An automated system for the preparation of Large Size Dried (LSD) Spikes

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Abstract *–Large size dried (LSD) spikes have been produced to fulfil the existing requirement for reliable and traceable isotopic reference materials for nuclear safeguards.*
A system to produce certified nuclear isotopic reference material as a U/Pu mixture in the form of large size dried spikes, comparable to those produced using traditional methods has been installed in collaboration with Nucomat, a company with a recognized reputation in design and development of integrated automated systems. The major components of the system are a robot, two balances, a dispenser and a drying unit fitted into a glove box. The robot is software driven and designed to control all movements inside the glove-box, to identify unambiguously the penicillin vials with a barcode reader, to dispense the LSD batch solution into the vials and to weigh the amount dispensed.
The system functionality has been evaluated and the performance validated by comparing the results from a series of samples dispensed and weighed by the automated system with the results by manual substitution weighing. After applying the proper correction factors to the data from the automated system balance no significant difference was observed between the two. However, an additional component of uncertainty of $3 \cdot 10^{-4}$ *is introduced in the uncertainty budget for the certified weights provided by the automatic system.*

INTRODUCTION

Over many years, IRMM has acquired long term experience on the preparation Large Size Dried (LSD) Spikes. The LSD spikes have been produced to fulfil the existing requirement for reliable and traceable spikes in fissile material control of dissolved nuclear fuel.
Traditionally the preparation of the LSD spikes was accomplished manually. In the method employed at IRMM [1], very high quality isotopically enriched metals of uranium (natural and high-enriched) and plutonium are dissolved into a single large volume of batch solution. From this solution, individual spikes are made by aliquoting weighed amounts of the solution into penicillin vials and drying carefully. Finally, a cellulose layer is dried onto the spike material to retain the spike at the bottom of the vial during transport to the laboratory where it will be applied. The spikes are prepared in large batches of over 1000 units. The whole preparation process is carried out in a glove box and is a meticulous, time-consuming task.
An automated system to produce certified large size dried spikes, comparable to the spikes produced traditionally, has been considered for some time by IRMM [2]. The recently designed automated system consists of a robot, two balances, a dispenser and a drying unit fitted into a glove box. The robot is controlled by dedicated software which is developed to control all movements inside the glove-box, to identify unambiguously the penicillin vials with barcode reader, to dispense the LSD batch solution into vials and to weigh the amount dispensed.
An important step is to validate the system and show that it is capable of producing LSD spikes with the correct physical properties and, very importantly, to show that possible sources of uncertainty introduced by the automatic system are understood and of a small magnitude.
The system functionality has been evaluated and a validation of the automated system was accomplished by comparing the result from a series of sample aliquoted and weighed by the automated system with the result by manual substitution weighing. No significant difference is observed between the two datasets when applying the proper correction factors on the data from the automated system balance. To take this small correction into account an additional component of uncertainty of $3 \cdot 10^{-4}$ will be introduced in the uncertainty budget for the certified weights provided by the automatic system.

DESIGN OF THE AUTOMATED SYSTEM

The automated system was designed and built by Nucomat in close collaboration with IRMM. Nucomat is a company with a well recognized reputation as a system integrator providing customer tailored solutions to laboratory automation. Based on a modular concept of autonomous stations, combined with a real time scheduling architecture, Nucomat builds turnkey applications to customer specifications. These applications may incorporate equipment from various analytical instrument providers.
The automated system, as shown in Figure 1, consists of a robot with a gripper device (Figure 2) to pick up vials, two balances, a dispenser and a drying unit fitted into a glove-box. The robot is controlled by dedicated software which is developed to control all movements inside the glove-box, to unambiguously identify the penicillin vials with a barcode reader, to dispense the LSD batch solution into vials and to weigh the amount dispensed.

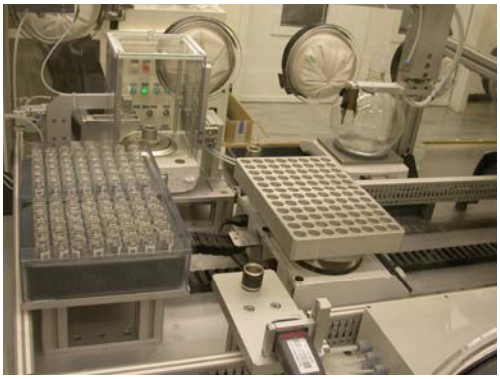

Figure 1: automated LSD production system

Robotic system

The central robot for vial transport is a 3-degrees-of-freedom Cartesian robot consisting of a belt-driven XY axis and a pneumatic Z axis to optimize usage of the workspace on the table. A pneumatic gripper placed on the eccentric arm of the Z axis can directly reach the balance pan. Robotic locations are derived from the 3D design drawings and no additional learning steps are required.

Distributed control

Specific hardware functions (robot, barcode reader, dispenser) are controlled by individual microcontroller modules named stations. Each of the stations communicates over an Ethernet port using UDP protocol with the central PC that performs the task distribution.

Nucomat Robin software

The LSD application program is based on the Nucomat Robin software architecture, an event driven command interpreter with underlying SQL server database. The operator interface uses Internet Explorer as a front end. Different operator levels can be configured to restrict parameter configuration to supervisors. Station communication and hardware status changes are continuously stored in databases for power failure recovery. Sample registration, robotic moves and stations dosing results are archived for traceability. The history of configuration changes is archived for validation purposes. During temporary downtime of the LIMS connection, the LSD system continues in stand-alone mode and will synchronise when the network connection re-establishes. Sample CLEANUP function is provided for robust and fault tolerant removal of samples.

Figure 2: gripper system

CONSIDERATIONS IN AUTOMATING THE PRODUCTION OF LSD SPIKES

During manual production of large series of LSD spikes there are a number of important constraints such as labour intensive manipulations in a glove-box requiring a lot of time spent in an uncomfortable position and also the exploitation license and regulations imposed to comply with the ALARA principle (as low as reasonably achievable) for exposure to radiation. The automated system was designed avoid such problems and to be capable of producing sets of certified nuclear isotopic reference material in the form of large size dried spikes, comparable to the spikes produced manually.
The requirements of the system were that the robot should be able to control all movements inside a glove-box, make an unambiguous identification of vials, dispense the LSD mother solution into vials and weigh the amount

dispensed with a sufficiently high degree of precision.
The development of the automated system to operate inside a glove box posed problems of a different nature such as maintaining the functionality of the system within a corrosive environment, the handling radioactive material and obviously the primary requirement of aliquoting 1200 units of 2.5 g into penicillin vials with uncertainty of less then $5 \cdot 10^{-4}$ on the certified mass.
Figure 3 shows the glove box in which the automated was installed.

Figure 3: glove box for automated system setup

In the initial design the electronic components inside the glove-box needed additional protection against the corrosive environment of nitric acid fumes. Special measures had to be taken to reduce fire hazard inside the glove-box, to avoid obstructions during manipulation and create sufficient storage place for aliquots prepared, to reduce radioactive waste produced and to allow easier replacement of electronic components.
A review by Nucomat in collaboration with IRMM resulted in major improvements to the design. The sensitive electronic parts were removed from the glove-box and placed in an external electrical cabinet. The electronic devices which remain inside the glove-box were protected in a closed box under an over-pressure by bleeding in compressed air required for the operation of the pneumatic system. The electronic parts were made easier to access. In addition a number of safety measures were installed such as alarm lights indicating the status of the system, over- and under-pressure alarms on the glove-box and manual emergency stop switches.

CALIBRATION OF SYSTEM BALANCE

Traceability to SI is ensured by weighing a reference weight, certified at IRMM against the IRMM kilogram, before and after filling a series of 96 vials, or at a higher frequency if needed. The reference weight is stored on a fixed position inside the box. Possible uncertainty contributions arise from the stability of the support cup on the balance pan, possible non-closure of the door of the weighing cabinet, air flow inside the box and the effect of vibrations within the glove-box.

VALIDATION OF FULL SYSTEM

The system was validated by dispensing and weighing 4 series of tests samples. In each series 2.5 g of mother solution was dispensed by the robot system into 10 preweighed vials. The vials were subsequently transferred into the mass metrology laboratory and weighed again using substitution weighing. The validation test consisted of comparing the results obtained from the system and those from the mass metrology service. The difference observed between the results from the balance in the automated system and those obtained by substitution weighing are shown for each series on Table 1.
A significant difference was observed after series 1 due to a mechanical problem arising from the balance door. After adapting the balance to correct for this, there still appeared to be a significant difference with series 2. A possible source of error was found to originate from the calibration or zero setting of the balance in the automated system which could not be performed properly. The weighing pan in the system contains a vial holder for positioning the penicillin vial. The correct zero adjustment and calibration can only be performed with an empty weighing pan. This problem was solved by the installation of a new pan balance.
Series 3 and series 4 show the dispensing with the correctly calibrated balance and adapted weighing pan. (A correction had to be applied to compensate the evaporation effect during time elapsing between Nucomat and mass metrology service weighing).
After evaluation there remained an unexplained difference of about 150 ppm or 5 scale divisions on the balance between the automated system and manual results after correction for all known sources of discrepancy such as balance adjustment and evaporation effects had been made.

A source of discrepancy needed to be considered which was related to a fundamental mass metrological concept. To make a direct readout from a balance comparable with the result of a substitution weighing, additional consideration has to given so that the balance readout is compensated for the assumed object density of 8000 $kg \cdot m^{-3}$.
Thus, instead of using the common balance equation

$$m_{display} = m_{sample} \cdot \left(1 - \frac{\rho_{air}}{\rho_{sample}} \right)$$

it is necessary to use the general balance equation and to use compensation for the balance manufacturer's assumed density of 8000 $kg \cdot m^{-3}$

$$m_{display} \cdot \left(1 - \frac{\rho_{air}}{8000} \right) = m_{sample} \cdot \left(1 - \frac{\rho_{air}}{\rho_{sample}} \right)$$

In this equation the factor the air density ρ_{air} = 1.2 $kg \cdot m^{-3}$. This additional factor in the equation is 150 ppm which explains the part of the difference between substitution weighing and direct weighing with the automated system.

(in mg)	A	B	C	D
Series 1	1.1	0.7	0.2	0.2
Series 2	0.9	0.5	0.0	0.1
Series 3	0.8	0.4	-0.1	0.1
Series 4	0.8	0.5	-0.0	0.3

Table 1

The columns A, B and C show the differences observed between the results from the balance in the automated system and those obtained by substitution weighing after applying proper corrections. A: buoyancy corrected, B: buoyancy and density corrected, C: evaporation effect corrected, D: standard uncertainty.

CONCLUSION

The system has proven to provide certified reference materials produced in a semi-automatic way in batches of 96 units. The uncertainties on the certified values of the reference materials delivered are comparable to those prepared manually.
Validation tests showed a bias in the weights of the dispensed spike aliquots between the automated system and mass metrology substitution weighing of less than $2 \cdot 10^{-4}$. The normal variability for a balance of the type used in the automated system is 3-5 scale divisions (0.3-0.5 mg).
Therefore no significant difference is observed between the two datasets on the condition of proper correction on the data from automated system balance. When evaluating or using weighing results for 2.5 g samples by the Nucomat system, an additional component of $3 \cdot 10^{-4}$ standard uncertainty will be introduced in the uncertainty budget due to the variability of the type of balances used.

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