

Monte Carlo Simulation of NDA for Fissile Materials Humidity

Valeriy I. BULANENKO, Vjacheslav B. POLEVOY

Federal State Unitary Enterprise State Scientific Center of the Russian Federation –

Institute for Physics and Power Engineering named after A.I. Leypunsky ,

Bondarenko sq.1, 249033, Obninsk, Kaluga region, Russia

The ensuring of the criticality safety during handling with nuclear materials (UO_2 or PuO_2) demands to measure values of moisture.

The optimization of the characteristics of the device for small values of moisture and influence estimation of the various factors using computer codes and mathematical modeling is preferable. Such model should reflect in detail real conditions of source-detector geometry and to be suitable for comparison of results of relative measurements on similar installations. For modeling the Monte Carlo method was chosen whose basic advantage against other numerical methods consists in direct numerical calculations for practically real source-detector geometry.

The complex of the programs MMKFK-2 developed in SSC IPPE was used for modeling. The task for point (in a case UO_2) or volumetric distributed (in a case PuO_2) source of neutrons with given spectrum was calculated for various parameters of the unit and properties of a powder included:

- * composition of materials of neutrons reflector and its thickness;
- * bulk density of a powder;
- * position of the neutrons detector;
- * isotope composition of nuclear materials.

Model and the experimental researches have shown that the usage of hydrogen reflector for the moisture measurement in powder oxide of uranium or plutonium less than 2 % is practically impossible. For realization of a neutron method of the moisture measurement in UO_2 or PuO_2 the presence of a metal reflector is obligatory.

KEYWORDS: *criticality safety, fissile materials, plutonium oxide powder, uranium oxide powder, moisture control, non-destructive assay, simulation Monte Carlo*

1. Framing the problem

Actually any technology of producing U or Pu oxide powder inevitably results in the presence of free or chemically bound hydrogen. Hydrogen sources could originate from different peculiar features of technological processes (the use of hydrogen-containing reagents, humidity sorption from the atmosphere, deviations from process conditions, emergency conditions with water leaks from cooling pipes, etc.). For instance, a significant value of moisture in PuO_2 and MOX powder was pointed out of the Euratom facilities in Sellafield and Mol.¹⁾

Hydrogen (moisture) control is required in order to solve two main problems:

1. To provide criticality safety when UO_2 and PuO_2 are handled.
2. To improve the NMC&A system.

According to the criticality safety requirements the hydrogen content is strictly limited because this value significantly impacts the value of oxide critical mass and thus, the characteristics of containers and storage facilities. In practice the hydrogen content is limited with ~ 0.2 wt %. To control hydrogen (moisture) is

extremely important when powder batches are formed before the containers go to be stored or transported or before the process of compacting nuclear fuel pellets. For water reprocessing conditions hydrogen will most probably be present in the form of water. The dynamics of UO_2 and PuO_2 fabrication process is such that the significant from the point of view of safety changes can only happen within the time exceeding several minutes. The powder moisture is one of the controlled parameters in the criticality safety.

The hydrogen (moisture) presence results in certain problems for the procedure of measurement verification in the NMC&A system. In this case the problem consists in the necessity to introduce the adequate corrections, which could be only obtained directly in measurements.

The main requirements imposed upon the method of hydrogen (moisture) measurement and the measurement devices are as follows:

- * a sufficient sensitivity and express nature;
- * no direct contact with NM powder;
- * possibility to automatize the process of material unloading to the container of favorable geometry or the product loading for

reprocessing under the conditions of increased moisture.

It is quite obvious that destructive methods of nuclear materials measurement do not satisfy the indicated requirements because of their drawbacks (the necessity to take samples, the analysis is not representative with local moisture spots, the destructive assay lasts quite long, etc.). Based on the world practice of using NDA, the most acceptable way to solve this problem will consist in neutron NDA methods.

It should be noted that as compared with other compounds nuclear materials present a more complicated task of determining their moisture due to the following reasons: low values of moisture under measurement; a rather small volume of powder; a relatively high density; variable uranium enrichment and Pu isotopic composition, neutron multiplication in the powder volume, etc. So specifically to control moisture in nuclear materials new designs of measuring devices with metal reflectors have been developed quite recently.^{2,3)}

The paper considers the specific features of numerical modeling of moisture meters like these by the Monte Carlo method. The main emphasis is made on the evaluation of those parameters, which affect the sensitivity of low enriched UO₂ moisture measurement. The numerical modeling of PuO₂ moisture meter only differs in the way how the volumetric neutron source and its energy spectrum are preset.

2. MMKFK-2 code package

The main task of the given physical modeling is to determine the parameters and to quantitatively estimate the effect of separate factors on the neutron detector response as well as to optimize the principal parameters of a neutron moisture meter.

The calculation physical model should be rather detailed and reflect the real measurement conditions, and it should be applicable to perform comparative relative measurements with various devices.

The final goal consists in demonstrating the possibilities to model a source-detector geometry with acceptable accuracy. It is quite obvious that a complicated design of moisture meter, a small volume under measurement, the measurement geometry and specific features in formation of neutron energy spectrum in each area make the task to determine the detector count efficiency rather labor consuming and analytically not solvable with acceptable accuracy. Based on that the Monte Carlo method was chosen for physical modeling. Its main advantage over the other methods consists in the direct numerical calculation under the conditions of nearly real geometry. The code package called MMKFK-2^{4,5)}, was used for modeling. Its version was adapted on the Pentium PC. These codes are designed to solve neutron transfer equations

in complex 3-D geometry, in multigroup approximation by the Monte Carlo method. The package is actively used to get critical parameters of fresh and spent fuel storage facilities, process equipment and transportation devices. A successful experience has been also gained in using the package for modeling physical NDA problems.

The MMKFK-2 package consists of the basic package, MMKFK-BASE, and a set of various physical, geometrical and functional blocks. The basic package, MMKFK-BASE, allows the solution of both a homogeneous conditionally critical problem and a non-homogeneous problem with the given volume and point source of unspecified spectrum.

The MMKFK-2 advantage consists in the availability of a great number of modules for various purposes which can be combined in a set for the specific problem, i.e. to calculate the sought functionals in the optimum way in terms of high-speed operation and labor cost.

When the neutron detector efficiency is determined the problem is solved either for the locally-placed source or for the volumetrically distributed source with a preset intensity profile. In both cases there is a possibility to specify an energy spectrum according to the choice. The MONO version calculates the neutron multiplication factor in the multiplying medium, i.e. the ratio of total number of generated neutrons to the number of initial neutrons of the source. The multiplication calculation is made in view of estimates in collisions, paths and absorptions. The linear combination with a minimum dispersion is calculated.

Neutron transfer in the epithermal energy region (with the energy above 1 eV) is modeled in the 26-group and subgroup approximations with the use of BNAB-78/85/90/93 nuclear data libraries in the system of nuclear data preparation, APAMAKO-C1, PRECON and MKPA. Elastic moderation is modeled in the approximation of effective atomic mass of nuclide mixture with 26 group calculations and with point kinematic scattering relations on each nuclide with subgroup calculations.

Neutron thermalization at the energy below 1 eV is modeled in the 40 group approximation with the physical model MOFITG and nuclear data library TEPKON-90 from the MCU package.⁶⁾ There is a vast set of specialized geometrical modules and the module of combined geometry, SCG, from the MCU package to describe the geometry. In the given problem of "source – an arbitrary located cylindrical detector" the PRSAZ module with axial parallel and coaxial cylinders and plates combination or the SCG module were used.

The efficiency of neutron detector count (the absorption reaction rate on ³He nuclei) was calculated in the mode of group fluxes post-processing in the sensitive volume of the detector: in the epithermal

region with the RRATE code and in the thermal region with the RRTER code.

All the functional values are normalized to the number of neutrons being generated in the element of symmetry with consideration of neutron multiplication. The total values in the specific regions of radiation spectrum, including the detector sensitive volume, were obtained with the convolution in the chosen energy intervals.

The functional determined in this case (the efficiency of detector with ^3He whose absorption cross-section follows the law of $1/v$) is rather sensitive to the dispersion of calculated neutron flux density in thermal and epithermal energy regions. So with numerical modeling at least 10^7 histories were played. As a result, the calculated detector efficiency or count rate per normalized neutron yield of the source had the statistical error below 2% (1 sigma).

It should be pointed out that any type of numerical modeling gives certain additional information first of all for interpretation of the data obtained and for revealing the effects of separate factors in those cases when the experimental studies cannot be performed for certain reasons. This modeling cannot replace full-scale experiments or model tests completely because any calculation method needs testing with benchmark experiments. The modeling of the given problem is not the exception either.

3. UO_2 moisture meter modeling

The calculation geometry to estimate UO_2 moisture is shown in fig.1.

The low enriched uranium dioxide powder under control goes from the recovery furnace to the measurement cavity (1), which is surrounded with metal neutron reflector (2). The cavity is supported by the support flange (4) of the unloading worm. The neutron source of the Pu-Be type with the yield of $5 \cdot 10^5$ n/s is located in the reflector material (6). Neutron detectors filled with ^3He are placed there as well [(7) and/or (8)]. In order to decrease the neutron leak, to additionally form the neutron energy spectrum and to provide radiation safety the additional flat plates (5) are installed. In the design they can be joined with the reflector.

In calculation variants the following factors that could change the observed effect varied with numerical modeling:

- UO_2 moisture content, W , % mass;
- Measurement cavity reflector material;
- Powder bulk density ρ , g/cm^3 ;
- Location of neutron counters in the reflector: D_1 and D_2 geometry;
- Uranium enrichment, ^{235}U , %;
- Powder matrix composition: UO_2 or U_3O_8 ;

- Hydrogen-containing material, water, zinc stearate;
- Powder level in the measurement cavity, h , mm.

The primary converter characteristics are determining during moisture measurements. So two variants of the location of ^3He neutron detectors (4 atm, 32 mm in diameter, 270 mm as a working length) were considered:

- * D_1 counter, located on the opposite periphery side of the reflector, near the measurement cavity wall surface, the so called "separate" geometry;
- * D_2 counter, located in the immediate vicinity of the fast neutron source, the so-called "joint" geometry.
- * Both detectors are located in the reflector, in the immediate vicinity of the outer side surface of the primary converter.

This complicated source-detector geometry is described with the geometrical module SCG of combinatorial geometry.

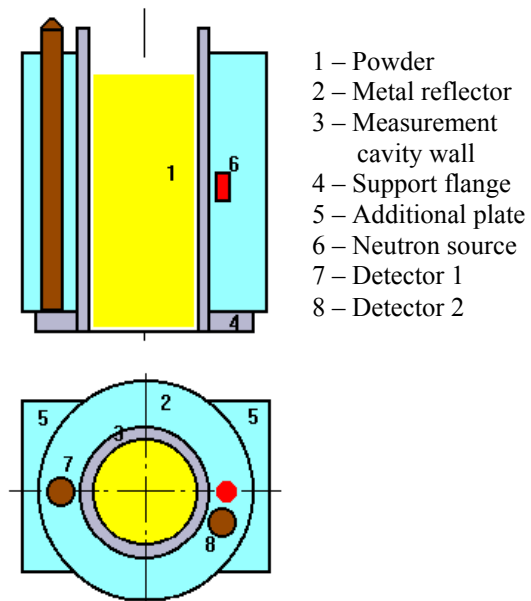


Fig.1. Calculation geometry to control UO_2 moisture. Neutron moisture meter with external source (Ni reflector).

Wet UO_2 powder increases the detector reading due to a total contribution of two principal effects:

- * neutron spectrum softening;
- * neutron multiplication.

Calculations show that even if there is no powder in the measurement cavity in the D_1 volume a rather hard neutron energy spectrum is formed due to metal reflector nuclei elastic scattering. This neutron spectrum gets somewhat softer when absolutely dry UO_2 powder is loaded. That is because of some

additional scattering on its nuclei. In case of wet powder being loaded to the measurement cavity the energy spectrum shift to the low energy region is even more profound. Finally the neutron counter readings are changing intricately with the change in the powder moisture content. For the sake of convenience the curves shown in Fig. 2 are normalized to one at the moisture of 1.0 wt %. As can be seen, a higher effect/background ratio is achieved in the “separate” geometry.

Calculations give a nonlinear function, $N = f(W)$, which only in the first approximation can have the linear function of the following type

$$N = N_0 + S_w \cdot W, \quad (1)$$

where $N = N_0$ corresponds to zero moisture ($W = 0$ wt %), $S_w = dN/dW$ – is the sensitivity of moisture control method.

The dominating contribution to the readings of both detectors comes from the energy region from 1 to 1000 eV. It is quite natural that the contribution to a lower energy region increases with the moisture growth. However the contribution of thermal neutrons with the energy below 1eV exceeds 20 % even at the moisture of 2.0 wt %. The direct experimental confirmation of the energy spectrum hardness consists in the measurement of cadmium ratio that is less than 1.25, i.e. differs insignificantly from 1.

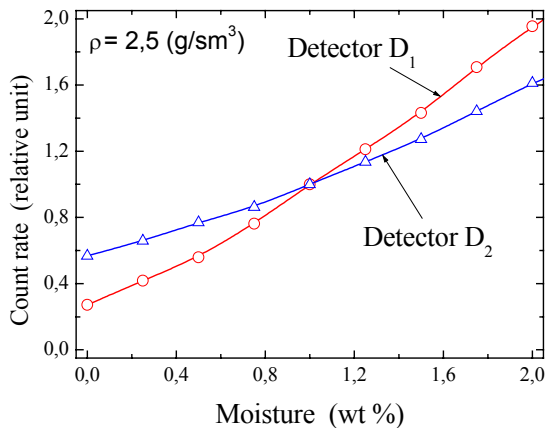


Fig.2 Count rate as a function of UO_2 moisture for two detectors, D_1 and D_2 .

At low values of moisture the nonlinear patterns of function $N = f(W)$ is related to the effect of recoil helium at elastic neutron scattering on detector helium nuclei. At high values of moisture the nonlinear function $N = f(W)$ is caused by the increase in the neutron flux density in the detector volume due to the of multiplication factor that varies from 1.05 to 1.15

within the controlled range of moisture ($0 \div 3$) wt % of low enriched uranium.

It is of no doubt that the adequate choice of material and measurement cavity side reflector thickness are determining in the design of neutron moisture meter. The main purpose of the reflector is to form the required energy spectrum in order to provide a high sensitivity of moisture control $S_w = dN/dW$, as well as to decrease neutron leakage from the powder volume and at the locations of both detectors. The importance of the optimum choice of these characteristics was experimentally demonstrated earlier².

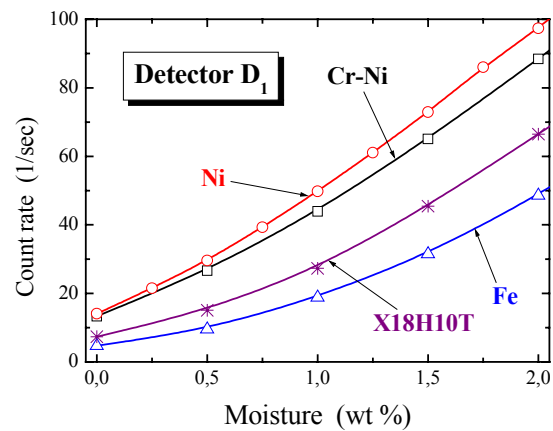


Fig.3 Count rate as a function of powder moisture. Density is 2.5 g/cm^3 .

The reflector material effect is shown in fig.3. Nickel and chrome-nickel alloys are seen to provide the highest sensitivity of moisture measurement. So in order to implement the moisture measurement method the reflector made of Ni or Cr-Ni alloy must be present. Other materials (e.g. stainless steel, iron) rank distinctly below in measurement sensitivity and accuracy. Besides, these materials give a more non-linear function $N = f(W)$, related to peculiarities of resonance structure of iron neutron elastic scattering cross-section.

The application of hydrogen containing compounds (polyethylene, caprolactone, etc.) does not actually allow low values of UO_2 moisture (below 2 wt %) to be controlled because the ratio $S_w / N_0 < 0,03$. In this case $N = N_0$ corresponds to zero moisture content ($W = 0$ wt %), $S_w = dN/dW$ – is the sensitivity of moisture assay method. Though the count efficiency turns out to be about 50 times higher in this case it is extremely doubtful that the signal could be developed from the wet powder.

In order to quantitatively estimate the effect of Ni reflector thickness and that of additional Ni layer on

the measurement sensitivity variation the functional dependence of the count rate on moisture, i.e. $N = F(W)$, was calculated.

The calculation shows that with the increase in the side reflector thickness the sensitivity grows nonlinearly and demonstrates an obvious tendency towards saturation, i.e. it tends to a constant value. Fig.4 illustrates this fact.

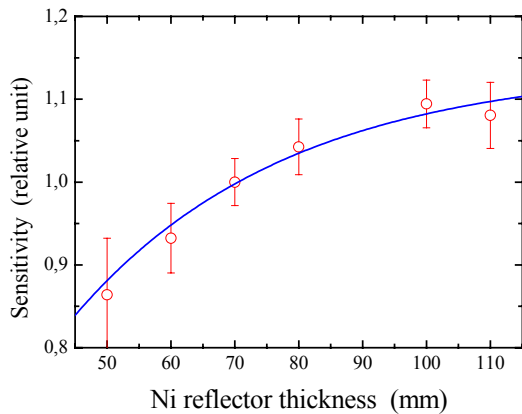


Fig.4 Ni reflector thickness effect on moisture assay sensitivity.

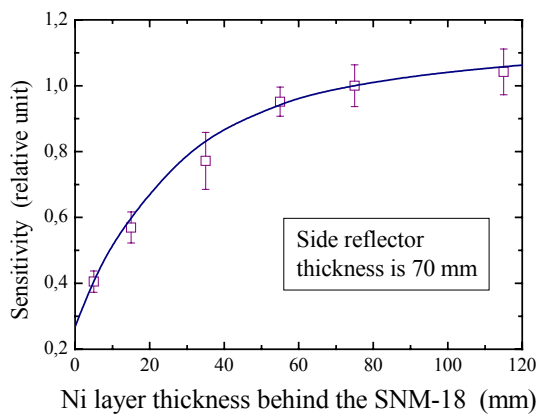


Fig.5 Effect of Ni layer thickness behind the SNM-18 neutron counter.

Sensitivity also goes up with the increase in the thickness of Ni reflector behind the neutron detector (fig.5), actually equally both on the detector side and on the side of fast neutron source. This fact was confirmed experimentally when additional flat nickel plates were located behind the fast neutron source and detector D_1 . Calculations show a more radical pattern of changes in sensitivity in case of additional

thickness behind the detector. In this case the situation of saturation is achieved much faster.

Based on the functions presented the optimum thickness of the side shielding equal to $(50 \div 70)$ mm has been chosen. The additional shielding behind the detector D_1 was chosen equal to $(40 \div 60)$ mm.

Both in the course of measurements during moisture meter operation and during its calibration the powder matrix composition can vary: UO_2 , U_3O_8 , wet powder or powder containing zinc stearate. With the same value of bulk density the nuclide concentration can be somewhat different, thus resulting in certain changes in moisture meter reading.

The results of quantitative estimation of numerical modeling in passing from U_3O_8 to UO_2 are shown in fig.6. As it can be seen, when U_3O_8 is replaced with UO_2 , the readings decrease by about 8 %, and that is confirmed by measurements. The dependence of this correction on the powder moisture is rather weak. The average value can be taken with the equal level of reliability.

As the neutron spectrum is rather hard, the effect is primarily caused by the increase in uranium nuclei concentration by 4 % for UO_2 ; the oxygen effect is negligibly small. So if this effect is neglected, it will result in underestimating the moisture content by $(0.05 \div 0.07)$ wt % in the measured range up to 2 wt %. The similar calculations for zinc stearate with the equivalent moisture instead of the wet powder demonstrate a good agreement within the calculation error, i.e. the data deviation does not exceed $1.5 \div 2.0$ wt %.

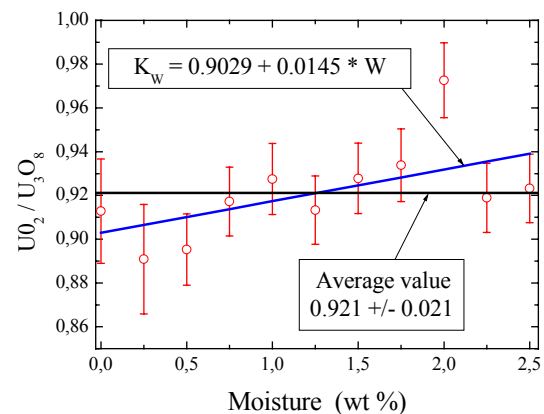


Fig.6 UO_2 composition effect.

The bulk density will evidently result in changes in the moisture meter reading because actually the total moisture content is measured in the measurement cavity. The numerical modeling confirms a rather distinct change in the reading if the density goes up, especially at the moisture above 2.0 wt %. However

the response of two detectors (D_1 and D_2) is different. A different pattern of detectors' response to the bulk density variation makes it actually possible to implement the two-parametric option of neutron moisture meter, when the powder moisture and density values are measured in a wide range simultaneously. Along with that, in case of the moisture meter option with only one detector D_1 and with the moisture limit to 2 wt %, it is allowable to use the reduced moisture adjusted for the average bulk density of the powder.

The estimation of uranium enrichment effect also deserves attention. This effect is very small and does not exceed 4 % when uranium enrichment varies from 3 % to 5 % for D_1 . If uranium enrichment grows the measurement sensitivity goes down.

4. Conclusion

Numerical modeling of neutronics problems dedicated to the assay methods is becoming a general practice. The advantages of these calculations are first of all determined by the possibility to quantitatively estimate many factors. Especially it is important in those cases when direct experimental measurements or full-scale testing turn out to be too burdensome and expensive. The accuracy obtained in calculations is usually comparable with the results of test measurements.

The calculated estimations of many factors related to the moisture assay method that are given in this paper are quite predictable and confirmed experimentally. For instance, the dependence of information signal on moisture, the effect of powder bulk density and its composition.

5. References

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