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Development and Implementation of GloveBox Cleanout Assistance Tool (BCAT) to Detect the Presence of MOX by Computational Approach

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Abstract − *In order to implement facility nuclear material accountancy (NMA) and safeguards properly, it is important to understand where and how much holdup deposit in the process is present for the planning of the cleanout before PIT. JAEA and LANL developed and implemented a GloveBox Cleanout Assistance Tool (BCAT) to assist cleanout activity (Recovering MOX powder in a glovebox) for invisible holdup effectively by computational approach which is called distributed source-term analysis (DSTA). To know the holdup location and activity, the BCAT tool uses a set of simple neutron slab detectors and a matrix showing the relation between source activity and measured intensity, The matrix is defined by the MCNPX simulation using known source activity (be check source) at 53 source voxels and measured intensity at 57 measurement positions almost uniformly distributed in a process room. The model of MCNPX for entire process is very precisely established.*

We have implemented and experimentally proved that the BCAT tool can direct the operator to recoverable holdup that would otherwise be accounted for as MUF. Reducing facility MUF results in a direct improvement of the facility NMA. It is expected that using the BCAT tool over time will reduce the holdup in the conversion room and the total MUF declaration at the facility, as well as enable the staff to significantly improve their knowledge of the locations of residual holdup in the process area. JAEA would like to use this application for dismantling of the glovebox with transparency in the future.

I. INTRODUCTION

When a facility operator performs cleanout activities for Physical Inventory Taking (PIT) twice a year or timely maintenance, scattered or deposited nuclear materials in a glovebox are called 'holdup.' The cleanout activities to find, recover and move holdup to (a) measureable container(s) carefully and efficiently are very important for the operator to reduce radiation exposure and also for the manager to carry out Nuclear Material Accountancy (NMA) properly and completely. However, due to the heavy shielding required to produce Plutonium-Uranium Mixed Dioxide (MOX) powder at a rate of 10 kgPu+U/day in the highest plutonium/uranium ratio that is close to but less than one as is done at the Plutonium Conversion Development Facility[1] (PCDF), there are many limitations on the cleanout activities. There are two kinds of holdup – holdup that is located inside the process equipment and holdup located outside of the equipment which is called 'loose holdup' accumulated during operation. Cleanout activities are done to the best of operator's ability within the limited days before the PIT. Especially for loose holdup, activities of finding and recovery the holdup outside the process equipment require more time and more effort. This results in increasing risks of contamination, unnecessary neutron and gamma radiation exposure and slight radiation damages of rubber gloves.

The most probable solution to this challenge is to find out location and estimate the mass of holdup by nondestructive assay (NDA) prior to the cleanout activities. Large assay systems called GloveBox Assay System[2] (GBAS /HBAS /Improved HBAS) have been developed, implemented and used for NMA to determine the mass of holdup (mass of spontaneous fission nuclei in holdup after cleanout) by passive coincidence neutron counting; however, it is difficult to apply the systems because only two large detectors (1m (W) x 1.7m (H)) are set beside a glovebox and thus two separated deposits cannot be distinguished because neutron signal from those deposits is not so different, so the system cannot be used for location finding. The location of holdup has been predicted based on the history of operation and the operational experiences deeply related to the knowledge of process equipment. However, some of the loose holdup is difficult to find and/or difficult to be recovered, which results in a bias to the Material Unaccounted For (MUF).

Therefore, we have developed and implemented a new tool composed of small total neutron counting detectors set at 57 points around the gloveboxes at the process room in PCDF and Distributed Source-Term Analysis[3] (DSTA) technique developed by Los Alamos National Laboratory.

We call it Glovebox Cleanout Assistant Tool (BCAT) that helps operator to find locations of holdup, to consider methods and tools to recover it prior to cleanout activity, and to perform the activity carefully and efficiently. It is expected that the tool has the following benefits: i) Reduce mass of loose holdup; ii) Reduce measurement uncertainty of recovered holdup because the uncertainty for the container with recovered holdup by Fast Carton Assay System[4] (FCAS) or a similar system is around ten-times smaller than the one for unrecovered holdup by the glovebox assay system; iii) Reduce time of cleanout activity and reduce radiation exposure; and iv) Reduce the bias to MUF. Another benefit is that operator can verify the presence or absence of holdup predicted by the conventional method. The BCAT tool is based on the DSTA but has been remarkably strengthened for many voxels. DSTA has been used in a variety of safeguards applications to determine the location and the quantity of neutron-producing materials contained in a large volume (ex. process room, waste storage room).

II. SYSTEM DEVELOPMENT

Development of BCAT system has been done according to the following steps:

1. Process Description and Measurement Points

Figure 1 shows the top view of MOX conversion room (A126；W 30 m × D 9.7 m × H 5.2 m) and glovebox locations (PXX is the glovebox ID). The gloveboxes with holdup are distributed entire the conversion room. Each glovebox is surrounded by neutron and gamma shields (multilayer of 7 mm-thick lead, 5 cm-thick or 2 cm-thick polyethylene-boron (BPE) and 6 mm-thick stainless steel) equipped with small thick windows (a square of side 43 cm and 4 cm-thick lead glass). In this figure, the P13 gloveboxes equipped with 2 cm BPE are used to denitrate Pu-U mixed solution using a ceramics tray[5] at a rate of five batches (2kgPu+U/batch) a day by microwave direct denitration method[6]. The denitrated solid (like a hard-cake) is then crunched in P13 and transferred to P14 by a pneumatic conveying system. P14B01 equipped with 5 cm BPE is used to shift the denitrated material to heat-resistant dishes before calcination and reduction. The P14 furnace, made of INCONEL alloy, is used for calcination and reduction of the denitrated material to MOX powder at a rate of one batch (10kgPu+U/batch) a day. P14B02 also equipped with 5 cm BPE and used for recycling the dishes and conveying pneumatically the MOX powder. P15B01 equipped with 5 cm BPE is used for grinding, and P16B01 equipped with only 2 cm BPE is used to carry a canister filled with 10kgPu+U MOX powder to intermediate storage before homogenization. Inside widths of these gloveboxes are unified into 1.0 m or 1.2 m. The average primary particle size of MOX powder is less than 0.1μm and there is little change in size before/after the calcination, reduction and grinding. Thus, the particles can easily be blown up by slow ventilation for negative-pressure control. To solve the problem, a new method to produce MOX granule with one tray/dish and without pneumatic conveying system has been studied[7]. P17 for homogenization is located in another process room due to its large inventory and radioactivity.

In order to establish BCAT measurement, as shown in the Figure 1, 57 predetermined and almost uniformly distributed locations in the conversion room for BCAT are set to cover entire room.

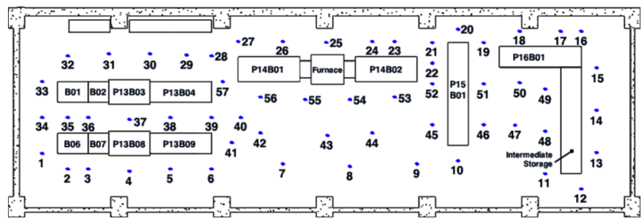
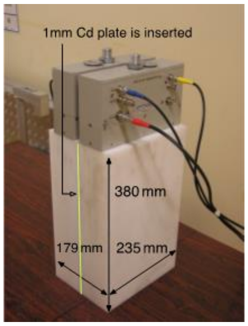


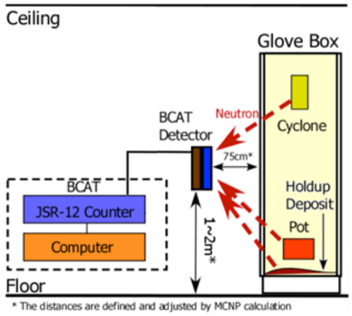
Fig.1. Top view of MOX conversion room (A126) and 57 measurement points

2. Survey of neutron intensity

We have developed a hand-held neutron detector for BCAT shown in Figure 2, which is a conventional passive total neutron counter composed of two banks where each bank has 6 one-inch 3He tubes of 33 cm active length. Between the two banks, 1 mm Cd shield is set to obtain desired directionality. The detector is set at 1-2 m high from the floor of process room and horizontally 75 cm away from the surface of glovebox in order to achieve equivalent counting efficiency both for the deposit in a cyclone (most probable in-process holdup) that is a part of pneumatic conveying system and for the deposits on a glovebox floor (most probable loose holdup). The detector has 1% efficiency for a point source at a distance of 30 cm. The most probable mass of holdup includes several 100s grams to 1 kilogram plutonium and equivalent uranium, so an operator can quickly scan a process room by doing short measurements (~2 minutes per location). The count time or position depends mainly on internal structure depends on characteristics of MOX powder (wet or dry, secondary particle size etc.) and internal structure of equipment in a glovebox. As a result, we obtain a map of Singles count rates at 57 positions shown in the Figure 1.



BCAT Detector
(Two Detector Banks)



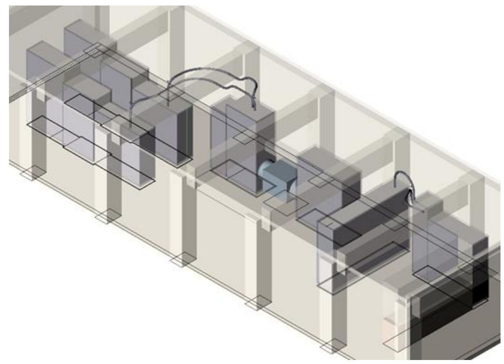
BCAT System Configuration

Measurement Scene

Fig.2. A hand-held neutron detector for BCAT and measurement scene

3. Establish MCNPX simulation model

A detailed MCNPX model for the process room composed of positions/dimensions /materials of the gloveboxes and the BPE shields, walls /floor/ceiling/beams of the room was established. The A126 model and internal view are shown in Figure 3. A test was done to validate the model by setting a certified Cf check source to the locations of holdup and loose holdup in real gloveboxes. The MCNPX model was improved so that the difference between calculation and validation was small and the difference has little bias. As a result, the sensitivity at the location of highest intensity (normally at the closest position to the check source) was 0.00059 counts/neutrons per unit time and 0.00027 counts/neutrons per unit time for 2 cm shield and 5 cm shield, respectively. The total uncertainty was estimated to be around 15%, and most of the uncertainty arose from a solid angle problem because a single mass is supposed distributed uniformly in a small region called 'source-voxel'. A total of 53 voxels were defined as shown in Figure 4. In such a case, it is inevitable that the solid angle from the supposed location to the detector and the one from the corner of the voxel to the detector are different by more than a little, which results in the difference of counting efficiency. The second effect on the total uncertainty is that the MCNPX model does not have definitions of any of the process equipment located inside the various gloveboxes. This omission may have a strong effect on the neutron scattering in the individual gloveboxes and thus limit the spatial fidelity of the material-localization results. Another effect on the total uncertainty is that Pu solution tank located one level below the west end of A126 and P17 glovebox located to the east of A126. If there is only one holdup in the process room, we can find the location and the mass by simply applying the model. However, the holdup is distributed in many voxels and we do not know clearly where the locations are even though they are predicted in the model.



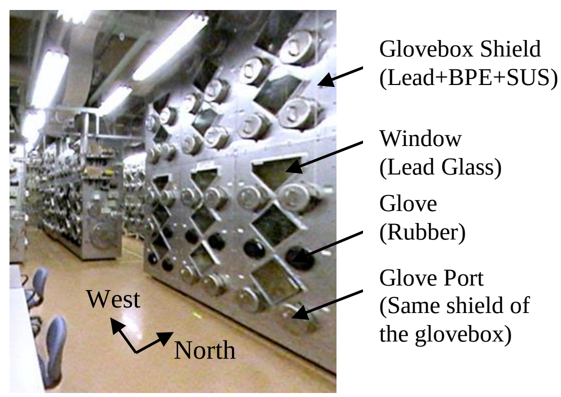


Fig.3. MCNPX simulation model and internal view of A126 room

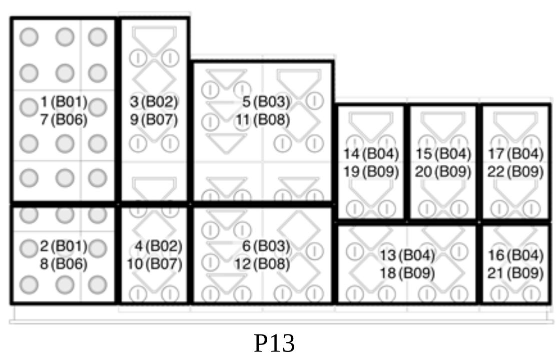
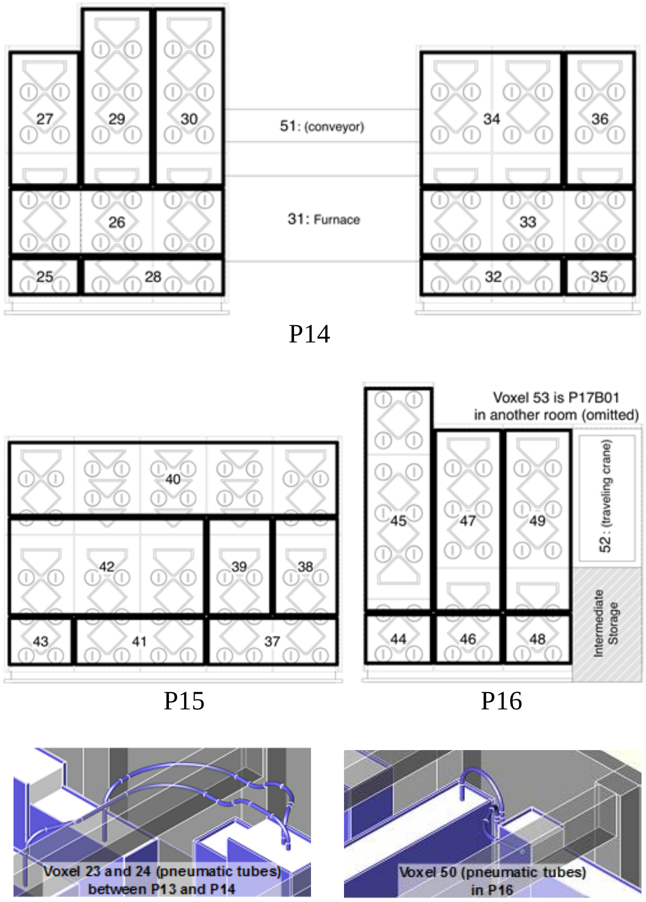


Fig.4. Definition of 53 source-voxels

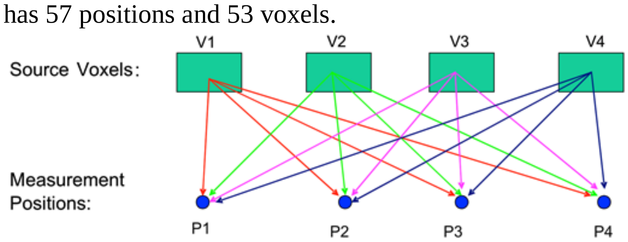


Pneumatic Tubes

Fig.4 (contd.). Definition of 53 source-voxels

4. Establish source-to-detector coupling matrix: $\mathbf{R}_{PV}$

Once the locations of holdups are discretized to 'source-voxels', we can define a matrix composed of measurement-position elements and source-voxel elements. For example, four voxels and four positions are drawn in Figure 5, and each line connecting a measurement-position (P) to a source-voxel (V) is an element of response matrix ($\boldsymbol{R}_{pv}$) considering the source-to-detector coupling. The real matrix has 57 positions and 53 voxels.



$$\rightarrow \quad \boldsymbol{R}_{PV}\boldsymbol{A}_V = \boldsymbol{M}_P$$

Fig.5. An example of four voxels and four positions, and response matrix ($\boldsymbol{R}_{pv}$)

where $\boldsymbol{A}_v$ is the vector of neutron activity (neutrons/s) at V voxels, $\boldsymbol{M}_p$ is the vector of singles count rate (counts/s) at P positions and $\boldsymbol{R}_{pv}$ is the response matrix. The final $\boldsymbol{R}_{pv}$[8] is shown in Figure 6 as a contour graph, where x-axis is source-voxel number, y-axis is measurement-position number and z-axis is response (%). Bright points which are red circled are the gloveboxes without shield. However, there is normally no holdup in these gloveboxes, so the red circled points are not bright in normal operation. The brightness unrelated to glovebox is less than 0.00005 counts/neutrons per unit time whereas the brightness corresponding to glovebox is 0.00027 for 5cm BPE or 0.00059 for 2cm BPE, which is sufficiently strong to detect Pu.

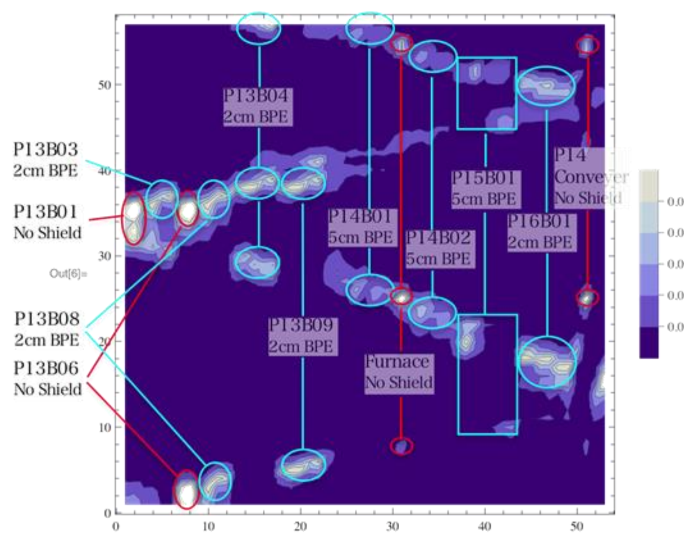


Fig.6. The final $\boldsymbol{R}_{pv}$ as a contour graph

5. Data reduction

Matrix $\boldsymbol{R}_{pv}$ is not square nor symmetrical, so inverse matrix can be obtained but it cannot be used to solve $\boldsymbol{A}_v$. In general, $\boldsymbol{A}_v$ is expressed using the transposed matrix as:

$$\boldsymbol{A}_V = \left(\boldsymbol{R}_{PV}^T \cdot \boldsymbol{R}_{PV}\right)^{-1} \cdot \boldsymbol{R}_{PV}^T \cdot \boldsymbol{M}_P \qquad (1)$$
$$\because \boldsymbol{R}_{PV}^T \cdot \boldsymbol{R}_{PV} \cdot \boldsymbol{A}_V = \boldsymbol{R}_{PV}^T \cdot \boldsymbol{M}_P$$

However, it is hard to solve this equation using the software for BCAT though a commercial software for doing Mathematics (ex. *Mathematica*®) can do this. Also, we are concerned that this equation is sensitive to the measurement uncertainty. An example of the effect of the measurement uncertainty on voxel activity is shown in Figure 7, where a) is the response vector corresponds to voxel 31, b) is the result of eq.1 when $\boldsymbol{M}_p$ is equal to the response vector a), and c) is an example of response vector with uncertainty distributed normally (σ=15%) and d) is the result of eq.1 when $\boldsymbol{M_p}$ is equal to the response vector c).

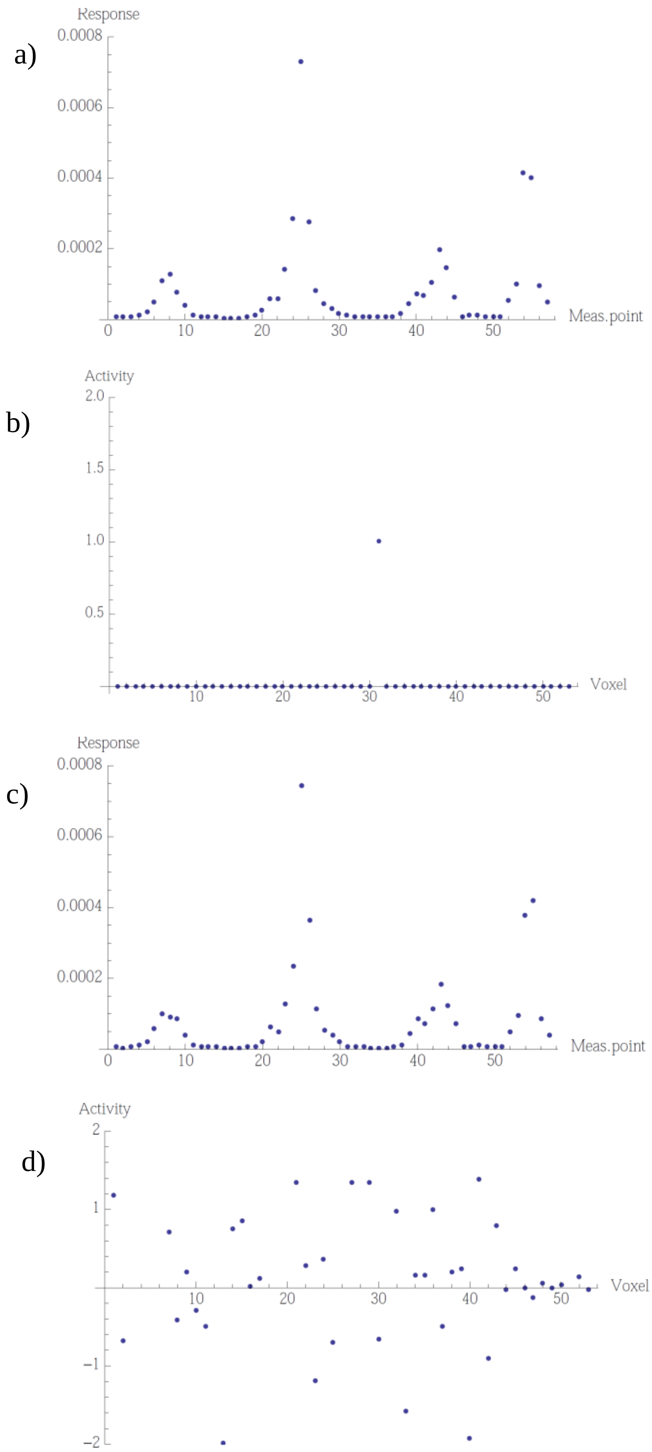


Fig. 7. An example of the effect of measurement uncertainty on voxel activity

Therefore, it became clear that the solving method using a transposed matrix was not suitable to the real measurement data with its associated uncertainty. So, we have introduced an ordinary least-square regression to find the minimum of the variance of $\boldsymbol{M_p}$ while changing $\boldsymbol{A_v}$ shown as:

$$wLSR = \sqrt{\sum_P \frac{(M_P - \sum_V R_{PV} A_V)^2}{M_P}}\Bigg|_{min} \quad (2)$$

where each $\boldsymbol{M_p}$ $\boldsymbol{A_v}$ $\boldsymbol{R_{pv}}$ is a scalar value and *wLSR* is the minimal weight of least-square regression while changing $\boldsymbol{A_v}$. The basic operation of BCAT involves numerically building the relationship shown in eq.2 from measurement data and $\boldsymbol{R_{pv}}$ matrix, and to use the Excel® solver function[9] to iteratively determine the values of $\boldsymbol{A_v}$ required to drive the relationship to a minimum.

6. An example of the initial results

Figure 8 shows an example of the initial results[10]. In this test, the source-voxels in P13 are set at Top, Middle and Bottom region that are different from the final configuration in Figure 4.

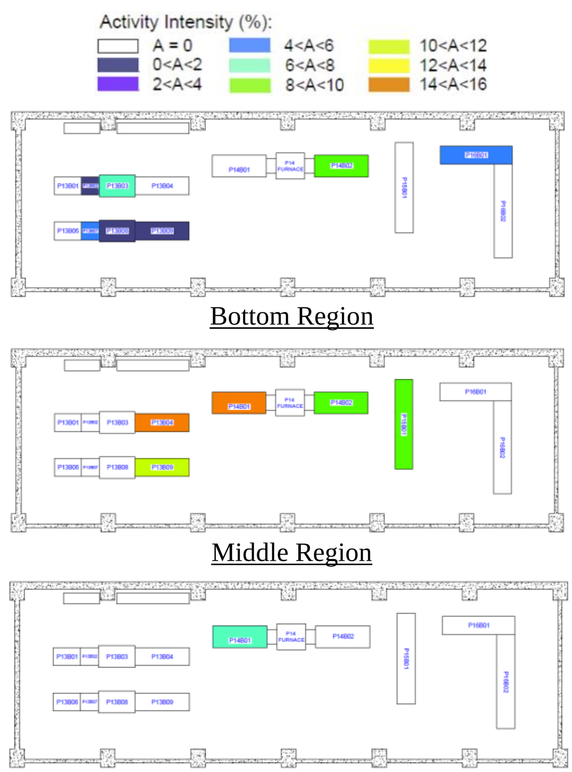


Fig.8. An example of the initial results

Based on these results (Figure 8), we found that: 1) Relatively high intensity is observed in the middle of P13B04, P13B09, P14B01, P14B02 and P15B01, also in the bottom of P14B02; and 2) Relatively low intensity is observed in P13B02, P13B03, P13B07, P13B08, P13B09 and P16B01, as well as in the top region of P14B01. This result matches our experimental prediction except for P13B02 and P13B07. As a result of visual confirmation, we found small holdup due to droplets of Pu-U mixed solution from a pipeline, which was not accounted for in our prediction. Therefore, the location finding capability of BCAT was confirmed and certified. After all, P13B04 and P13B09 are horizontally separated into three voxels and unified vertically into two voxels as shown in Figure 4.

III. EXPERIMENTAL RESULTS

The BCAT system was developed in 2010. After that, JAEA is mainly using this tool for the preparation of PIT to achieve the cleanout target. If the target is not met, we conduct BCAT measurements to determine the location of hidden holdup. In some cases, we repeat the measurement. Since one BCAT measurement activity for the entire room needs 57 points measurement points and detector moving time, the entire activity takes about 5 hours (all measurement is finished during 1 day). After the measurements are completed, every operator can know the activity of each location of each glovebox by using the neutron count rates (Singles), the uncertainties and Excel (to convert Singles to the neutron activity of each source voxel) software. Then, the operator can determine the relative Pu inventory distributions throughout the conversion room. This knowledge is helpful to decide the cleanout plan by the operator without increasing the radiation exposure. In addition, when we first introduced the BCAT into the process, we were able to find out unmeasured (unknown) inventory successfully. It seems that it is very difficult to convert from the activity to Pu mass directly due to large uncertainty of correction factor (Am build-up, (α, n) reactions and solid angle, etc.).

IV. DSTA APPLICATION

The DSTA technique has been used in a variety of safeguards applications to determine the location and quantity of material contained within large volumes. In the actual nuclear fuel cycle field, several applications using DSTA technique were being introduced to solve safeguards subjects and nuclear material accountancy[3]. In order to estimate unmeasured nuclear material in the enrichment cascade halls and low active solid waste storage areas, the application which consists of a neutron detector and MCNP simulations were developed and implemented. As a result, we were able to estimate their inventories successfully. Recently, in order to solve the actual nuclear material accountancy issue, we developed the “Dynamic Cross-Talk Correction (DCTC)” methodology using DSTA technique and introduced with authorization by the IAEA. The DCTC improved PCDF nuclear material accountancy by eliminating the double-counting of material that stems from cross-talk in the holdup assay data and thus eliminating this source of bias in the assay results[11]. The DCTC methodology can be used to determine the cross-correlation among multiple inventories in small areas and substantially reduce cross-talk-induced biases in the assay results.

V. CONCLUSIONS

This BCAT tool is routinely used during each cleanout activity to help the operator identify the physical locations where holdup material can be recovered. We have implemented and experimentally proved that the BCAT tool can direct the operator to recoverable holdup that would otherwise be accounted for as MUF. Reducing facility MUF results in a direct improvement of the facility NMA. It is expected that using the BCAT tool over time will reduce the holdup in the conversion room and the total MUF declaration at PCDF, as well as enable the staff to significantly improve their knowledge of the locations of residual holdup in the process area. JAEA would like to use this application for dismantling of the glovebox with transparency in the future.

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