spent PWR fuel assemblies that satisfies the reference DUPIC fuel composition.

A DUPIC fuel process is described in Sec. 2 and a combination method is presented in Sec. 3. In Sec. 4, the proposed optimal combination method is tested by using a total of 3598 spent fuel assemblies collected from PWR plants in Korea up to 1996 and test results are compared. Finally, Sec. 5 summarizes and concludes the paper.

2. DUPIC Fuel Process

II.A Spent PWR Fuel Characteristics

The DUPIC fuel is made of spent PWR fuel material and, therefore, the isotopic composition data of spent PWR fuels, generated for Younggwang nuclear power plants (NPPs) 1 and 2, Kori NPPs 3 and 4, and Uljin NPPs 1 and 2 up to year 1996, were collected. The total number of spent PWR fuel assemblies included in this database is 3598, which was processed by Microsoft (MS) Access Software as shown in Fig. 1. Then the distribution of each isotopic composition was analyzed, which is shown in Table 1 in terms of statistical variables. In Table 1, the statistical variables are given for ten isotopes which have strong effects on the reactivity of the DUPIC fuel lattice from the nuclear physics viewpoint. Compared to the fissile content of natural uranium (0.71 wt%), the average fissile content of the spent PWR fuel (0.91 wt% of ^{235}U, 0.54 wt% of ^{239}Pu and 0.05 wt% of ^{241}Pu) is much higher, which can provide high excess reactivity to the reactor system. Table 1 also shows that the variations (variance) of ^{235}U and ^{239}Pu content are relatively higher compared with those of other isotopes, which are 0.1063 and 0.0031, respectively, which inform that the variance of ^{235}U is much higher than that of ^{239}Pu. Figure 2 shows the content distribution of two major fissile isotopes in 3598 spent PWR fuel assemblies.

II.B Physical Modeling

A commercial DUPIC fuel fabrication facility has been conceptually designed by Korea Atomic Energy Research Institute.[4] In the conceptual design study, a spent fuel transfer cask is used in the main process building to retrieve each fuel assembly from the shipping cask. The as-received spent PWR fuels are inspected and stored in the storage vault. Then an appropriate number of spent fuels are selected and transported to the process hot cell. In the process hot cell, the fuel material is recovered from the spent PWR fuel through disassembling and decladding by thermal and mechanical process. The powder preparation, called OREOX (Oxidation REduction of OXide fuel), is the key process that produces resinterable powder feedstock from the spent PWR fuel. Once the resinterable powder is prepared, the fuel pellet and rod are manufactured following processes which are almost the same as those used for conventional pellet/rod production of oxide fuel. During the fuel fabrication process, the fuel composition homogeneity should be retained to satisfy constraints of fuel and reactor performance. So far two approaches have been considered to keep the uniform neutronic performance of the DUPIC fuel, the conventional method and optimum combination technique, which are described as follows:

- Conventional Method

In the conventional method, spent PWR fuels are classified by their fissile contents (characterized and evaluated by the fuel design data) and burnup characteristics, and stored in the designated area. Then the spent fuel assembly is dismantled and one batch (~ 400 kgHM) of fuel material is taken for the subsequent processes. The spent fuel pellet fragments and their debris go through three OREOX cycles to get a powder form of suitable characteristics for fuel fabrication. The resulting fuel powder is sampled and assayed for size distribution and fissile content measurements. At this stage, the fuel composition is adjusted by either fissile content or reactivity control method, which has already been developed.[2, 3] In these methods, spent PWR fuel powder is blended with fresh uranium such as SEU and/or DU to satisfy the fuel composition requirements.

- Optimum Combination Technique

The combination technique, proposed in this study, can be considered in two physical stages (rod-wise and assembly-wise combinations) of feedstock preparation as depicted in Fig. 3. In this process, the transportation schedule will be set up early through the assembly-wise optimum combination described in Sec. III when spent fuels are stored in the reactor storage pool. In the DUPIC fuel fabrication facility, spent PWR fuels are received in a shipping cask, unloaded and stored in the storage vault following the transportation schedule. The capacity of the storage vault is 100 metric ton (MT) of spent PWR fuels, which are divided into five sectors as shown in Fig. 3. Then the spent PWR fuels are transferred to the main process hot cell by an overhead crane. In the hot cell, structural components in the top-end fitting are removed, and fuel rods are extracted and stored in a

temporary buffer storage after fuel composition is measured. Using the measurement data, rod-wise optimum combination is carried out to obtain uniform composition of DUPIC fuel. A batch of 400 kg is formed and transferred to the subsequent processes such as powder treatment by OREOX and sintering processes.

3. An Optimal Combination Method

The direct refabrication of spent PWR fuel to CANDU fuels results in heterogeneous fuel composition, depending on the enrichment, irradiation history, and discharge burnup of PWR fuels, which induces the reduction of the operational margin of DUPIC-fueled CANDU core. Therefore, differently from the previous methods[2,3] that resolve the heterogeneity problem but increase the fabrication cost due to uses of SEU or DU, a simple combination method was developed in this study to search for an optimum combination of only spent PWR fuel assemblies without using fresh uranium.

The objective of the optimization process is to group the spent PWR fuel assemblies such that the composition variability of the DUPIC fuel is minimized. Therefore a cost function for minimizing the composition variability can be written as follows:

$$J = \max\{\bar{x}_1, \bar{x}_2, \Lambda\ , \bar{x}_G\} - \min\{\bar{x}_1, \bar{x}_2, \Lambda\ , \bar{x}_G\}, \tag{1}$$

where

$$\bar{x}_j = \frac{\sum_{i=1}^{n} w_i y_{ij}}{\sum_{i=1}^{n} w_i}, \quad j = 1, 2, 3, ..., G \tag{2}$$

$\bar{x}_j$ = average value of x in group j,

w_i = importance of target isotope i,

y_{ij} = average content (wt%) of target isotope i in group j,

n = number of target isotopes, and

G = number of groups.

A combination method proceeds in two stages to minimize the composition variability of DUPIC fuels. In the first stage, an averaging technique is used as the simple grouping method to minimize the cost function. If the composition contents of several important isotopes of all spent PWR fuel assemblies are given, the spent PWR fuel assemblies can be sorted in an ascending order of composition for a specific target isotope. Here, the target isotope can be defined as either single isotope or importance-weighted average of several important isotopes. Then, as shown in Fig. 4, each assembly is selected and assigned to a specific fuel group, which was defined over a finite interval of the fuel assemblies sorted with the isotope composition value. Through this process, the group-averaged composition of the target isotope is substantially leveled and all groups have the similar mean values of composition for the target isotope. After the first stage, the fuel assemblies in each group are sorted in an ascending order of the group-wise average composition of the target isotope.

The combination process is continued in the second stage to accomplish more minute combination for minimizing the composition variability of DUPIC fuels. In the second stage, a spent fuel assembly of the highest composition value of the target isotope in the maximum group is exchanged with a spent fuel assembly of the lowest composition value of the target isotope in the minimum group. This process can minimize the difference of the average composition value of the target isotope between the maximum and minimum groups, which is the objective of the whole optimization process. The average composition value of the target isotope of all the groups is leveled by repeating the foregoing (second stage) process, which means that a spent fuel assembly of the highest composition value of the target isotope in the newly created maximum group is repeatedly exchanged with a spent fuel assembly of the lowest composition value of the target isotope in the newly created minimum

group.

However, if this process is conducted many times, sometimes this process may result in oscillations without reaching a minimal point (refer to Fig. 5). For example, after the fuel assemblies are exchanged between the maximum and minimum groups, the previous minimum group can be a new maximum group and the previous maximum group can become a new minimum group. Then the difference of the isotope composition between the new maximum and minimum groups may not be reduced continuously and rather increase even if the optimization step proceeds. In order to prevent such an oscillation, if that situation takes place, it is generally required to reduce the momentum effect. This is accomplished by exchanging a spent fuel assembly of less higher target value in the maximum group with a spent fuel assembly of less lower target value in the minimum group as the optimization step proceeds (refer to Fig. 6). This second stage is repeated until a specific cost function value will be accomplished. The overall fuel combination procedure including the first and second stages is shown in Fig. 7.

4. Application to Fuel Composition Adjustment

In the optimal combination of spent PWR fuel assemblies, a total of 3598 spent fuel assemblies collected from PWR plants in Korea up to 1996, were used. Table 2 shows the importance of each isotope used for cost function calculation. The target weight content of each isotope is an average of isotopic contents calculated from 3598 fuel assemblies and the importance was determined from the sensitivity of reactivity to the isotopic composition. Table 2 also shows that ^{235}U and ^{239}Pu are the most important isotopes from the viewpoint of nuclear physics design when the fuel composition is adjusted.

The proposed combination method was applied to four cases to find the best option for determining target variable:

Case 1) using only ^{239}Pu as a target isotope,

Case 2) using only ^{235}U as a target isotope,

Case 3) using both ^{235}U and ^{239}Pu as target isotopes, and

Case 4) using periodic change between ^{235}U and ^{239}Pu as target isotopes as the optimization step continues.

Since the average weight of a spent fuel assembly is ~430 kg and the capacity of the hot cell is ~20 MT uranium, the 3598 fuel assemblies were divided into 80 groups. Therefore, each group consists of 44 or 45 fuel assemblies.

Figure 8 shows that the difference of the target isotopic composition between the maximum and minimum groups and the standard deviation decrease as the optimization step continues. This is the result of the case using ^{239}Pu as a target isotope. Figure 9 shows the weight content of important isotopes for the case using only ^{239}Pu as the target isotope (Case 1). The ^{239}Pu weight contents of 80 different groups remain in a horizontal line, which means that all groups have almost the same weight content. The maximum difference of the ^{239}Pu weight content is 1.11×10^{-6} at the most (refer to Table 3) and its relative error (two standard deviations) is 0.012%. However, the ^{235}U content and the importance-weighted average content of ^{235}U and ^{239}Pu are distributed sparsely because the objective of this case is to minimize only the ^{239}Pu weight content difference between the maximum and minimum groups.

Figure 10 shows the weight content of important isotopes for the case using only ^{235}U as the target isotope (Case 2). The maximum difference of the ^{235}U weight content is 1.67×10^{-5} and its relative error is 0.12% (refer to Table 3). The ^{235}U weight content difference between the maximum and minimum groups is much less than that of the first case but the ^{239}Pu weight content difference is much greater than that of the first case. Figure 11 shows the weight content of important isotopes for the case using importance-weighted average of ^{235}U and ^{239}Pu as target isotopes (Case 3). The result shows an intermediate behavior of Cases 1 and 2. Figure 12 shows the weight content of important isotopes for the case using periodic change between ^{235}U and ^{239}Pu as target isotopes (Case 4). In this case, the relative errors of ^{239}Pu and ^{235}U are 1.40% and 0.11%, respectively. Considering the results of the weight content variation of both important isotopes ^{235}U and ^{239}Pu, it is determined that Case 4 is the best. In all these four cases, all groups have similar weight as shown in Table 3. Figures 13 and 14 show the histograms of ^{239}Pu and ^{235}U weight contents for Case 4, respectively. The distribution of ^{239}Pu content is close to

the nominal distribution function, while the distribution of ^{235}U content is very uniform.

The sensitivity of composition variability to the capacity change of a hot cell was also analyzed as given in Table 4 for the case using periodic change between ^{235}U and ^{239}Pu as target isotopes (Case 4), which is known to be the best. The result shows that the weight content difference between the maximum and minimum groups decreases as well as the relative errors of ^{235}U and ^{239}Pu weight content variation as the hot cell capacity increases (the group number decreases), which is due to the physical fact that the more homogeneity is attainable when the batch size increases.

The proposed combination method was applied to the assembly-wise combination only. However, if there are rod-wise measurement data in the second stage of the DUPIC fuel process, it is expected that the proposed combination method can significantly reduce the variations of the DUPIC fuel composition by being applied to the rod-wise combination in the same way as the assembly-wise combination.

If the proposed combination method instead of the conventional method described in Sec. II is taken into service in the DUPIC fuel fabrication facility, two benefits can be achieved. At first it is possible to fully utilize the spent PWR fuels generated in Korea, contrary to the conventional method that produces additional spent fuel (even though the amount of extra spent fuel is less than 20% of spent PWR fuel used for DUPIC fuel fabrication) by using fresh uranium. Secondly, because no fresh uranium is needed, the DUPIC fuel fabrication cost decreases. Previous study[5] has shown that the fabrication unit costs with and without using fresh uranium are 654 and 616 $/kgHM, respectively. Therefore, a unit fabrication cost saving of 6% can be achieved through this combination method.

5. Summary and Conclusion

In this work, a combination method of spent PWR fuel has been developed and applied to the minimizing process of the DUPIC fuel composition variability. The method proceeds in two stages. The grouping method of the first stage is to use a simple averaging technique that after the spent PWR fuel assemblies are sorted in an ascending order of composition for a specific target isotope, the sorted fuel assemblies over a finite interval are assigned to each group. In a second stage, a spent fuel assembly of the highest composition value of the target isotope in the maximum group is exchanged with a spent fuel assembly of the lowest composition value of the target isotope in the minimum group. Also, as optimization step continues, a momentum effect is decreased to avoid continuous oscillations (if any) without reaching an optimal point.

The proposed combination method was applied to four cases according to target isotopes: i) ^{239}Pu, ii) ^{239}Pu, iii) ^{235}U and ^{239}Pu, and iv) periodic change between ^{235}U and ^{239}Pu. The comparison among four cases has shown that Case 4 has the best performance to minimize the composition variation of the major fissile isotopes ^{235}U and ^{239}Pu. The simulation results have shown that the combination method can reduce the composition variation of ^{235}U and ^{239}Pu to 0.11% and 1.40%, respectively, through assembly-wise mixing operation only. Therefore, it is concluded that the proposed method reduces the composition variation of the DUPIC fuel caused by directly reusing spent PWR fuel as CANDU fuel feedstock. In addition, the combination method can reduce the DUPIC fuel fabrication cost compared with conventional techniques proposed earlier.

In the future the acceptability of the composition variation achieved by the combination method should be confirmed by the reactor physics calculation. The composition variation results in uncertainty in power distribution, which will eventually reduces the operational margin of the reactor. Therefore this composition variability should be quantified in relation to the reactor trip set point. At the same time, both the conventional and proposed combination methods require a fissile content measurement system that can process the fuel material within a reasonable measurement time so that the whole fabrication process is not delayed

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Table 1. Results of statistical treatment of isotopic content in spent PWR fuels.

Items	^{235}U	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	^{241}Pu	^{103}Rh	^{143}Nd	^{151}Sm	^{155}Gd
Total mass (kg)	14019	8381.9	3267.04	790.16	1143.23	790.17	592.05	1132.17	19.21	16.36
Average content (wt%)	0.9103	0.5442	0.2121	0.0513	0.0742	0.0513	0.0384	0.0735	0.0012	0.0011
Standard error	0.0054	0.0009	0.0007	0.0003	0.0003	0.0003	0.0001	0.0003	0.0000	0.0000
Median	0.8460	0.5629	0.2200	0.0530	0.0788	0.0530	0.0408	0.0787	0.0013	0.0011
Standard deviation	0.3260	0.0558	0.0443	0.0171	0.0188	0.0171	0.0088	0.0155	0.0002	0.0004
Variance	1.1063	0.0031	0.0020	0.0003	0.0004	0.0003	0.0001	0.0002	0.0000	0.0000
Tortuosity	9.6098	1.1280	0.8727	-0.5898	0.7489	-0.5898	0.5148	0.6351	0.3713	-0.4373
Skewness	2.5700	-1.2796	-1.0871	-0.3725	-0.9973	-0.3725	-1.1214	-1.2415	-0.986	-0.1703
Minimum value	0.2070	0.2856	0.0336	0.0025	0.0074	0.0025	0.0077	0.0232	0.0006	0.0002
Maximum value	2.9082	0.6350	0.3208	0.0871	0.1324	0.0871	0.0531	0.0947	0.0016	0.0027

Table 2. Input data for optimum combination.

Element	Target content (wt%)	Importance	Element	Target content (wt%)	Importance
^{235}U	0.9103	0.2813	^{149}Sm	0.0003	0.0150
^{239}Pu	0.5440	0.2352	^{143}Nd	0.0735	0.0110
^{240}Pu	0.2120	0.0222	^{241}Am	0.0742	0.0107
^{241}Pu	0.0513	0.0370	^{151}Sm	0.0012	0.0061
^{155}Gd	0.0011	0.0159	^{103}Rh	0.0384	0.0045

Table 3. Comparison of four cases with different target isotopes (80-group model).

		Case 1	Case 2	Case 3	Case 4
^{239}Pu	Maximum content (wt%)	5.44607E-3*	5.63102E-3	5.55935E-3	5.54148E-3
	Minimum content (wt%)	5.44496E-3	5.26312E-3	5.33329E-3	5.33152E-3
	Standard deviation (wt%)	3.16782E-7	7.34586E-5	5.04057E-5	3.80209E-5
	Relative error (%)	1.16346E-2	2.69795E+0	1.85128E+0	1.39641E+0
^{235}U	Maximum content (wt%)	1.05634E-2	9.13859E-3	9.21567E-3	9.13949E-3
	Minimum content (wt%)	8.27067E-3	9.12188E-3	9.04087E-3	9.12201E-3
	Standard deviation (wt%)	4.39570E-4	5.47832E-6	4.10708E-5	5.10891E-6
	Relative error (%)	9.62868E+0	1.20001E-1	8.99645E-1	1.11910E-1
Importance-weighted average of ^{235}U + ^{239}Pu	Maximum content (wt%)	8.23276E-3	7.53658E-3	7.45719E-3	7.49241E-3
	Minimum content (wt%)	6.98410E-3	7.36826E-3	7.44746E-3	7.39614E-3
	Standard deviation (wt%)	2.39392E-4	3.39416E-5	3.64846E-6	1.76518E-5
	Relative error (%)	6.42453E+0	9.10888E-1	9.79134E-2	4.73719E-1
Weight	Maximum content (wt%)	1.95640E+7	1.96600E+7	1.95410E+7	1.95640E+7
	Minimum content (wt%)	1.88660E+7	1.87040E+7	1.86570E+7	1.88660E+7
	Standard deviation (wt%)	1.30412E+5	1.65268E+5	1.70543E+5	1.30412E+5
	Relative error (%)	1.35479E+0	1.71689E+0	1.77169E+0	1.35479E+0

* Read as 5.45039×10^{-3}

Table 4. Sensitivity calculation on hot cell capacity (Case 4).

		60 groups (~25 MTU)	80 groups (~20 MTU)	100 groups (~15 MTU)
^{239}Pu	Maximum content (wt%)	5.51533E-3*	5.54148E-3	5.56066E-3
	Minimum content (wt%)	5.34708E-3	5.33152E-3	5.31314E-3
	Standard deviation (wt%)	3.68174E-5	3.80209E-5	4.85025E-5
	Relative error (%)	1.35221E+0	1.39641E+0	1.78138E+0
^{235}U	Maximum content (wt%)	9.13529E-3	9.13949E-3	9.17608E-3
	Minimum content (wt%)	9.12570E-3	9.12201E-3	9.08541E-3
	Standard deviation (wt%)	3.18949E-6	5.10891E-6	1.13228E-5
	Relative error (%)	6.98651E-2	1.11910E-1	2.48022E-1
Importance-weighted average of ^{235}U + ^{239}Pu	Maximum content (wt%)	7.48642E-3	7.49241E-3	7.51090E-3
	Minimum content (wt%)	7.40768E-3	7.39614E-3	7.39026E-3
	Standard deviation (wt%)	1.71276E-5	1.76518E-5	2.30464E-5
	Relative error (%)	4.59653E-1	4.73719E-1	6.18494E-1
Weight	Maximum weight (g)	2.60380E+7	1.95640E+7	1.56510E+7
	Minimum weight (g)	2.52390E+7	1.88660E+7	1.50410E+7
	Standard deviation (wt%)	1.66356E+5	1.30412E+5	1.08039E+5
	Relative error (%)	1.29615E+0	1.35479E+0	1.40296E+0

* Read as 5.51533×10^{-3}

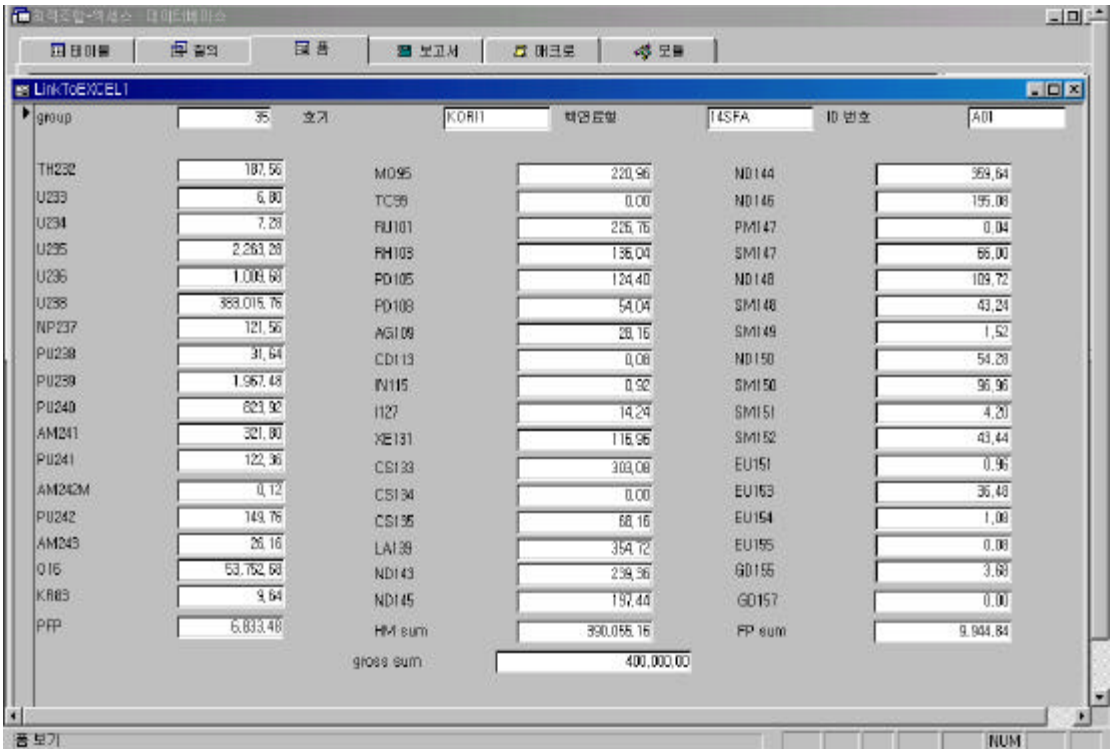
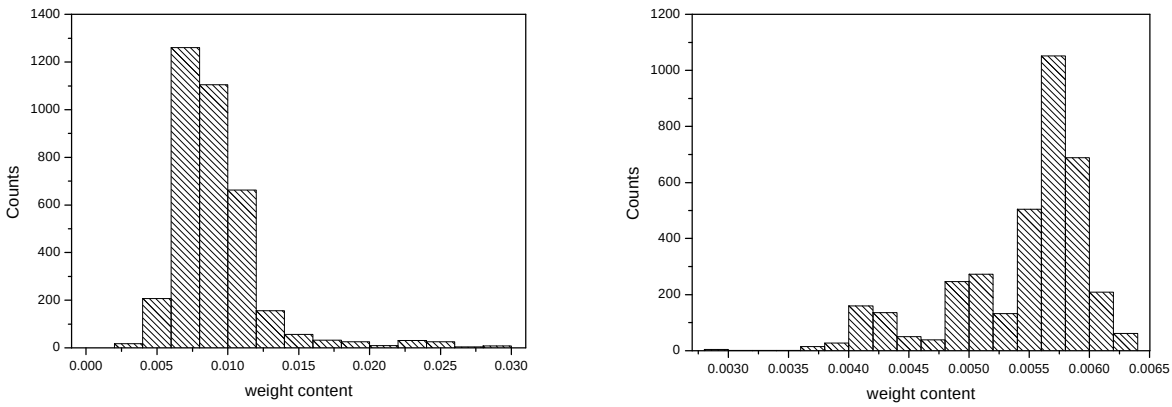


Fig. 1 Database form of spent PWR fuels.



(a) ^{239}Pu (b) ^{235}U

Fig. 2 Distribution of isotopic content for spent PWR fuels in Korea (3598 assemblies).

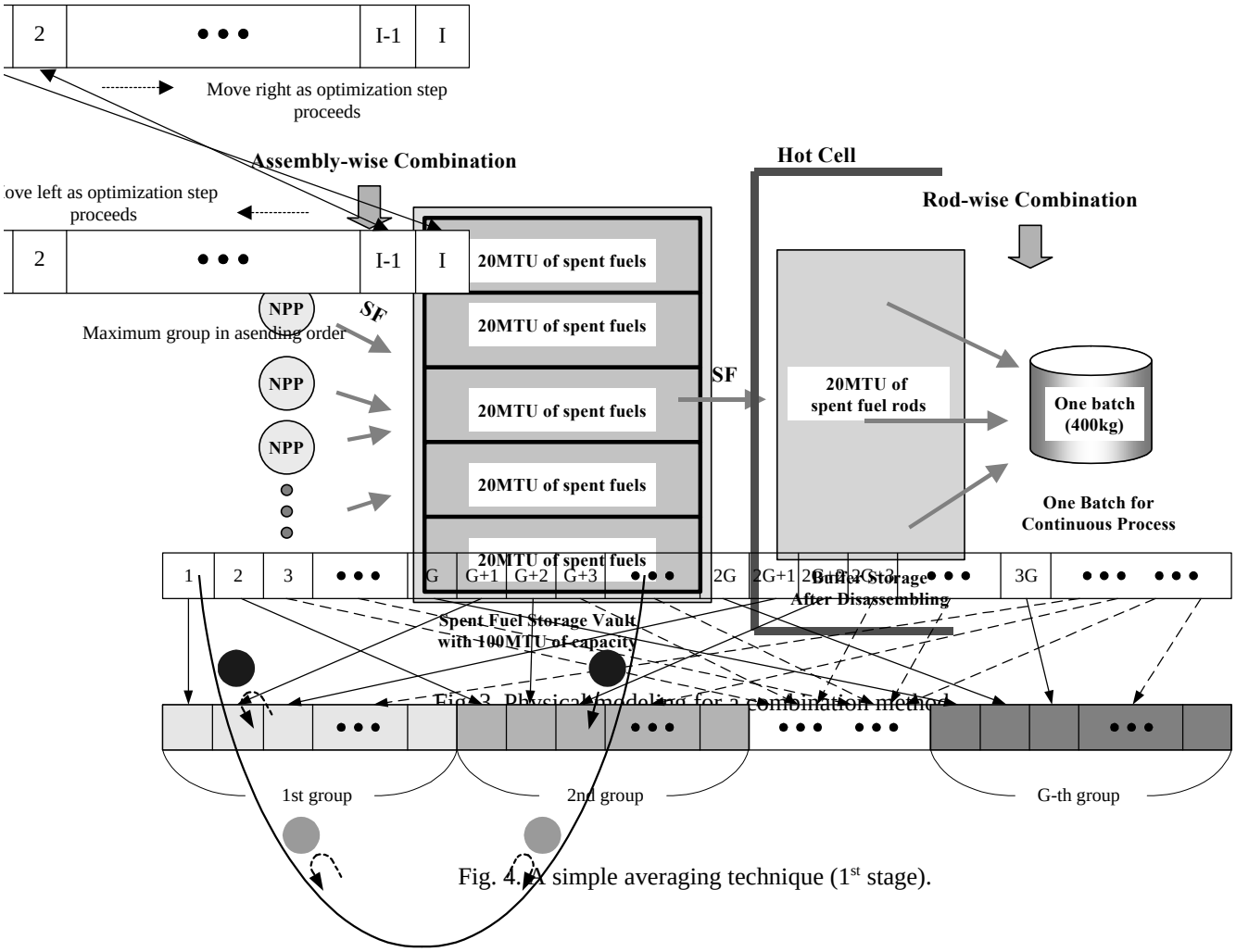


Fig. 4. A simple averaging technique (1st stage).

Fig. 5. Oscillations without reaching a minimal point.

Fig. 6. An exchange method for preventing continuous oscillations (2^{nd} stage).

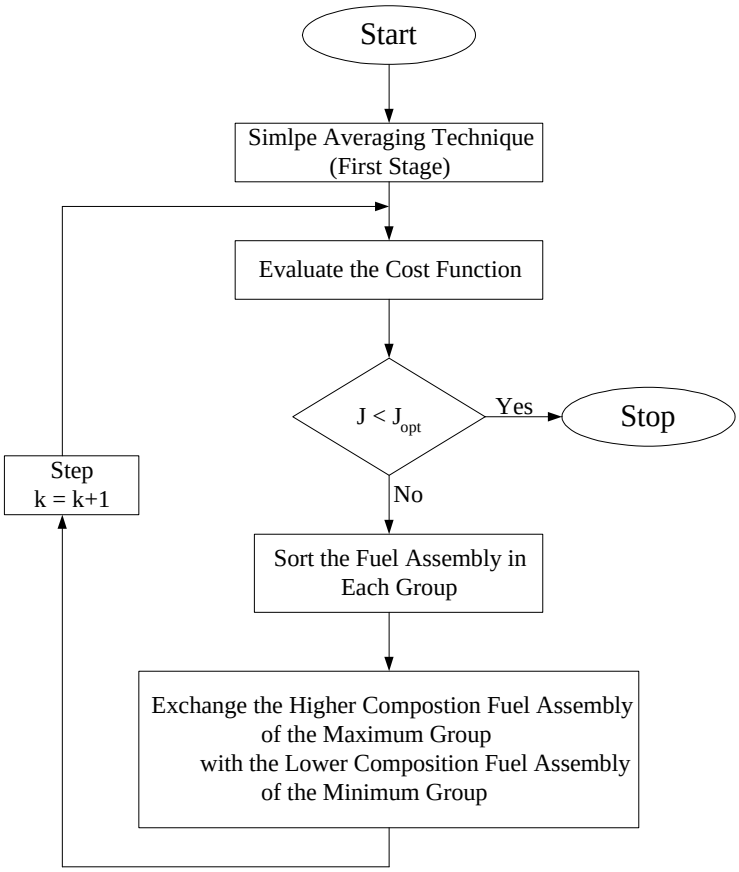


Fig. 7. Overall fuel combination procedure.

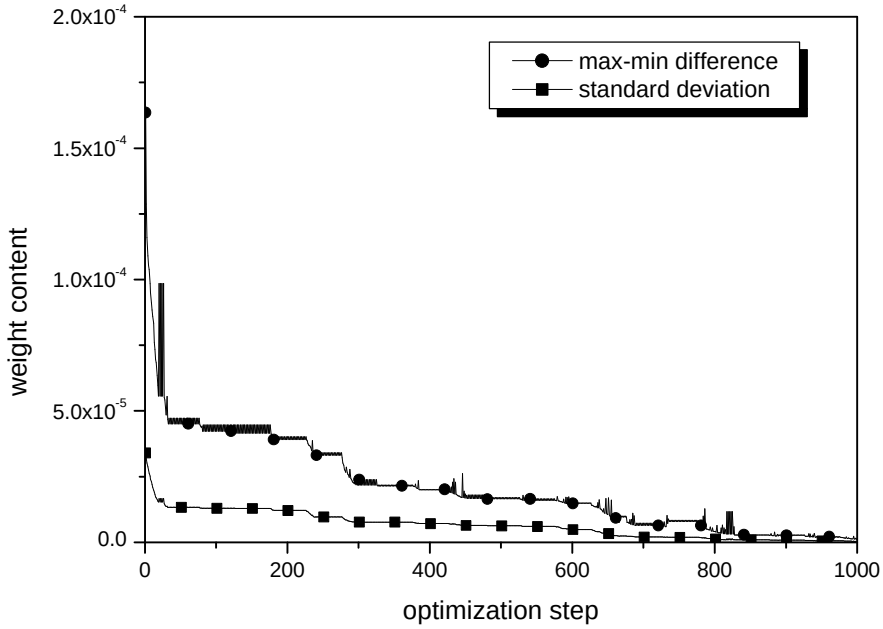


Fig. 8. Difference of isotopic content between maximum and minimum groups and standard deviation.

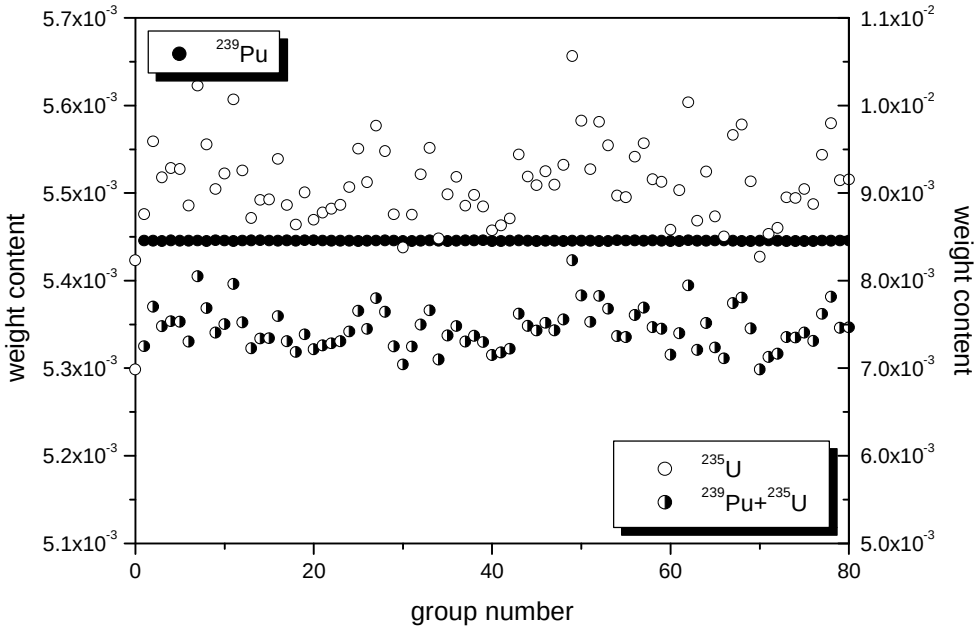


Fig. 9. Weight content of important isotopes for each group (target isotope: ^{239}Pu).

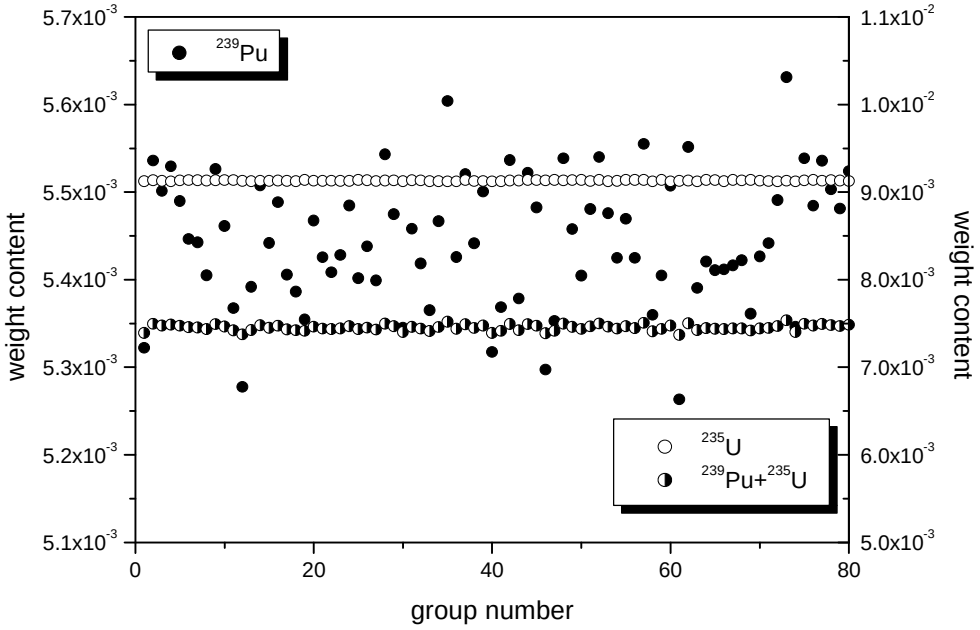


Fig. 10. Weight content of important isotopes for each group (target isotope: ^{235}U).

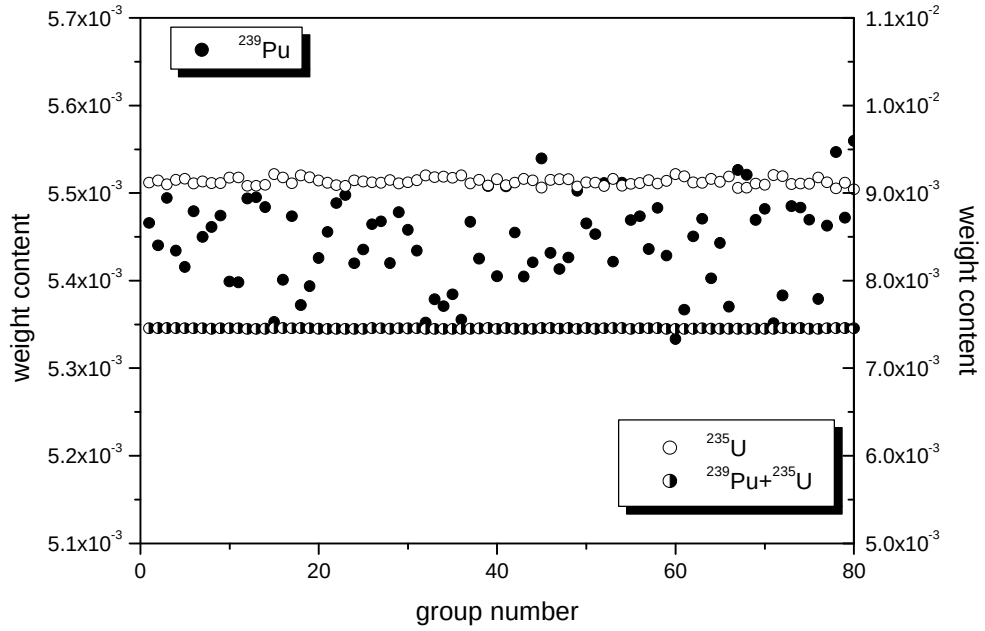


Fig. 11. Weight content of important isotopes for each group (target isotope: ^{239}Pu and ^{235}U).

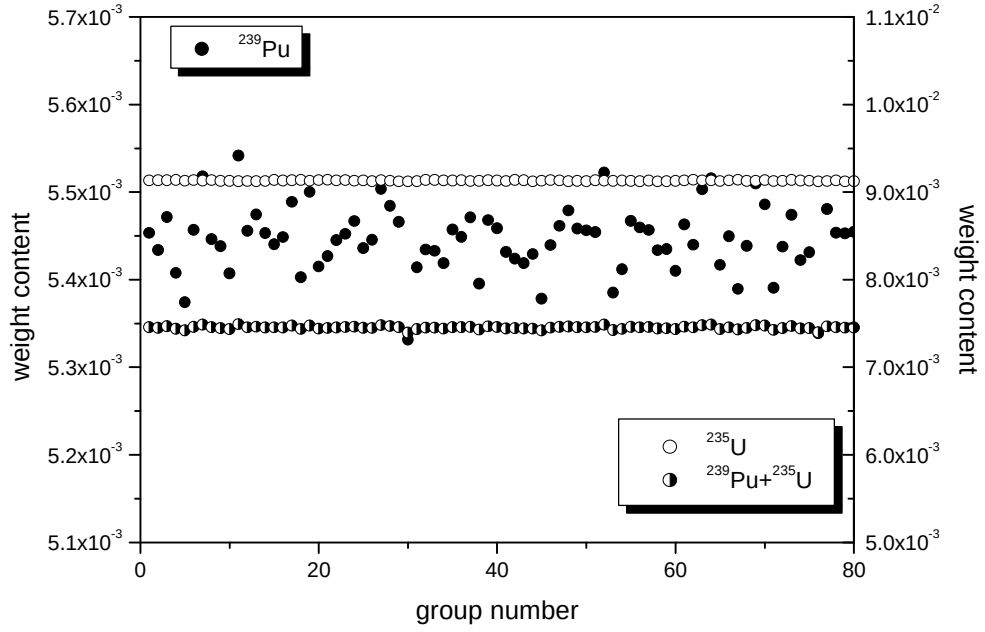


Fig. 12. Weight content of important isotopes for each group
(target isotope: periodic change between ^{239}Pu and ^{235}U).

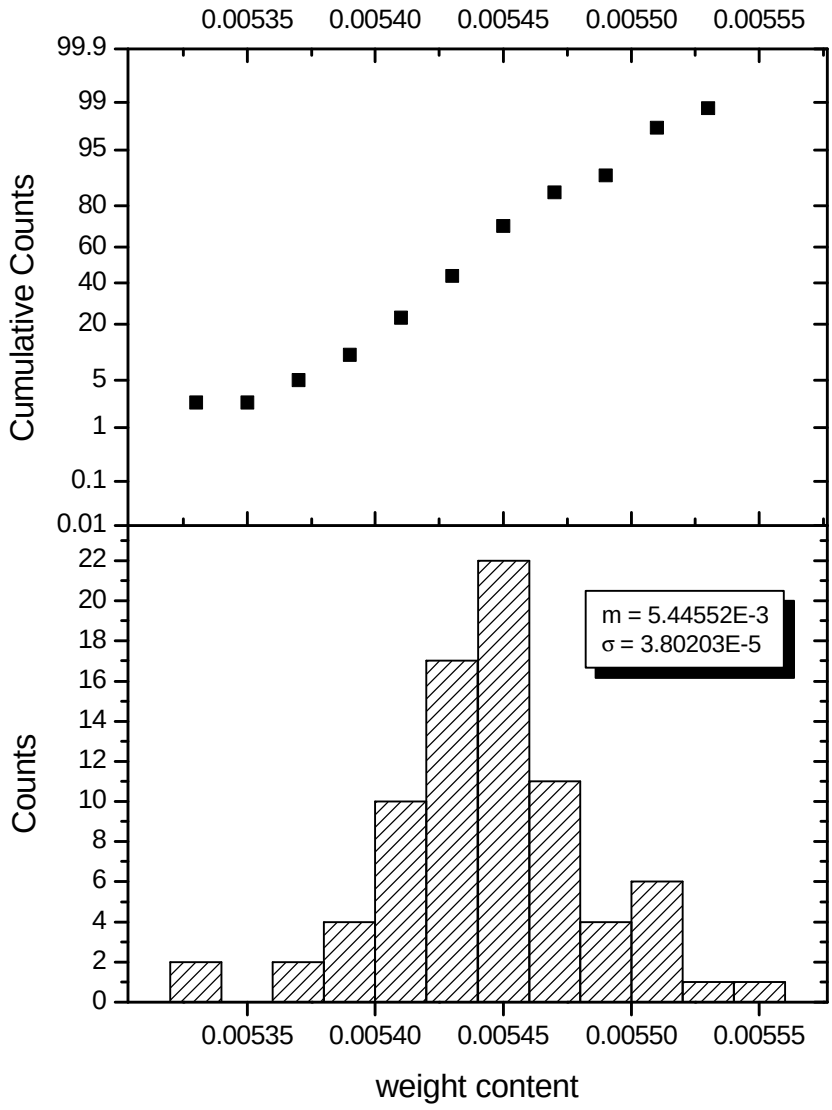


Fig. 13. Histogram of ^{239}Pu content for each group (target isotope: periodic change between ^{239}Pu and ^{235}U).

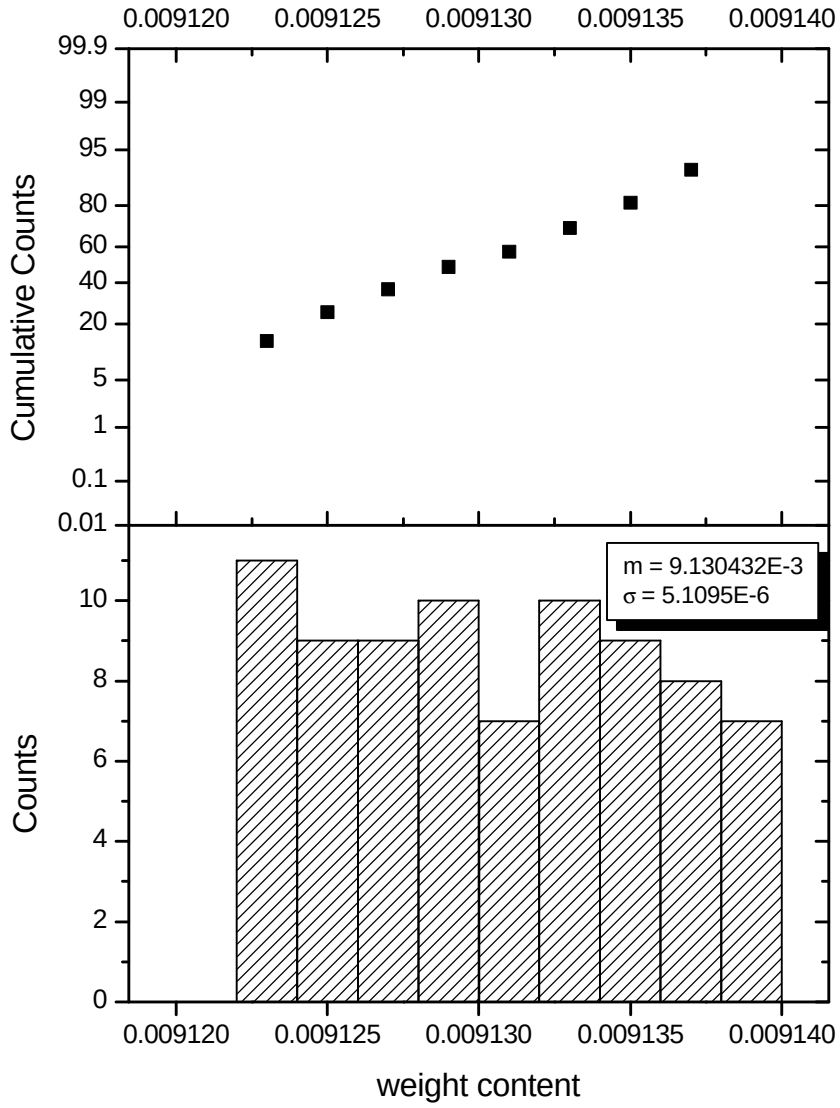


Fig. 14. Histogram of ^{235}U content for each group (target isotope: periodic change between ^{239}Pu and ^{235}U).