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Title: **ADVANCED MATERIAL ACCOUNTANCY GLOVE BOX ASSAY SYSTEM FOR PROCESS MATERIAL AT PFPF**

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NATIONAL LABORATORY

Advanced Material Accountancy Glove Box Assay System for Process Material at PFPF

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1. Abstract

The Material Accountancy Glove Box Assay System (MAGB) is a neutron coincidence counting system that has been developed under the agreement between Japan Nuclear Cycle Development Institute (JNC) and Los Alamos National Laboratory (LANL) in order to measure plutonium in the MOX transfer container in the glove box at Plutonium Fuel Production Facility (PFPF) in Japan. The system was installed at PFPF in August 1989 and afterwards it has been using for verification of plutonium in the transfer container by inspectorate during inspection

Process equipment to improve its performance has been installed in the PFPF fabrication line. As a result the number of sample required for inspection increased. Since MAGBs were only used, it was anticipated that the reduction of the time of sampling and movement time of nuclear materialfor verification would be difficult. Furthermore, the sample has to be taken from the transfer containers that contain a large amount of plutonium. Therefore, in order to reduce the time for sampling, time required for the transfer container to arrive at sampling point, and personal radiation exposure, the integrated MAGB system combined with High resolution gamma-ray spectroscopy (HRGS), called AMAGB, was developed.

This system was installed at PFPF in August 2000, and then, the functional test and the calibration were carried out in the presence of inspectorate. In the course of these activities, the data that is necessary for evaluating the system were fully acquired. The system is now being in the process of evaluation whether it can be used as an attended safeguards system for International Atomic Energy Agency (IAEA) and Japan Safeguards Office (JSGO).

2. Introduction

PFPF is the facility which adopted remote automatic operation equipments to demonstrate its use in a plant of mass production technology of MOX fuel. This facility supplies fuels to the prototype FBR "MONJU" and the experimental FBR "JOYO". Safeguards systems at this facility were developed under JNC/DOE joint study in order to increase the efficiency of inspection activities, reduce the radiation exposure dose during inspection activities and accommodate the remote automatic operation equipment. As one of these systems, MAGB[1], which is a nondestructive assay system, was developed to measure plutonium in the MOX transfer container in the glove box by neutron coincidence counting technique[2]. This system was installed at PFPF in August 1989 and afterwards it was used for verification by inspectorate during inspection. However, the following difficulties have become obvious recently.

1. Increase of number of samplings in inspection
2. Measurement of sample contains additive material that affects precision.

As a solution to these difficulties, the development of AMAGB was started in 1996 with the purpose of improving the objectives such as efficiency, precision and impact of inspection activities. This system was installed at PFPF in August 2000, and then, the performance test and the calibration were carried out in the presence of inspectorate.

3. Overview of AMAGB

3.1 AMAGB

AMAGB was developed to measure plutonium in the transfer container and this system measures by detecting the time-correlated neutrons emitted from the even isotopes of plutonium. The ^{240}Pu effective mass is evaluated based on the calibration curve of mass as a function of the coincidence count rate. Concerning the improvement of the measurement efficiency, in order to measure plutonium in the transfer container that contains a lot of low-z additive material, additional slabs compared with MAGB were installed and multiplicity technique[3] was adopted in conjunction with known alpha technique. The higher efficieny helps reduce the long count time needed for multiplicity assay.

Two AMAGB systems were developed and installed at PFPF. The AMAGB system consists of neutron coincidence counter to assay the ^{240}Pu-eff mass, and electronics cabinet including personal computers, shift register, printer and so on. Figure 1 shows a schematic diagram of the AMAGB system.

The first unit, AMAGB#1, has 4 slab type detector and is located around supplemental transfer glove box which is the by-pass route connecting the process equipments directly. Figure 2 shows a photograph and a specification of AMAGB#1. In case of maintenance, the top detector slab is rotated 90 degrees so that the junction box is accessible from the front of the glove box. In addition, the front detector that faced globe panel can slide to side so that the parts of transfer equipment in the glove box can be repaired if necessary.

The second unit, AMAGB#2, has 6 slab type detector and is located around a material accountancy glove box equipped with a load cell same as MAGB. Figure 3 shows a photograph and a specification of AMAGB#2. From the point of view of the efficiency, the tubes used in the four small detector slabs of AMAGB#2 contain 10-atm of ^{3}He gas. The other two detectors slabs contain 4atm of ^{3}He gas. The detector slabs were design to be slightly undermoderated to take advantage of neutron moderation that occurs in the polyeurathane walls of the glove box. Cadmium liners are on the front and backsides of all the detector slabs.

3.2 HRGS

It is necessary to verify isotope information of inventory by using the NDA measurement. The isotopic information is used to convert the ^{240}Pu effective mass obtained from the neutron assay to total grams Pu. Typically, the operator declared isotopics are used because they have better precision, but the isotopics from NDA are used to verify the operator decalred values. Usually, the transfer container was moved to specified sampling glove box in the process area and then a small sample was manually taken from the transfer container. The small sample was measured by HRGS in the specified glove box at the analytical laboratory. Therefore, a lot of time was spent in these activities. For reducing the impact of these activities, the HRGS was installed at the location of the AMAGB station.

In addition to the neutron assay, both AMAGB systems have HRGS for measuring the plutonium isotopic composition of the samples at the AMAGB station. The HRGS systems adopted mechanical cooling systems in place of liquid nitrogen to keep the germanium crystals at cryogenic temperatures during operation because the HRGS systems must be mounted on a mechanical lifter to position the detector on the center of the sample (about 3 m above the floor).

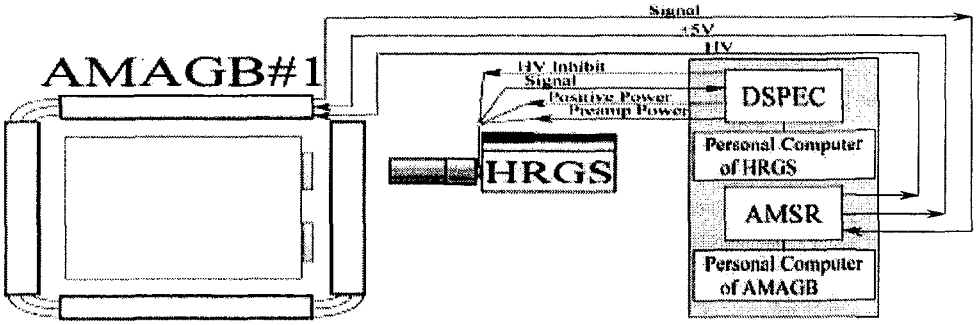

Fig. 1 Schematic diagram of AMAGB.

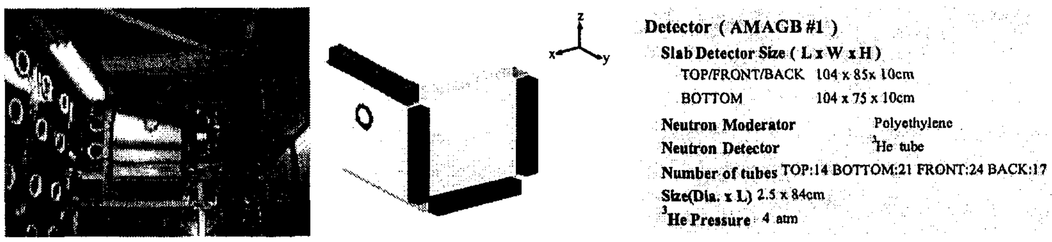

Fig. 2 Photograph and a specification of AMAGB#1.

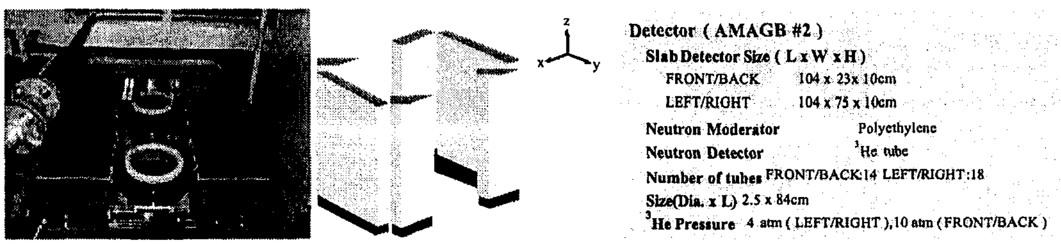

Fig. 3 Photograph and a specification of AMAGB#2.

3.3 Software of AMAGB and HRGS

The ability to review multiple types of data in a quick and easily understandable manner is needed. Figure 4 shows the software configuration of AMAGB system with new HRGS system. Neutron measurement data is evaluated by the Integrated Review, Radiation Review, INCC and Operator Review. Gamma-ray measurement data is evaluated by the VB-MGA or FRAM. Figure 4 illustrates the data-matching process and interaction between four different review tools and communication tool.

Especially, Integrated Review software provides a common starting, importing, comparison and interaction interface for all of the detailed review tools.

As a result, these softwares can provide a means to display the data and results graphically or to display a subset of the data in the database for inputting ID information and isotope ratios.

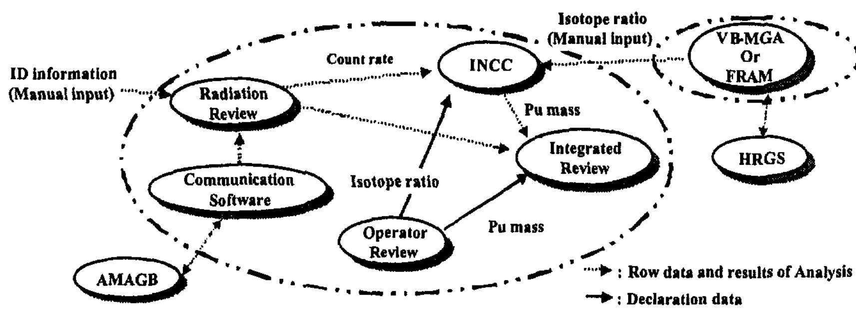

Fig. 4 The software configuration of AMAGB system with new HRGS system.

4. Functional Test and Calibration (attended mode)

The functional test and calibration were carried out from 9/18/00 to 9/27/00 at PFPF. These examinations were executed by JSGO, IAEA, LANL and JNC. The acceptance test was also performed by IAEA at the same time.

4.1 Efficiency

The Efficiency of the AMAGB detectors was measured by using a calibrated Cf-252 source in the center of the detectors before installing AMAGB around the glove box. The measured totals rate from the shift register data were background and deadtime corrected and divided the decay-corrected activity of the source. The results are reported in Table 1. The efficiencies of AMAGB#1 and #2 are, respectively, 1.85 and 1.63 times better than that of MAGB.

Table 1 Efficiency Calculation for ^{252}Cf source.

Detector	Singles Rate (cps)	Background (cps)	Initial Rate (cps)	Efficiency	Efficiency (MCNP)	Efficiency (MAGB)
AMAGB#1	4519.773 ± 1.534	77.421 ± 0.051	39894.10	11.1%	10.7%	6%
AMAGB#2	4059.076 ± 2.392	138.717 ± 0.318	40096.08	9.8%	9.46%	

4.2 Axial and Radial Response Profiles

The spatial response function of the AMAGB instrument was characterized by moving a ^{252}Cf source along each axis centered between the slabs, and by the generation of a response surface corresponding to the summation of the three axes to the slab detectors. Figures 5 and 6 compare the Monte Carlo generated coincidence response curve with the actual measured data for the vertical (z-axis) and horizontal (x,y-axis) direction. The axis directions are shown in Figure 2. The measured data are plotted as circular points and MCNP[4] results are plotted as triangle-up points. It is clear from figures that both the results agree very well and the efficiency profiles are flat to within ± 5 % over the transfer container.

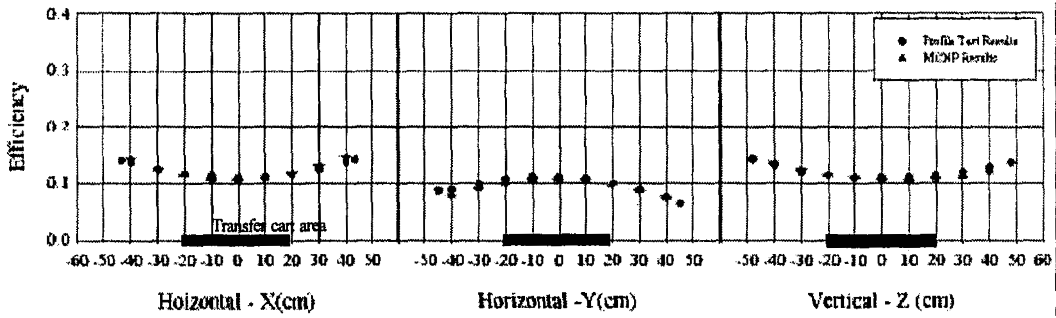

Fig. 5 Coincidence response of AMAGB#1 displacement of ^{252}Cf source.
Triangle-up points are MCNP-generated simulation and points are actual measured data.

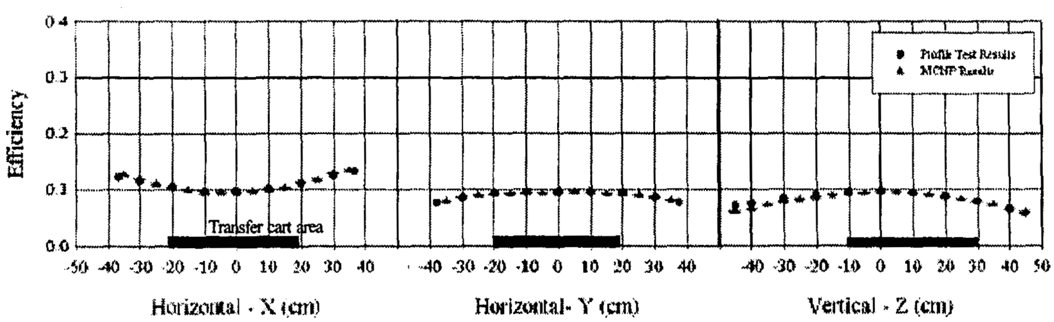

Fig. 6 Coincidence response of AMAGB#2 displacement of ^{252}Cf source.
Triangle-up points are MCNP-generated simulation and points are actual measured data.

4.3 Calibration

4.3.1 Standards

Table 2 lists the material types to be measured during the calibration. The MOX that is placed in the AMAGB systems is contained in either powder or pellet transfer containers. The type of material are categorized on the basis of powder, pellet and recovered scrap that contains dirty powder and pellets. Feed powder was not measured with the AMAGBs because feed powder will never be processed in the areas where the AMAGBs are located due to the criticality safety concerns. Currently, green pellets are not available at PFPF because the production line is being shutdown. This material will be calibrated at a later date once green pellets are available.

Table 2 PFPF material types measured in the AMAGBs.

MATERIAL TYPE	CONTAINER TYPE	MATERIAL TYPE	CONTAINER TYPE
POWDERS		PELLETS	
Feed Powder	(not measured AMAGBs)	Sintered Pellets	Molybdenum
Blended Powder	Aluminum(New)	Sintered Pellets	Aluminum
Dry Recovered Powder	Aluminum(New)	Green Pellets	(none available now)
RECOVERED SCRAP			
Dirty Powder and Pellets	Aluminum(Old)		
Dirty Powder and Pellets	Stainless Steel		

Tables 3 and 4 list the isotopic composition and mass attributes of the calibration samples. The isotopic compositions were supplied by JNC and will be verified by the IAEA using destructive analysis techniques. Sample ID's beginning with 'R' are powder containers, and ID's beginning with 'T' are pellet containers. The pellet containers are square and have multiple trays that hold the pellets.

Table 3 Uranium and Plutonium mass of calibration standards measured in AMAGB #1 and #2.

Sample ID	MOX Mass (g)	Pu Mass (g)	$^{240}Pu_{EFF}$(g)	U Mass (g)	^{235}U Mass (g)	Container Type	Material Type
R023	981	202.68	71.80	538.18	17.18	Stainless Steel	Dirty Scrap
R027	18,009	4,563.66	1627.78	10,816.23	1,999.38	Stainless Steel	Powder
R159	21,230	4,886.50	1686.56	12,902.95	120.00	Aluminum	Dirty Scrap
R210	6,889	1,693.49	572.82	4,180.92	777.23	Aluminum	Powder
R215	18,980	4,764.22	1696.79	11,492.40	2,094.03	Aluminum	Powder
R219	9,660	2,377.77	804.28	5,870.31	1091.29	Aluminum	Powder
T027	4,690	1,226.70	439.26	2,898.95	528.25	Aluminum	Pellet
T098	11,752	3,055.52	1090.84	7,253.33	1,332.36	Aluminum	Pellet
T127	6,680	1,735.58	611.34	4,138.80	719.28	Molybdenum	Pellet
T148	1,440	374.14	131.79	892.19	155.05	Molybdenum	Pellet

Table 4 Isotopic composition of calibration standards measured in AMAGB #1 and AMAGB #2.

Sample ID	^{238}Pu (wt%)	^{239}Pu (wt%)	^{240}Pu (wt%)	^{241}Pu (wt%)	^{242}Pu (wt%)	Pu Date	^{241}Am(ppm)	Am Date
R023	1.224	62.753	24.897	6.696	4.430	98/07/22	29,000	98/07/22
R027	1.262	61.457	24.918	7.857	4.506	97/07/30	15,900	97/07/31
R159	1.187	63.857	24.486	6.281	4.189	98/07/06	32,620	98/07/06
R210	1.090	62.630	24.190	7.990	4.100	94/09/12	12,700	94/09/14
R215	1.251	61.493	24.933	7.841	4.482	97/06/19	15,500	97/06/20
R219	1.090	62.630	24.190	7.990	4.100	94/09/12	12,700	94/09/14
T027	1.282	61.389	24.947	7.840	4.542	97/05/15	17,300	97/05/15
T098	1.274	61.380	24.910	7.924	4.512	97/05/15	16,000	97/05/14
T127	1.215	62.081	24.824	7.512	4.368	97/07/24	17,700	97/07/25
T148	1.215	62.081	24.824	7.512	4.368	97/07/24	17,700	97/07/25

4.3.2 Neutron Calibration Results (Known-alpha analysis technique)

A total of ten samples were measured at each station. The IAEA took samples of the calibration standards to verify the declared plutonium isotopic composition. The standards were also weighed on a scale calibrated with IAEA mass standards to verify the total mass of each standard. Four calibration curves were generated for each material type to be assayed in the AMAGBs: blended powder, recovered powder, sintered pellets, and recovered scrap. The known alpha analysis method was used to determine the calibration coefficients. With the known alpha analysis, the measured doubles rate is corrected for neutron multiplication in the sample by assuming the value of alpha can be calculated from the known isotopic composition of the sample. This assumption reduces the number of unknowns to two - mass and multiplication. Alpha is defined as the ratio of the number of neutrons produced from (alpha,n) interactions to the number of neutrons produced from spontaneous fission reactions within the sample. With two unknowns and two measured quantities (singles and doubles rates), the sample mass and sample multiplication can be determined. The multiplication corrected doubles rate is then plotted as a function of Pu-240 effective mass to obtain the calibration curve. Since the count rates have been corrected for neutron multiplication, the calibration curves are linear and have the following form.

Dmc = a + bm

where Dmc is the multiplication corrected doubles rate, m is the of Pu-240 effective mass, a is the intercept of the calibration curve, and b is the slope of the calibration curve.
Table 5 summarizes the calibration coefficients from the known-alpha analysis technique for both AMAGB #1 and AMAGB #2. Figures 7 through 10 show plots of these calibration curves for each of the four material types for both AMAGBs.

Table 5 AMAGB calibration coefficients for the known-alpha analysis technique.

Detector	Material Type	Material Description	a	Var (a)	b	Var (b)	Cov (a-b)
AMAGB #1	BP	Blended Powder	1.3654E-6	2.3314E-5	9.3820	1.8502E-5	-3.5297E-8
AMAGB #1	RP	Recovered Powder	-2.5021E-7	6.8818E-5	8.9647	3.2715E-5	-4.1624E-8
AMAGB #1	SP	Sintered pellets -Al/Mo	4.309E-4	7.1701E-3	8.3500	7.6591E-4	-2.1032E-5
AMAGB #1	RS	Recovered Scrap	7.8345E-5	1.2277E-3	10.4790	7.8399E-4	-8.4426E-7
AMAGB #2	BP	Blended Powder	6.6019e-6	1.1455E-3	8.7208	3.4562E-4	-1.7687E-6
AMAGB #2	RP	Recovered Powder	-5.3733E-6	1.8577E-2	8.1604	5.4523E-3	-1.1178E-5
AMAGB #2	SP	Sintered pellets -Al/Mo	-3.9026E-3	2.5200E-2	7.7787	4.3342E-4	-3.0346E-5
AMAGB #2	RS	Recovered Scrap	-1.0810E-4	1.1210E-3	9.3019	7.8758E-4	-8.929E-7

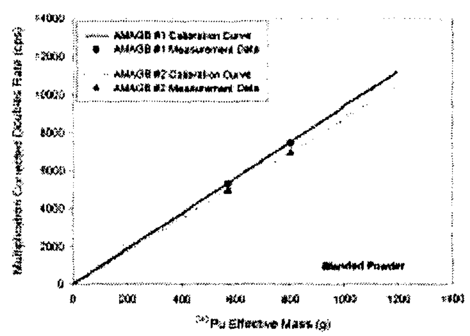

Fig. 7 AMAGB#1 and #2 Known -alpha calibration curves for blended powder. The measurement points used to generate the curves are also shown on the plot.

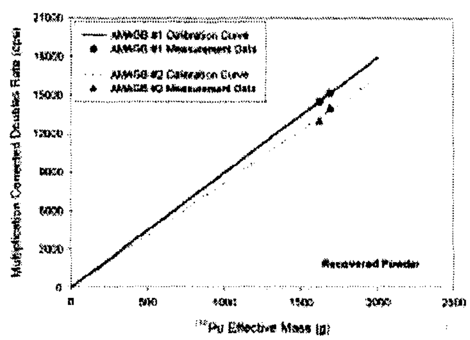

Fig. 8 AMAGB#1 and #2 Known-alpha calibration curves for recovered powder The measurement points used to generate the curves are also shown on the plot.

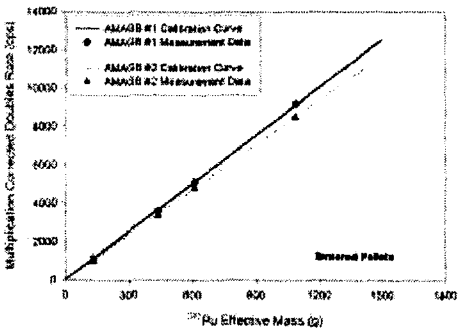

Fig. 9 AMAGB#1 and #2 Known-alpha calibration curves for sintered pellets. The measurement points used to generate the curves are also shown on the plot.

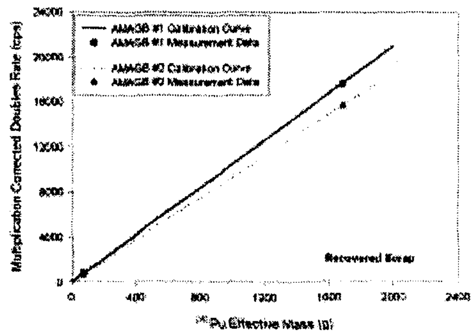

Fig. 10 AMAGB#1 and #2 Known-alpha calibration curves for recovered scrap. The measurement points used to generate the curves are also shown on the plot.

5. Future Plan

Functional test and acceptance test for the AMAGB, neutron counter and associated HRGS in attended mode, were successfully performed at PFPF in September 2000. However, the long-time measurement for by adopting multiplicity technique could not be performed during this test because of the preparation of JOYO fuel fabrication. This measurement for multiplicity technique is needed to determine parameters.

AMAGB is scheduled to be measured by unattended mode. The camera system which takes a picture of ID in the transfer container are needed to set AMAGB stations. This system is scheduled to be installed by IAEA. It is scheduled to do final acceptance test in unattended mode at PFPF in near future.

Moreover, in order to meet the final goal of this system that can be operated automatically and can be evaluated by visiting facility remotely, further developments are necessary. Especially, it is necessary to remodel and integrate the isotopic software.

6. Conclusion

A number of unattended mode NDA systems for inspection use have been developed and installed in PFPF. These systems would be a powerful tool to improve the implementation of safeguards effectively and efficiently. AMAGB combined with new HRGS would contribute to the further strengthened safeguards measure.

JNC is making continuous efforts to improve the safeguards technology not only for the implementation of safeguards in PFPF/JNC but also for international safeguards regime.

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