



HAL
open science

Application of Machine Learning to MICADO Passive and Active Neutron Measurement System for the Characterization of Radioactive Waste Drums

Quentin Ducasse, Cyrille Eleon, Bertrand Perot, Nadia Perot, Pierre-Guy Allinei

► To cite this version:

Quentin Ducasse, Cyrille Eleon, Bertrand Perot, Nadia Perot, Pierre-Guy Allinei. Application of Machine Learning to MICADO Passive and Active Neutron Measurement System for the Characterization of Radioactive Waste Drums. IEEE Transactions on Nuclear Science, 2024, 71 (5), pp.1084 - 1090. 10.1109/tns.2024.3351275 . hal-04694745

HAL Id: hal-04694745

<https://hal.science/hal-04694745v1>

Submitted on 11 Sep 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License

AUTHOR QUERIES

AUTHOR PLEASE ANSWER ALL QUERIES

PLEASE NOTE: We cannot accept new source files as corrections for your article. If possible, please annotate the PDF proof we have sent you with your corrections and upload it via the Author Gateway. Alternatively, you may send us your corrections in list format. You may also upload revised graphics via the Author Gateway.

Carefully check the page proofs (and coordinate with all authors); additional changes or updates **WILL NOT** be accepted after the article is published online/print in its final form. Please check author names and affiliations, funding, as well as the overall article for any errors prior to sending in your author proof corrections.

AQ:1 = Please confirm or add details for any funding or financial support for the research of this article.

AQ:2 = Please confirm whether the edits made in the current affiliation of all the authors are correct.

AQ:3 = Please provide the title and accessed date for Refs. [1], [6], and [9].

AQ:4 = Please provide the organization name and organization location for Refs. [3], [4], and [5].

AQ:5 = Please provide the publisher name and publisher location for Ref. [8].

AQ:6 = Please provide the volume no. for Ref. [11].

AQ:7 = Please provide the page range for Ref. [14].

Application of Machine Learning to MICADO Passive and Active Neutron Measurement System for the Characterization of Radioactive Waste Drums

Quentin Ducasse¹, Cyrille Eleon¹, Bertrand Perot¹, Nadia Perot, and Pierre-Guy Allineï¹

Abstract—A passive and active neutron measurement system has been developed within the Measurement and Instrumentation for Cleaning and Decommissioning Operation (MICADO) H2020 project to estimate the nuclear material mass inside legacy waste drums of low and intermediate radioactivity levels. Monte–Carlo simulations were performed to design a transportable neutron system allowing both passive neutron coincidence counting and active interrogation with the differential die-away technique (DDT). However, the calibration coefficients (CCs) representing the signal of interest (due to nuclear material) in these two measurement modes may vary by a large amount depending on the properties of the matrix of the nuclear waste drum. Therefore, this article investigates matrix effects based on 104 Monte–Carlo calculations with different waste drums, based on Taguchi experimental design with a range of densities, material compositions, filling levels, and nuclear material masses. A matrix correction method is studied using machine learning algorithms. The matrix effect on the neutron signal is deduced from the signal of internal neutron monitors located inside the measurement cavity and from a transmission measurement with an AmBe neutron source. Those quantities can be assessed experimentally and are used as explanatory variables for the definition of a predictive model of the simulated CC, either in passive or in active mode. A multilinear regression model of the CC based on ordinary least square (OLS) is built and compared to the random forest (RF) machine-learning algorithm and to the multilayer perceptron (MLP) artificial neural network. In passive neutron coincidence counting, the residual error of the regression is lower for the MLP and RF than for OLS. The agreement between the predicted CCs of four mockup drums used as test is better than 17% and 3%, respectively, with the MLP and RF methods, while three predictions are out of the 95% confidence level range with OLS. In active neutron interrogation, similar conclusions are drawn. The prediction of the CC for the four mockup drums is

better than 12%, 35%, and 72% for the respective MLP, RF, and OLS methods. In conclusion, the MLP and RF regression model demonstrates more accurate results of the quantities of interest than the traditional OLS method. The future steps will focus on matrix heterogeneities, experimental validation, improving our models and testing new regression approaches.

Index Terms—Active neutron interrogation, calibration coefficient (CC), experimental design, linear regression, multilayer perceptron (MLP), passive neutron coincidence counting.

I. INTRODUCTION

THE Measurement and Instrumentation for Cleaning and Decommissioning Operations (MICADOs) project [1] of H2020 Research and Innovation Program aims to propose a cost-effective and comprehensive solution for nondestructive characterization of nuclear waste. The project is developing a platform composed of different measurements (gamma camera, gamma-ray spectroscopy, passive and active neutron measurements, and photofission interrogation) and of modern analysis technologies, such as AI and Bayesian methods, to combine experimental results in view of reducing the uncertainty in the determination of the nuclear material content inside the nuclear waste package. The neutron system aims at quantifying nuclear material (plutonium and uranium) in legacy technological wastes resulting from the exploitation of nuclear plants. A neutron system prototype was recently designed and optimized by Monte–Carlo simulations [2] using the MCNP code [3] for the nuclear material mass determination of a wide range of nuclear waste by combining both passive neutron coincidence counting [4], [5] and active neutron interrogation [5]. This work also showed that the neutron signal of interest coming from the nuclear material is strongly impacted by the properties of the nuclear waste drum matrix. Therefore, we investigate the matrix effects with Monte–Carlo calculations based on an experimental design to figure out possible corrections to determine more accurately the nuclear material mass.

II. MICADO NEUTRON SYSTEM MEASUREMENT

The design of a neutron measurement system is strongly influenced by the characteristics of the nuclear waste drums to

Manuscript received 28 September 2023; revised 14 December 2023; accepted 27 December 2023. This work was supported by the European Union's Horizon 2020 Research and Innovation Program under Grant 847641. (Corresponding author: Quentin Ducasse.)

Quentin Ducasse was with CEA, DES, IRESNE, Nuclear Measurement Laboratory, Cadarache, F-13108 Saint-Paul-lez-Durance, France. He is now with the Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSE-SANTE/SDOS/LMDN, Cadarache, F-13108 Saint-Paul-lez-Durance, France (e-mail: quentin.ducasse@irsn.fr).

Cyrille Eleon, Bertrand Perot, and Pierre-Guy Allineï are with CEA, DES, IRESNE, Nuclear Measurement Laboratory, Cadarache, F-13108 Saint-Paul-lez-Durance, France.

Nadia Perot is with CEA, DES, IRESNE, DER, SESI, LEMS, Cadarache, F-13108 Saint-Paul-lez-Durance, France.

Color versions of one or more figures in this article are available at <https://doi.org/10.1109/TNS.2024.3351275>.

Digital Object Identifier 10.1109/TNS.2024.3351275

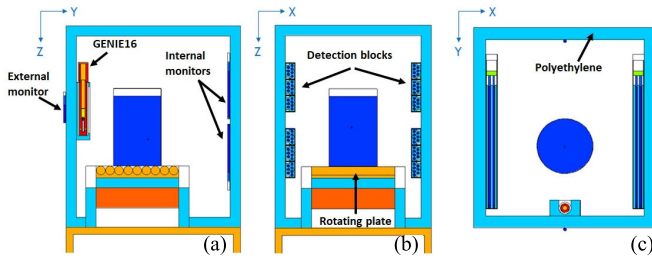


Fig. 1. MCNP model of the MICADO neutron system design. (a) Side view. (b) Front view. (c) Upper view.

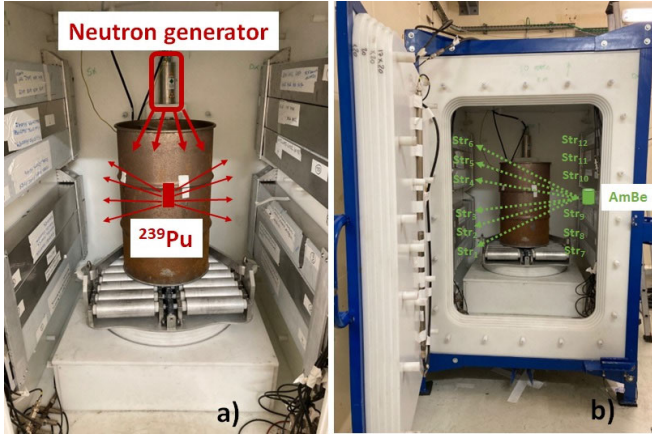


Fig. 2. Experimental setup of the MICADO neutron system. Measurement in active neutron interrogation using (a) neutron generator and (b) transmission.

of neutron absorbing materials (mainly stainless steel) and is covered by a layer of 10 cm of polyethylene to mitigate this effect. A 14-MeV DT neutron generator (GENIE16 from SODERN, [7]) used in the active mode is fixed on the wall on a polyethylene support. An additional boron-coated proportional counter called “external monitor,” located outside the neutron cell, is used to normalize all measurements and thus correct for potential fluctuations of the neutron generator emission rate.

The determination of the nuclear material mass in a drum derives from the value of a calibration coefficient (CC) representing the useful neutron signal obtained for 1 g of nuclear material in the drum, which is calculated by Monte-Carlo simulation [2]. This signal is highly affected by the properties of the matrix of the drum, namely, the presence of neutron thermalizing and/or absorbing materials that impact the useful signal in both passive neutron coincidence counting and active neutron interrogation. For instance, a matrix with rich-in-hydrogen materials, such as polyethylene, thermalizes generator fast neutrons and thus increases the fission rate, but neutrons absorbers, such as boron, cadmium, iron, or hydrogen itself, have the opposite effect. On the other hand, as hydrogen slows down the neutrons to be detected, coming from spontaneous or induced fissions, it reduces their detection efficiency. Indeed, thermalized neutrons are absorbed by the cadmium layer surrounding the detection blocks (see Fig. 1). To monitor thermal neutron absorbers, two boron-coated detectors (called “internal monitors,” see Fig. 1) are fixed at two different heights on the opening door of the neutron cell. They are sensitive to the thermal neutron flux inside the measurement cavity, which depends on the waste materials, and can be used to correct matrix effects in active neutron interrogation mode, in view to reduce the uncertainty on the nuclear mass estimation [10]. We also use the signal of an AmBe neutron source transmitted across the waste drum, with the opposite ^3He detection blocks (see Fig. 2), which is sensitive to the thermalization properties of the matrix. Both internal monitors and transmission signals are used to monitor matrix effects in passive neutron coincidence counting and in active neutron interrogation.

III. EXPERIMENTAL DESIGN

The investigation of matrix effects on the CC requires a large number of Monte-Carlo simulations based from the numerical model presented in Fig. 1 for a variety of nuclear waste drums. For this purpose, a Taguchi experimental design [8] is used to investigate how different matrix features affect the mean and variance of the CC variable of interest. The aim is to build a predictive model of CC for waste matrices with properties in the scope of the experimental design. The experimental design proposed by Taguchi involves orthogonal arrays to organize the parameters affecting the variable of interest and their levels of variation. Compared to a factorial design, the Taguchi method only tests pairs of combinations of parameters, allowing determining which ones most affect the variable of interest with a limited number of simulations. In this study, we perform the simulation for 104 waste drums of different matrix compositions, densities, filling heights, and nuclear material masses (see Table I).

be measured. Hundreds of thousands of nuclear waste packages (produced by EDF, ORANO, CEA, etc.) are currently stored in several areas in France [6]. We focus our study on 118 L technological legacy waste drums from nuclear fuel fabrication or spent fuel reprocessing plants, which contains plutonium and uranium. The MCNP numerical model of MICADO neutron system is based on the prototype described in [2], with only a few modifications to consider the measurement cell as built. Figs. 1 and 2 show the MCNP model and the experimental setup (not used in this article), respectively.

The neutron system consists of a $150 \times 170 \times 230$ cm cell that can contain drums up to 400 L. The 10-cm-thick walls of the cell are made of polyethylene to thermalize neutrons and thus increase the fission rate for the differential die-away technique (DDT) [5]. The neutron system prototype includes a total of 84 gas proportional counters filled with ^3He , embedded by groups of seven detectors in 12 polyethylene blocks, in order to thermalize the neutrons to be detected. These neutrons are fission prompt neutrons induced in fissile nuclei like ^{239}Pu and ^{235}U , in active neutron interrogation, and spontaneous fission neutrons from odd nuclei like ^{240}Pu , in passive neutron coincidence counting. The detection blocks are embedded in cadmium for the DDT technique. They are disposed horizontally on the two sidewalls of the system, to give an indication of the vertical location of neutron sources in the drum, in view to reduce uncertainties on the nuclear material mass determination. The drum is placed on a rotating plate made of aluminum that is relatively transparent to neutrons. The motor that commands the rotating plate is composed

TABLE I

PARAMETERS AND THEIR VARIATION LEVELS OF THE L32 AND THE L72 TAGUCHI EXPERIMENTAL DESIGNS FOR 118 L DRUMS

Parameters	L104 experimental design	
	L32 Taguchi	L72 Taguchi
Matrix composition (mass fractions)	Polyethylene (100 %) ; Stainless steel (100 %) ; Mixed 33% PE - 67 % steel ; Mixed 67 % PE - 33 % steel	Metallic ; Zircaloy (Zr alloy) ; MELOX (organic waste) ; C ₆ H ₁₀ O ₆ (50 %) – CH ₂ (50 %) ; PVC ; borated silicon carbide SiC (99 %) - B (1 %)
Density (g/cm³)	0.1 ; 0.3 ; 0.5 ; 0.7	0.1 ; 0.2 ; 0.35 ; 0.45 ; 0.6 ; 0.7
Filling level (%)	50 ; 65 ; 80 ; 100	50 ; 65 ; 80 ; 100
Nuclear material mass (g)	0.1 ; 1 ; 10 ; 50	0.1 ; 10 ; 100

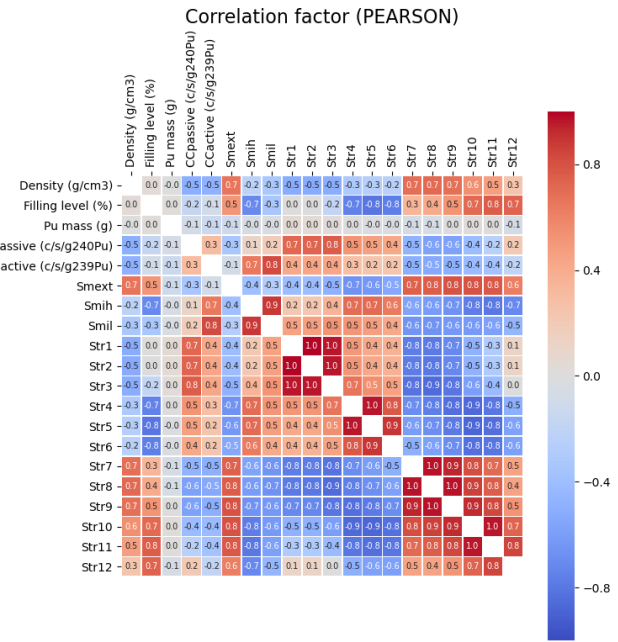


Fig. 3. Correlation matrix between the CC (CC ²³⁹Pu and CC ²⁴⁰Pu) and the explanatory variables (signal of the internal monitors Smih and Smil and neutron signal in the 12 ³He detection blocks Str_i measured with AmBe source).

Note that the nuclear material mass is not strictly speaking a matrix effect, but it is introduced in the study to take into account the combined self-absorption and multiplication effects in nuclear material. In the end, this combined effect was found to be negligible in this study because nuclear materials are distributed homogeneously in the whole matrix. The 104 simulations are split into an orthogonal array of 32 and 72 configurations, namely, L32 and L72 [9]. These two experimental designs have then been merged into a unique L104 experimental design to obtain a more robust database for the regression techniques described later in this article. The target value is the CC both in passive neutron coincidence counting and in active neutron interrogation of 118 L drums. The L32 and the L72 propose four and up to six levels of variation of their parameters, respectively. The parameters and their levels of variation fully cover the scope of characteristics defining the nuclear waste drums likely to be measured in the MICADO project. They are presented in Table I.

The experimental designs involve a large variety of matrices, mainly composed of metallic or organic elements present as primary components in technological wastes of nuclear plants (pipes, rods, gloves, etc.). In this study, the matrix and the distribution of nuclear materials inside the drum are assumed to be homogeneous. In practice, partial information of the drum characteristics might be collected from the drum provider before performing a neutron measurement. Information can also be assessed with complementary measurements [11] performed within the MICADO project (gamma spectroscopy and tomography). In the following, we will assume that no a priori information is known, so as not to introduce any bias in the analysis of the experimental design simulations.

IV. COMPARISON OF REGRESSION MODELS

A. Definition of the Explanatory Variables

The measured transmitted and internal monitor signals are used as explanatory variables to assess the properties of the waste matrix. The neutron signal transmitted through the drum is measured by the six ³He detection blocks (Str₁, Str₂, Str₃, Str₄, Str₅, and Str₆) located at the opposite side with respect to the AmBe source (Fig. 2). It reflects the thermalizing power of the waste matrix inside the drum. In passive neutron coincidence counting, only this neutron transmission information

is implemented in the regression models, as thermal neutron absorption in the waste matrix has a minor effect when using detection blocks wrapped in cadmium for the purpose of the active interrogation. For this last measurement, however, the signal measured by the two internal monitors inside the cell is used as additional explanatory variables in the regression models. Indeed, these internal monitors give information on the absorbing properties of the matrix for thermal interrogating neutrons, respectively, in the lower part (Smil) and in the higher part (Smih) of the drum. Fig. 3 shows the correlation matrix between the CC (in passive and active modes) and the explanatory variables. The CC in passive mode (CC_{passive}) is little correlated to the signal of the internal monitors, while it shows the highest correlation with the transmitted signal (Str₁–Str₃) located at the opposite side of the AmBe source. Finally, we can also observe the strong correlation (factor > 0.7) between CC in the active mode (CC_{active}) and internal monitors.

B. Regression Techniques

Regression techniques for the prediction of the CC can be applied using the explanatory variables showing the highest correlations with CC. In this work, we illustrate the benefit of new regression approaches with respect to multilinear regression with ordinary least squares (OLSs), which is traditionally used in our laboratory. We compare its results with the multilayer perceptron (MLP) [11] and random forest (RF) [12] algorithms from the Scikit-Learn Python library [13].

Linear regression with the OLS provides a straightforward and interpretable framework for estimating relationships between dependent and multiple independent variables. OLS provides an estimation of the linear model coefficients by



Fig. 4. Mockup drums modeled in MCNP. From left to right: wood–PVC–polyethylene–stainless steel.

minimizing the sum of squared differences between observed and predicted values of the CC (here, CC_{MCNP} and CC_{pred}). CC_{pred} is defined as an analytical function of quadratic and polynomial value (up to the second order) of the most relevant explanatory variables (see Section IV-A).

An MLP is a class of artificial neural networks characterized by multiple layers of interconnected nodes, or “neurons,” which process input data to produce desired outputs [13]. The efficacy of MLPs in regression modeling stems from their capacity to approximate highly nonlinear mappings between input and output variables. Unlike traditional multilinear regression models, MLPs can capture intricate patterns, interactions, and nonlinearity present in the data.

On the practical side, the optimization process involves defining hyperparameters, such as the number of hidden layers, the number of neurons for each hidden layer, the activation function, and the learning rate [13]. These are determined through a cross-validation process, which is exclusively applied to the experimental design L72 that presents a larger number of density and matrix levels in comparison to L32 (see Table I).

The RF algorithm is a powerful asset in predictive modeling, particularly for complex and high-dimensional datasets. By building a large set of decision trees and aggregating their predictions, RF enhances prediction accuracy, reduces overfitting, and provides valuable insights into variable importance. The hyperparameters include the number of trees, the criterion, the minimum number of samples required to split a node, and among others [13]. These are also determined through a cross-validation process, which is exclusively applied to the experimental design L72.

C. Tests of the Regression Models With Homogeneous Mockup Drums

In this work, the predictions of CC calculated by the regression models will be checked using calculations with the MCNP models of realistic homogenous waste drums. For this purpose, we use the MCNP models of four mockup drums filled with different matrices (see Fig. 4), which will be used in the experimental tests of MICADO cell at the Nuclear Measurement Laboratory of CEA Cadarache.

These four matrices are representatives, in terms of neutron moderation and absorption, of the materials constituting radioactive technological wastes. In addition, their characteristics are included in the range of the parameters of the experimental design regarding matrix composition, density, filling level, and nuclear mass content (see Table II).

In the following, the unique L104 experimental design (as described in Section III) is directly used for the training

TABLE II
118 L MOCKUP DRUMS DESCRIPTION

Designation	Matrix material	Composition	Apparent density ($\text{g}\cdot\text{cm}^{-3}$)	Filling level
Mockup 1	Stainless steel	Fe (70%) – Ni (18%) – Cr (8%)	0.63	82%
Mockup 2	Wood	$\text{C}_6\text{H}_{10}\text{O}_5$	0.35	95%
Mockup 3	Polyethylene	CH_2	0.5	92%
Mockup 4	PVC	$\text{C}_2\text{H}_3\text{Cl}$	0.27	91%

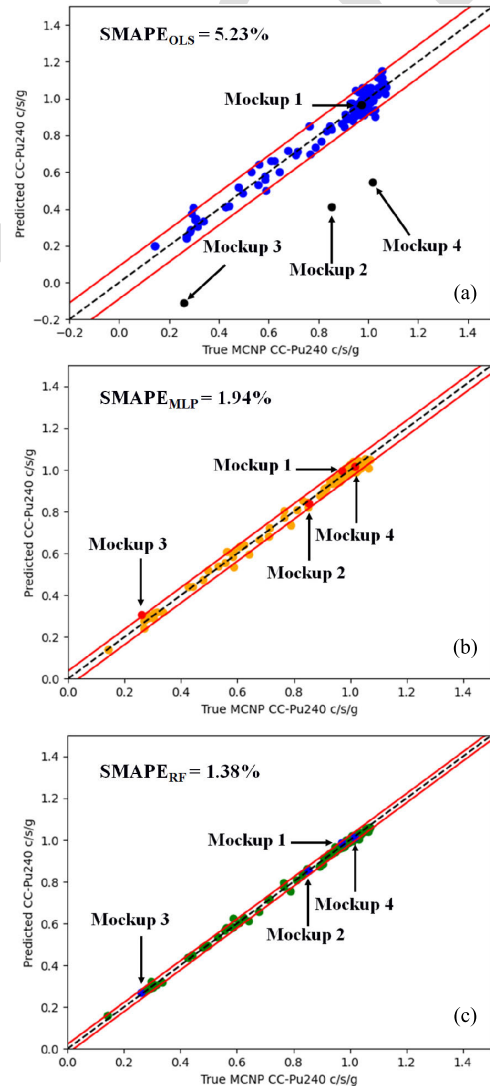


Fig. 5. Predicted CC-240Pu of the mockup cases (labeled points) in passive neutron coincidence counting obtained with (a) OLS, (b) MLP, and (c) RF algorithms. The two red lines are the interval $\pm 2 \sigma_{res}$.

database, and the prediction performance of the regression techniques is assessed using simulation results performed for the four mockup drums. The result of the regression models (called predicted CC or “ CC_{pred} ”) is compared to the MCNP simulated CCs (called “ CC_{true} ”) for the 104 configurations in Figs. 5 and 6 for passive neutron coincidence counting and for active neutron interrogation, respectively. The predicted CC and the true CC of the four mockup drums are also indicated.

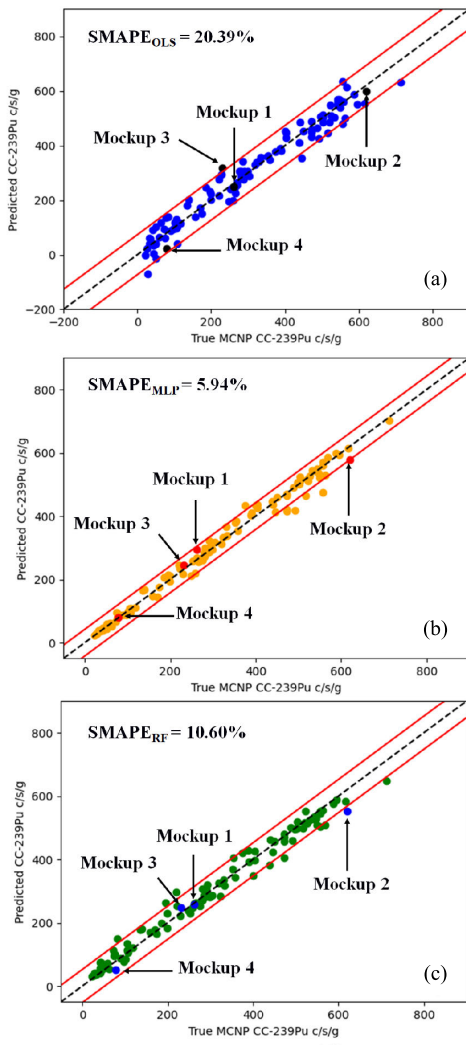


Fig. 6. Predicted CC-239Pu of the mockup cases (labeled points) in active neutron interrogation obtained with (a) OLS, (b) MLP, and (c) RF. The two red lines are the interval $\pm 2 \sigma_{\text{res}}$.

The predicted CC (CC_{pred}) by the three regression methods is represented against the simulated (CC_{true}). The residual error of the multilinear regression with the interval is $\pm 2 \sigma_{\text{res}}$. The absolute value of σ_{res} is deduced from the square root of the mean square error (mse). Values covered by the CC are largely distributed and are not organized in clusters, demonstrating the relevance of the description of the experimental design. The symmetric mean absolute percentage error (SMAPE) demonstrates a better quality of the regression using MLP and RF methods compared to OLS, in both passive coincidence counting (SMAPE_{OLS} = 5.23%, $\sigma_{\text{res,OLS}}$ = 0.0446 c.s⁻¹.g⁻¹; SMAPE_{MLP} = 1.94%, $\sigma_{\text{res,MLP}}$ = 0.0185 c.s⁻¹.g⁻¹; SMAPE_{RF} = 1.38%, $\sigma_{\text{res,RF}}$ = 0,0111 c.s⁻¹.g⁻¹) and active interrogation (SMAPE_{OLS} = 20.39%, $\sigma_{\text{res,OLS}}$ = 37 c.s⁻¹.g⁻¹; SMAPE_{MLP} = 5.94%, $\sigma_{\text{res,MLP}}$ = 21 c.s⁻¹.g⁻¹; SMAPE_{RF} = 10.60%, $\sigma_{\text{res,RF}}$ = 26 c.s⁻¹.g⁻¹). Additionally, a few negative, nonphysical values of CC_{pred} arise from the linear regression [“Mockup 3” on Fig. 5 and a few others (blue points) on Fig. 6], disqualifying the validity of the OLS method in this region. Table III compares the metrics of

TABLE III

COMPARISON METRICS OF THE DIFFERENT MULTILINEAR REGRESSION MODELS IN PASSIVE NEUTRON COINCIDENCE COUNTING AND ACTIVE NEUTRON INTERROGATION FOR THE FOUR MOCKUP DRUMS

	Mock-up number	CC_{true} (MCN P) [c.s ⁻¹ .g ⁻¹]	$CC_{\text{pred,OLS}}$ (c.s ⁻¹ .g ⁻¹ and relative difference in %)	$CC_{\text{pred,MLP}}$ (c.s ⁻¹ .g ⁻¹ and relative difference in %)	$CC_{\text{pred,RF}}$ (c.s ⁻¹ .g ⁻¹ and relative difference in %)
Passive neutron coincidence counting	Mock-up 1	0.972	0.964 (-1%)	0.995 (2%)	0.986 (1%)
	Mock-up 2	0.851	0.411 (-52%)	0.842 (-1%)	0.854 (0%)
	Mock-up 3	0.261	0.110 (-58%)	0.306 (17%)	0.270 (3%)
	Mock-up 4	1.017	0.545 (-46%)	1.017 (0%)	1.011 (-1%)
Active neutron coincidence counting	Mock-up 1	262	250 (-5%)	294 (12%)	259 (-1%)
	Mock-up 2	621	600 (-3%)	579 (-7%)	553 (-11%)
	Mock-up 3	231	319 (38%)	245 (6%)	248 (7%)
	Mock-up 4	79	22 (-72%)	80 (1%)	51 (-35%)

accuracy for CC_{pred} using the OLS, MLP, and RF methods in passive coincidence counting and active interrogation.

In the passive mode, results indicate that the agreement in the predicted CC in the four mockup drums lies in the [1%–58%], [0%–17%], and [0%–3%] ranges in passive coincidence counting and in the [3%–72%], [1%–12%], and [1%–35%] ranges in active interrogation for the OLS, MLP, and RF methods, respectively. The model based on the OLS method provides poor prediction results in passive coincidence counting, where three of the four drum cases are outside the interval $\pm 2 \sigma_{\text{res}}$. This indicates an overfitting of the OLS model and reducing the amount of input parameters, as well as the polynomial order, enables to mitigate this disagreement (while nonetheless degrading the residual error of the fit). On the other hand, the MLP and RF methods forecast all the results inside the interval $\pm 2 \sigma_{\text{res}}$. In active neutron interrogation, a better accuracy and consistency of the predictions is also observed for the MLP and RF methods.

D. Model Prediction Extension to Heterogeneous Waste Cases

To assess the validity and limitations of the prediction regression models established with homogenous waste matrices, ORANO La Hague provided two realistic heterogeneous drums case studies, together with their detailed Monte–Carlo simulation model, in the frame of MICADO project [14]. The first realistic case (RC1) consists of a 118 L alpha waste drum divided into five bags containing decontamination wipes. The activity of the plutonium inside the drum is heterogeneous. The matrix is also moderately heterogeneous and exclusively constituted of humid plastic C₃H₆(H₂O) of densities varying from 0.32 to 0.53. The second realistic case (RC2) is a 118 L alpha waste drum containing 50 stainless steel pipes with a low contamination level. The average density is 1.08 g.cm⁻³ and about 87% of the plutonium activity is concentrated in a valve located at the bottom of the drum. Fig. 7 shows a view of the RC1 and RC2 Monte–Carlo models.

The result of the regression models trained with the L104 data set is illustrated in Figs. 8 and 9, in passive neutron coincidence counting and in active neutron interrogation, respectively.

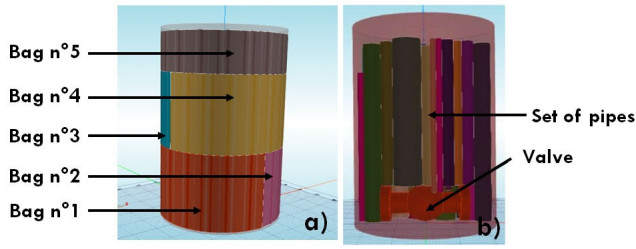


Fig. 7. MCNP models of (a) RC1 and (b) RC2 waste drums. The different colors correspond to different types of material.

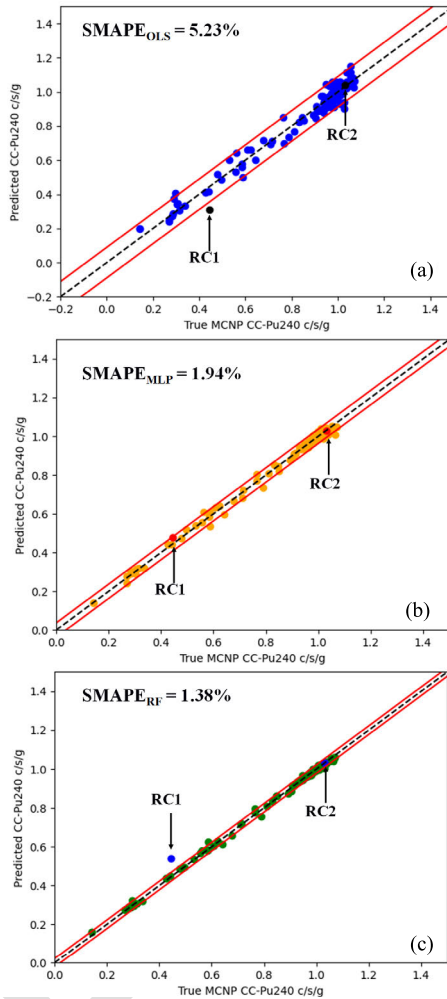


Fig. 8. Predicted CC-240Pu of the realistic drum cases (labeled points) in passive neutron coincidence counting obtained by (a) OLS, (b) MLP, and (c) RF algorithms. The two red lines are the interval $\pm 2 \sigma_{res}$.

348 Table IV compares the accuracy metric results for CC_{pred}
349 using the OLS, MLP, and RF methods in passive coincidence
350 counting and active interrogation.

351 As previously mentioned, both drums present certain level
352 of heterogeneities (matrix and distribution of the plutonium
353 mass) that was not simulated in the experimental design
354 calculations. Thus, the regression algorithms have not been
355 trained with heterogeneous matrices and nuclear material
356 distributions. This being said, CC_{pred} agrees within [0%–30%]
357 in passive neutron coincidence counting, regardless of the
358 regression method, but results are significantly better using

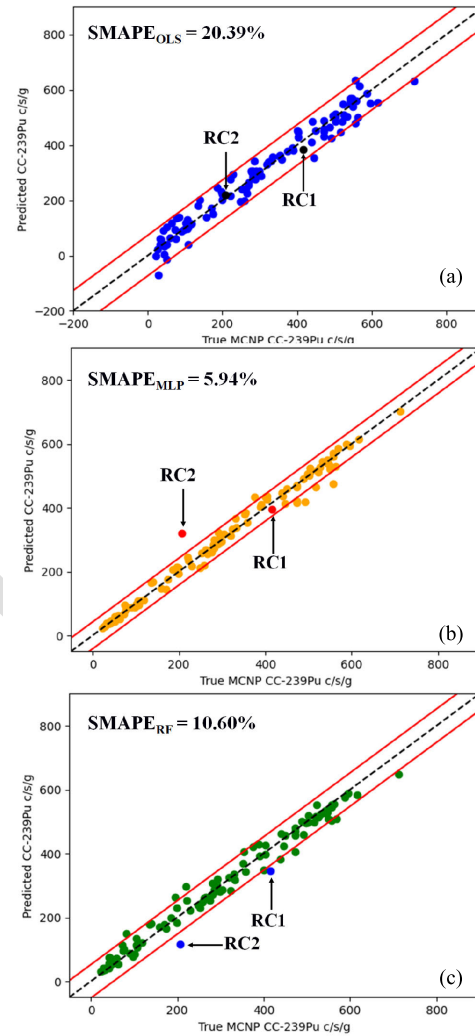


Fig. 9. Predicted CC-239Pu of the realistic drum cases (labeled points) in active neutron interrogation obtained by (a) OLS method, (b) MLP, and (c) RF algorithm. The two red lines are the interval $\pm 2 \sigma_{res}$.

TABLE IV
COMPARISON METRIC RESULTS OF THE DIFFERENT REGRESSION MODELS
IN PASSIVE NEUTRON COINCIDENCE COUNTING AND ACTIVE
NEUTRON INTERROGATION FOR THE
REALISTIC DRUM CASES

	Mock-up number	CC_{true} (MCN P) [$c \cdot s^{-1} \cdot g^{-1}$]	$CC_{pred,OLS}$ ($c \cdot s^{-1} \cdot g^{-1}$ and relative difference in %)	$CC_{pred,MLP}$ ($c \cdot s^{-1} \cdot g^{-1}$ and relative difference in %)	$CC_{pred,RF}$ ($c \cdot s^{-1} \cdot g^{-1}$ and relative difference in %)
Passive neutron coincidence counting	RC 1	0.445	0.310 (-30%)	0.477 (7%)	0.539 (21%)
	RC 2	1.032	1.039 (1%)	1.027 (0%)	1.033 (0%)
Active neutron coincidence counting	RC 1	416	383 (-8%)	394 (-5%)	345 (17%)
	RC 2	207	220 (6%)	321 (55%)	115 (-44%)

359 MLP and RF than OLS. In active neutron interrogation,
360 predictions are within [5%–55%] and OLS provides better
361 predictions of CC for RC2 (6% against 55% and 44% for
362 the MLP and RF methods, respectively). Beyond the fact that

RC2 shows heterogeneities in matrix composition and nuclear mass distribution, these poor predictions for MLP and RF algorithms are probably due to the average density of RC2, which is out of the boundary limits defined in the experimental design (i.e., 1.08 versus 0.7 g.cm⁻³ maximum in the training set). Overall predicted results of realistic heterogeneous drum cases are thus logically less accurate than the prediction for the previous homogeneous mockup drums.

E. Conclusion and Outlooks

The MICADO neutron measurement system prototype has been designed by MCNP simulation, with the objective to estimate the nuclear material mass in a wide range of radioactive waste drums by passive neutron coincidence counting and active neutron interrogation. The useful neutron signal (namely, the CC in count per second and per gram of ²⁴⁰Pu or ²³⁹Pu, respectively) was calculated by Monte-Carlo simulations for a series of matrix compositions, density, and filling levels defined with an experimental design. The obtained 104 simulation results were used to establish regression models with three different algorithms to reduce the uncertainty on the nuclear mass estimation. These models were based on internal matrix monitors and a neutron transmission measurement. Within the context of this issue, MLP and RF regression techniques show a clear advantage over conventional OLS (linear regression with OLSs), both in passive coincidence counting and active interrogation techniques. This behavior may be attributed to the strong correlation between some of the predictors knowing that one of the conditions to use classical linear regression method is the independence of the predictors. The residual error and the SMAPE on the linear regression of the MLP and RF techniques are lower, and the predicted CC never results in nonphysical negative values. Therefore, the uncertainty on the determination of the nuclear mass can significantly be reduced by comparison to standard OLS method usually employed in our laboratory.

In the next steps, we will investigate more regression methods and we will enlarge the training database with additional Monte-Carlo simulations, including more waste materials and heterogeneous matrices as well. Currently, the use of experimental designs helps to reduce the number of MCNP simulations while providing maximum information for constructing regression models. However, this approach only works if the range of variation for influential parameters (density, filling level, matrix material, etc.) covers the entire range of waste barrels to be measured. When dealing with homogeneous matrices, this approach is relatively robust. However, accounting for matrix heterogeneities and radioactive materials becomes very challenging, even when using

experimental designs driven by MCNP simulations. At present, the idea is to consider extreme cases in terms of heterogeneities. These cases are unlikely to be measured in practice, but it allows to cover a broad range of variations. Despite this, it is still expected that regression algorithms (based on interpolation between different points of the experimental design) will exhibit poorer performance compared to homogeneous cases. In this specific scenario, simulating all the possible heterogeneous configurations to train the models is very complex, if not impossible. Work is ongoing to evaluate the performance for these heterogeneous waste scenarios. Another important prospect is to perform laboratory measurements with mockup drums in DANAIDES casemate of TOTEM nuclear facility, at CEA Cadarache, both to validate our numerical Monte-Carlo calculation models and to test with experimental data the regression models established with simulation data.

ACKNOWLEDGMENT

The work presented in this article reflects only the author's views and the Commission is not liable for any use that may be made of the information contained therein.

REFERENCES

- [1] [Online]. Available: <https://www.micado-project.eu/>
- [2] Q. Ducasse et al., "Design of MICADO advanced passive and active neutron measurement system for radioactive waste drums," *Nucl. Instrum. Methods Phys. Res. A, Accel. Spectrom. Detect. Assoc. Equip.*, vol. 1005, Jul. 2021, Art. no. 165398.
- [3] *MCNP User's Manual Code Version 6.2*, Oct. 2017.
- [4] N. Ensslin, "Principles of neutron coincidence counters," Tech. Rep., LA-UR-90-732, 1991.
- [5] J. T. Caldwell, "The Los Alamos second-generation system for passive and active neutron assays of drum-size containers," Tech. Rep., LA-10774-MS, 1986.
- [6] [Online]. Available: <https://inventaire.andra.fr/>
- [7] *Sodern Tubes and Generators*. Accessed: Sep. 2023. [Online]. Available: <https://sodern.com/en/tubes-neutroniques/>
- [8] R. Sabre, "Taguchi experimental design," in *Engineering Techniques*, Mar. 2007.
- [9] [Online]. Available: <https://support.minitab.com/fr-fr/minitab/18/help-and-how-to/modeling-statistics/doe/supportingtopics/taguchi-designs/catalogue-of-taguchi-designs/#132-21-49>
- [10] L. Lepore et al., "The MICADO integrated gamma station for radioactive waste packages radiological characterization," in *Proc. EPJ Web Conf.*, vol. 288, 2023, p. 07015, doi: [10.1051/epjconf/202328807015](https://doi.org/10.1051/epjconf/202328807015).
- [11] G. E. Hinton, "Connectionist learning procedures," *Mach. Learn.*, pp. 555–610, Jan. 1990, doi: [10.1016/B978-0-08-051055-2.50029-8](https://doi.org/10.1016/B978-0-08-051055-2.50029-8).
- [12] L. Breiman, "Random forests," *Mach. Learn.*, vol. 451, pp. 5–32, Oct. 2001, doi: [10.1023/A:1010933404324](https://doi.org/10.1023/A:1010933404324).
- [13] F. Pedregosa et al., "Scikit-learn: Machine learning in Python," *J. Mach. Learn. Res.*, vol. 12, pp. 2825–2830, Nov. 2011.
- [14] E. Breuil and A. Bielen, "WP2: Virtual cases & WP8: Combination of measurements and uncertainty assessment," in *Proc. Micado Final Demonstration Conf.*, Jan. 2023.

AUTHOR QUERIES

AUTHOR PLEASE ANSWER ALL QUERIES

PLEASE NOTE: We cannot accept new source files as corrections for your article. If possible, please annotate the PDF proof we have sent you with your corrections and upload it via the Author Gateway. Alternatively, you may send us your corrections in list format. You may also upload revised graphics via the Author Gateway.

Carefully check the page proofs (and coordinate with all authors); additional changes or updates **WILL NOT** be accepted after the article is published online/print in its final form. Please check author names and affiliations, funding, as well as the overall article for any errors prior to sending in your author proof corrections.

AQ:1 = Please confirm or add details for any funding or financial support for the research of this article.

AQ:2 = Please confirm whether the edits made in the current affiliation of all the authors are correct.

AQ:3 = Please provide the title and accessed date for Refs. [1], [6], and [9].

AQ:4 = Please provide the organization name and organization location for Refs. [3], [4], and [5].

AQ:5 = Please provide the publisher name and publisher location for Ref. [8].

AQ:6 = Please provide the volume no. for Ref. [11].

AQ:7 = Please provide the page range for Ref. [14].

Application of Machine Learning to MICADO Passive and Active Neutron Measurement System for the Characterization of Radioactive Waste Drums

Quentin Ducasse¹, Cyrille Eleon¹, Bertrand Perot¹, Nadia Perot, and Pierre-Guy Allineci¹

Abstract—A passive and active neutron measurement system has been developed within the Measurement and Instrumentation for Cleaning and Decommissioning Operation (MICADO) H2020 project to estimate the nuclear material mass inside legacy waste drums of low and intermediate radioactivity levels. Monte–Carlo simulations were performed to design a transportable neutron system allowing both passive neutron coincidence counting and active interrogation with the differential die-away technique (DDT). However, the calibration coefficients (CCs) representing the signal of interest (due to nuclear material) in these two measurement modes may vary by a large amount depending on the properties of the matrix of the nuclear waste drum. Therefore, this article investigates matrix effects based on 104 Monte–Carlo calculations with different waste drums, based on Taguchi experimental design with a range of densities, material compositions, filling levels, and nuclear material masses. A matrix correction method is studied using machine learning algorithms. The matrix effect on the neutron signal is deduced from the signal of internal neutron monitors located inside the measurement cavity and from a transmission measurement with an AmBe neutron source. Those quantities can be assessed experimentally and are used as explanatory variables for the definition of a predictive model of the simulated CC, either in passive or in active mode. A multilinear regression model of the CC based on ordinary least square (OLS) is built and compared to the random forest (RF) machine-learning algorithm and to the multilayer perceptron (MLP) artificial neural network. In passive neutron coincidence counting, the residual error of the regression is lower for the MLP and RF than for OLS. The agreement between the predicted CCs of four mockup drums used as test is better than 17% and 3%, respectively, with the MLP and RF methods, while three predictions are out of the 95% confidence level range with OLS. In active neutron interrogation, similar conclusions are drawn. The prediction of the CC for the four mockup drums is

better than 12%, 35%, and 72% for the respective MLP, RF, and OLS methods. In conclusion, the MLP and RF regression model demonstrates more accurate results of the quantities of interest than the traditional OLS method. The future steps will focus on matrix heterogeneities, experimental validation, improving our models and testing new regression approaches.

Index Terms—Active neutron interrogation, calibration coefficient (CC), experimental design, linear regression, multilayer perceptron (MLP), passive neutron coincidence counting.

I. INTRODUCTION

THE Measurement and Instrumentation for Cleaning and Decommissioning Operations (MICADOs) project [1] of H2020 Research and Innovation Program aims to propose a cost-effective and comprehensive solution for nondestructive characterization of nuclear waste. The project is developing a platform composed of different measurements (gamma camera, gamma-ray spectroscopy, passive and active neutron measurements, and photofission interrogation) and of modern analysis technologies, such as AI and Bayesian methods, to combine experimental results in view of reducing the uncertainty in the determination of the nuclear material content inside the nuclear waste package. The neutron system aims at quantifying nuclear material (plutonium and uranium) in legacy technological wastes resulting from the exploitation of nuclear plants. A neutron system prototype was recently designed and optimized by Monte–Carlo simulations [2] using the MCNP code [3] for the nuclear material mass determination of a wide range of nuclear waste by combining both passive neutron coincidence counting [4], [5] and active neutron interrogation [5]. This work also showed that the neutron signal of interest coming from the nuclear material is strongly impacted by the properties of the nuclear waste drum matrix. Therefore, we investigate the matrix effects with Monte–Carlo calculations based on an experimental design to figure out possible corrections to determine more accurately the nuclear material mass.

II. MICADO NEUTRON SYSTEM MEASUREMENT

The design of a neutron measurement system is strongly influenced by the characteristics of the nuclear waste drums to

Manuscript received 28 September 2023; revised 14 December 2023; accepted 27 December 2023. This work was supported by the European Union’s Horizon 2020 Research and Innovation Program under Grant 847641. (Corresponding author: Quentin Ducasse.)

Quentin Ducasse was with CEA, DES, IRESNE, Nuclear Measurement Laboratory, Cadarache, F-13108 Saint-Paul-lez-Durance, France. He is now with the Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSE-SANTE/SDOS/LMDN, Cadarache, F-13108 Saint-Paul-lez-Durance, France (e-mail: quentin.ducasse@irsn.fr).

Cyrille Eleon, Bertrand Perot, and Pierre-Guy Allineci are with CEA, DES, IRESNE, Nuclear Measurement Laboratory, Cadarache, F-13108 Saint-Paul-lez-Durance, France.

Nadia Perot is with CEA, DES, IRESNE, DER, SESI, LEMS, Cadarache, F-13108 Saint-Paul-lez-Durance, France.

Color versions of one or more figures in this article are available at <https://doi.org/10.1109/TNS.2024.3351275>.

Digital Object Identifier 10.1109/TNS.2024.3351275

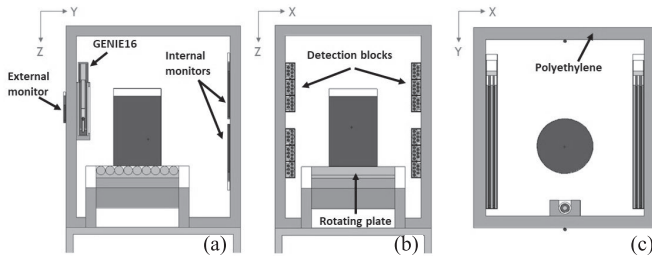


Fig. 1. MCNP model of the MICADO neutron system design. (a) Side view. (b) Front view. (c) Upper view.

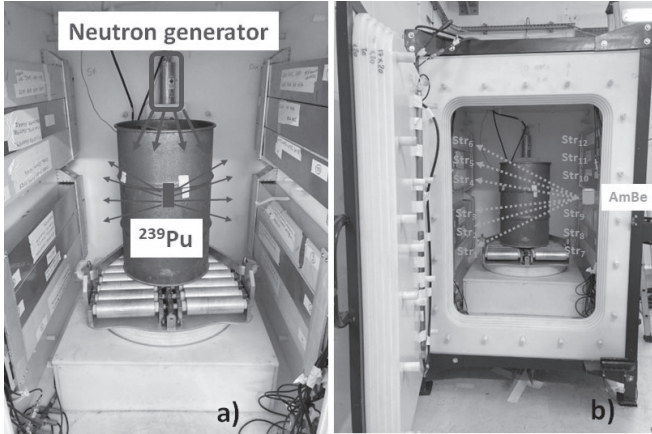


Fig. 2. Experimental setup of the MICADO neutron system. Measurement in active neutron interrogation using (a) neutron generator and (b) transmission.

of neutron absorbing materials (mainly stainless steel) and is covered by a layer of 10 cm of polyethylene to mitigate this effect. A 14-MeV DT neutron generator (GENIE16 from SODERN, [7]) used in the active mode is fixed on the wall on a polyethylene support. An additional boron-coated proportional counter called “external monitor,” located outside the neutron cell, is used to normalize all measurements and thus correct for potential fluctuations of the neutron generator emission rate.

The determination of the nuclear material mass in a drum derives from the value of a calibration coefficient (CC) representing the useful neutron signal obtained for 1 g of nuclear material in the drum, which is calculated by Monte-Carlo simulation [2]. This signal is highly affected by the properties of the matrix of the drum, namely, the presence of neutron thermalizing and/or absorbing materials that impact the useful signal in both passive neutron coincidence counting and active neutron interrogation. For instance, a matrix with rich-in-hydrogen materials, such as polyethylene, thermalizes generator fast neutrons and thus increases the fission rate, but neutrons absorbers, such as boron, cadmium, iron, or hydrogen itself, have the opposite effect. On the other hand, as hydrogen slows down the neutrons to be detected, coming from spontaneous or induced fissions, it reduces their detection efficiency. Indeed, thermalized neutrons are absorbed by the cadmium layer surrounding the detection blocks (see Fig. 1). To monitor thermal neutron absorbers, two boron-coated detectors (called “internal monitors,” see Fig. 1) are fixed at two different heights on the opening door of the neutron cell. They are sensitive to the thermal neutron flux inside the measurement cavity, which depends on the waste materials, and can be used to correct matrix effects in active neutron interrogation mode, in view to reduce the uncertainty on the nuclear mass estimation [10]. We also use the signal of an AmBe neutron source transmitted across the waste drum, with the opposite ^3He detection blocks (see Fig. 2), which is sensitive to the thermalization properties of the matrix. Both internal monitors and transmission signals are used to monitor matrix effects in passive neutron coincidence counting and in active neutron interrogation.

III. EXPERIMENTAL DESIGN

The investigation of matrix effects on the CC requires a large number of Monte-Carlo simulations based from the numerical model presented in Fig. 1 for a variety of nuclear waste drums. For this purpose, a Taguchi experimental design [8] is used to investigate how different matrix features affect the mean and variance of the CC variable of interest. The aim is to build a predictive model of CC for waste matrices with properties in the scope of the experimental design. The experimental design proposed by Taguchi involves orthogonal arrays to organize the parameters affecting the variable of interest and their levels of variation. Compared to a factorial design, the Taguchi method only tests pairs of combinations of parameters, allowing determining which ones most affect the variable of interest with a limited number of simulations. In this study, we perform the simulation for 104 waste drums of different matrix compositions, densities, filling heights, and nuclear material masses (see Table I).

be measured. Hundreds of thousands of nuclear waste packages (produced by EDF, ORANO, CEA, etc.) are currently stored in several areas in France [6]. We focus our study on 118 L technological legacy waste drums from nuclear fuel fabrication or spent fuel reprocessing plants, which contains plutonium and uranium. The MCNP numerical model of MICADO neutron system is based on the prototype described in [2], with only a few modifications to consider the measurement cell as built. Figs. 1 and 2 show the MCNP model and the experimental setup (not used in this article), respectively.

The neutron system consists of a $150 \times 170 \times 230$ cm cell that can contain drums up to 400 L. The 10-cm-thick walls of the cell are made of polyethylene to thermalize neutrons and thus increase the fission rate for the differential die-away technique (DDT) [5]. The neutron system prototype includes a total of 84 gas proportional counters filled with ^3He , embedded by groups of seven detectors in 12 polyethylene blocks, in order to thermalize the neutrons to be detected. These neutrons are fission prompt neutrons induced in fissile nuclei like ^{239}Pu and ^{235}U , in active neutron interrogation, and spontaneous fission neutrons from odd nuclei like ^{240}Pu , in passive neutron coincidence counting. The detection blocks are embedded in cadmium for the DDT technique. They are disposed horizontally on the two sidewalls of the system, to give an indication of the vertical location of neutron sources in the drum, in view to reduce uncertainties on the nuclear material mass determination. The drum is placed on a rotating plate made of aluminum that is relatively transparent to neutrons. The motor that commands the rotating plate is composed

TABLE I

PARAMETERS AND THEIR VARIATION LEVELS OF THE L32 AND THE L72 TAGUCHI EXPERIMENTAL DESIGNS FOR 118 L DRUMS

Parameters	L104 experimental design	
	L32 Taguchi	L72 Taguchi
Matrix composition (mass fractions)	Polyethylene (100 %) ; Stainless steel (100 %) ; Mixed 33% PE - 67 % steel ; Mixed 67 % PE - 33 % steel	Metallic ; Zircaloy (Zr alloy) ; MELOX (organic waste) ; C ₆ H ₁₀ O ₆ (50 %) – CH ₂ (50 %) ; PVC ; borated silicon carbide SiC (99 %) - B (1 %)
Density (g/cm ³)	0.1 ; 0.3 ; 0.5 ; 0.7	0.1 ; 0.2 ; 0.35 ; 0.45 ; 0.6 ; 0.7
Filling level (%)	50 ; 65 ; 80 ; 100	50 ; 65 ; 80 ; 100
Nuclear material mass (g)	0.1 ; 1 ; 10 ; 50	0.1 ; 10 ; 100

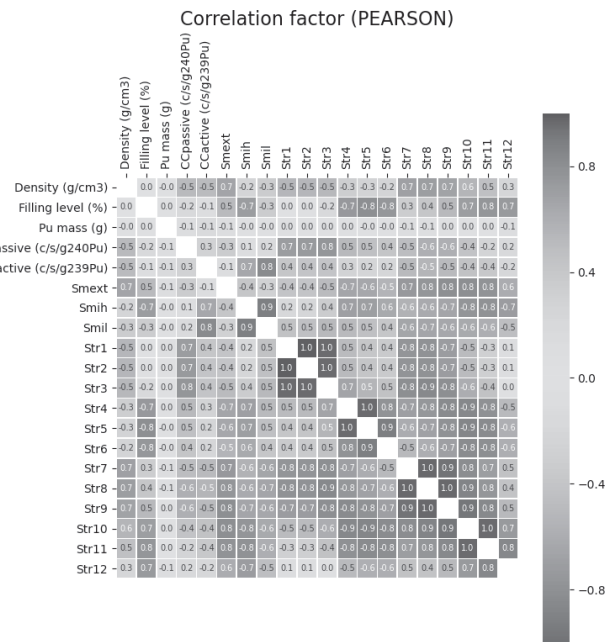


Fig. 3. Correlation matrix between the CC (CC ²³⁹Pu and CC ²⁴⁰Pu) and the explanatory variables (signal of the internal monitors Smih and Smil and neutron signal in the 12 ³He detection blocks Str_i measured with AmBe source).

Note that the nuclear material mass is not strictly speaking a matrix effect, but it is introduced in the study to take into account the combined self-absorption and multiplication effects in nuclear material. In the end, this combined effect was found to be negligible in this study because nuclear materials are distributed homogeneously in the whole matrix. The 104 simulations are split into an orthogonal array of 32 and 72 configurations, namely, L32 and L72 [9]. These two experimental designs have then been merged into a unique L104 experimental design to obtain a more robust database for the regression techniques described later in this article. The target value is the CC both in passive neutron coincidence counting and in active neutron interrogation of 118 L drums. The L32 and the L72 propose four and up to six levels of variation of their parameters, respectively. The parameters and their levels of variation fully cover the scope of characteristics defining the nuclear waste drums likely to be measured in the MICADO project. They are presented in Table I.

The experimental designs involve a large variety of matrices, mainly composed of metallic or organic elements present as primary components in technological wastes of nuclear plants (pipes, rods, gloves, etc.). In this study, the matrix and the distribution of nuclear materials inside the drum are assumed to be homogeneous. In practice, partial information of the drum characteristics might be collected from the drum provider before performing a neutron measurement. Information can also be assessed with complementary measurements [11] performed within the MICADO project (gamma spectroscopy and tomography). In the following, we will assume that no a priori information is known, so as not to introduce any bias in the analysis of the experimental design simulations.

IV. COMPARISON OF REGRESSION MODELS

A. Definition of the Explanatory Variables

The measured transmitted and internal monitor signals are used as explanatory variables to assess the properties of the waste matrix. The neutron signal transmitted through the drum is measured by the six ³He detection blocks (Str₁, Str₂, Str₃, Str₄, Str₅, and Str₆) located at the opposite side with respect to the AmBe source (Fig. 2). It reflects the thermalizing power of the waste matrix inside the drum. In passive neutron coincidence counting, only this neutron transmission information

is implemented in the regression models, as thermal neutron absorption in the waste matrix has a minor effect when using detection blocks wrapped in cadmium for the purpose of the active interrogation. For this last measurement, however, the signal measured by the two internal monitors inside the cell is used as additional explanatory variables in the regression models. Indeed, these internal monitors give information on the absorbing properties of the matrix for thermal interrogating neutrons, respectively, in the lower part (Smil) and in the higher part (Smih) of the drum. Fig. 3 shows the correlation matrix between the CC (in passive and active modes) and the explanatory variables. The CC in passive mode (CC_{passive}) is little correlated to the signal of the internal monitors, while it shows the highest correlation with the transmitted signal (Str₁–Str₃) located at the opposite side of the AmBe source. Finally, we can also observe the strong correlation (factor > 0.7) between CC in the active mode (CC_{active}) and internal monitors.

B. Regression Techniques

Regression techniques for the prediction of the CC can be applied using the explanatory variables showing the highest correlations with CC. In this work, we illustrate the benefit of new regression approaches with respect to multilinear regression with ordinary least squares (OLSs), which is traditionally used in our laboratory. We compare its results with the multilayer perceptron (MLP) [11] and random forest (RF) [12] algorithms from the Scikit-Learn Python library [13].

Linear regression with the OLS provides a straightforward and interpretable framework for estimating relationships between dependent and multiple independent variables. OLS provides an estimation of the linear model coefficients by

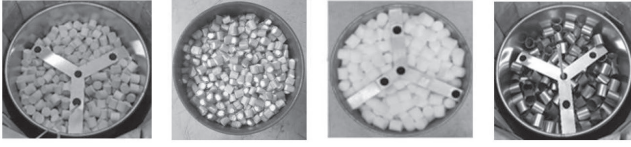


Fig. 4. Mockup drums modeled in MCNP. From left to right: wood–PVC–polyethylene–stainless steel.

minimizing the sum of squared differences between observed and predicted values of the CC (here, CC_{MCNP} and CC_{pred}). CC_{pred} is defined as an analytical function of quadratic and polynomial value (up to the second order) of the most relevant explanatory variables (see Section IV-A).

An MLP is a class of artificial neural networks characterized by multiple layers of interconnected nodes, or “neurons,” which process input data to produce desired outputs [13]. The efficacy of MLPs in regression modeling stems from their capacity to approximate highly nonlinear mappings between input and output variables. Unlike traditional multilinear regression models, MLPs can capture intricate patterns, interactions, and nonlinearity present in the data.

On the practical side, the optimization process involves defining hyperparameters, such as the number of hidden layers, the number of neurons for each hidden layer, the activation function, and the learning rate [13]. These are determined through a cross-validation process, which is exclusively applied to the experimental design L72 that presents a larger number of density and matrix levels in comparison to L32 (see Table I).

The RF algorithm is a powerful asset in predictive modeling, particularly for complex and high-dimensional datasets. By building a large set of decision trees and aggregating their predictions, RF enhances prediction accuracy, reduces overfitting, and provides valuable insights into variable importance. The hyperparameters include the number of trees, the criterion, the minimum number of samples required to split a node, and among others [13]. These are also determined through a cross-validation process, which is exclusively applied to the experimental design L72.

C. Tests of the Regression Models With Homogeneous Mockup Drums

In this work, the predictions of CC calculated by the regression models will be checked using calculations with the MCNP models of realistic homogenous waste drums. For this purpose, we use the MCNP models of four mockup drums filled with different matrices (see Fig. 4), which will be used in the experimental tests of MICADO cell at the Nuclear Measurement Laboratory of CEA Cadarache.

These four matrices are representatives, in terms of neutron moderation and absorption, of the materials constituting radioactive technological wastes. In addition, their characteristics are included in the range of the parameters of the experimental design regarding matrix composition, density, filling level, and nuclear mass content (see Table II).

In the following, the unique L104 experimental design (as described in Section III) is directly used for the training

TABLE II
118 L MOCKUP DRUMS DESCRIPTION

Designation	Matrix material	Composition	Apparent density ($\text{g}\cdot\text{cm}^{-3}$)	Filling level
Mockup 1	Stainless steel	Fe (70%) – Ni (18%) – Cr (8%)	0.63	82%
Mockup 2	Wood	$\text{C}_6\text{H}_{10}\text{O}_5$	0.35	95%
Mockup 3	Polyethylene	CH_2	0.5	92%
Mockup 4	PVC	$\text{C}_2\text{H}_3\text{Cl}$	0.27	91%

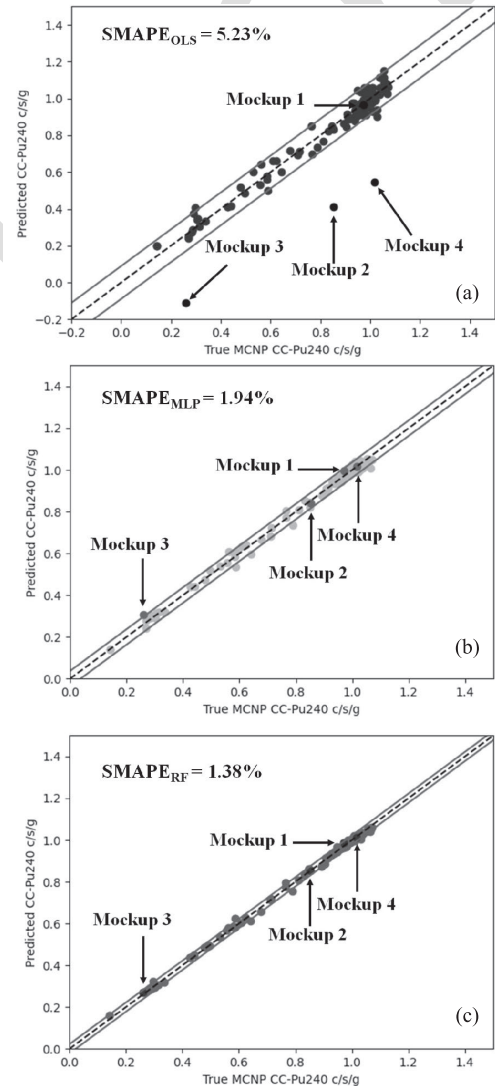


Fig. 5. Predicted CC-240Pu of the mockup cases (labeled points) in passive neutron coincidence counting obtained with (a) OLS, (b) MLP, and (c) RF algorithms. The two red lines are the interval $\pm 2 \sigma_{res}$.

database, and the prediction performance of the regression techniques is assessed using simulation results performed for the four mockup drums. The result of the regression models (called predicted CC or “ CC_{pred} ”) is compared to the MCNP simulated CCs (called “ CC_{true} ”) for the 104 configurations in Figs. 5 and 6 for passive neutron coincidence counting and for active neutron interrogation, respectively. The predicted CC and the true CC of the four mockup drums are also indicated.

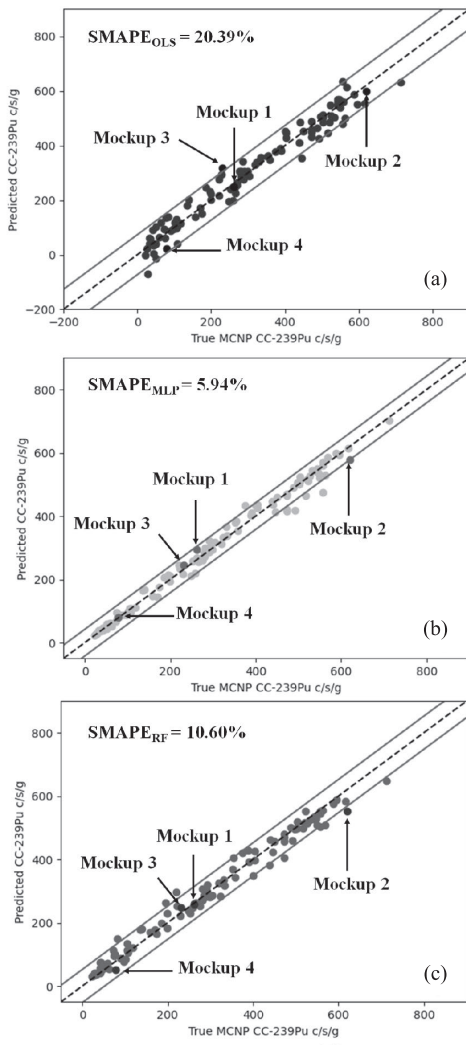


Fig. 6. Predicted CC-239Pu of the mockup cases (labeled points) in active neutron interrogation obtained with (a) OLS, (b) MLP, and (c) RF. The two red lines are the interval $\pm 2 \sigma_{res}$.

The predicted CC (CC_{pred}) by the three regression methods is represented against the simulated (CC_{true}). The residual error of the multilinear regression with the interval is $\pm 2 \sigma_{res}$. The absolute value of σ_{res} is deduced from the square root of the mean square error (mse). Values covered by the CC are largely distributed and are not organized in clusters, demonstrating the relevance of the description of the experimental design. The symmetric mean absolute percentage error (SMAPE) demonstrates a better quality of the regression using MLP and RF methods compared to OLS, in both passive coincidence counting (SMAPE_{OLS} = 5.23%, $\sigma_{res,OLS}$ = 0.0446 c.s⁻¹.g⁻¹; SMAPE_{MLP} = 1.94%, $\sigma_{res,MLP}$ = 0.0185 c.s⁻¹.g⁻¹; SMAPE_{RF} = 1.38%, $\sigma_{res,RF}$ = 0,0111 c.s⁻¹.g⁻¹) and active interrogation (SMAPE_{OLS} = 20.39%, $\sigma_{res,OLS}$ = 37 c.s⁻¹.g⁻¹; SMAPE_{MLP} = 5.94%, $\sigma_{res,MLP}$ = 21 c.s⁻¹.g⁻¹; SMAPE_{RF} = 10.60%, $\sigma_{res,RF}$ = 26 c.s⁻¹.g⁻¹). Additionally, a few negative, nonphysical values of CC_{pred} arise from the linear regression [“Mockup 3” on Fig. 5 and a few others (blue points) on Fig. 6], disqualifying the validity of the OLS method in this region. Table III compares the metrics of

TABLE III

COMPARISON METRICS OF THE DIFFERENT MULTILINEAR REGRESSION MODELS IN PASSIVE NEUTRON COINCIDENCE COUNTING AND ACTIVE NEUTRON INTERROGATION FOR THE FOUR MOCKUP DRUMS

	Mock-up number	CC_{true} (MCN P) [c.s ⁻¹ .g ⁻¹]	$CC_{pred,OLS}$ (c.s ⁻¹ .g ⁻¹ and relative difference in %)	$CC_{pred,MLP}$ (c.s ⁻¹ .g ⁻¹ and relative difference in %)	$CC_{pred,RF}$ (c.s ⁻¹ .g ⁻¹ and relative difference in %)
Passive neutron coincidence counting	Mock-up 1	0.972	0.964 (-1%)	0.995 (2%)	0.986 (1%)
	Mock-up 2	0.851	0.411 (-52%)	0.842 (-1%)	0.854 (0%)
	Mock-up 3	0.261	0.110 (-58%)	0.306 (17%)	0.270 (3%)
	Mock-up 4	1.017	0.545 (-46%)	1.017 (0%)	1.011 (-1%)
Active neutron coincidence counting	Mock-up 1	262	250 (-5%)	294 (12%)	259 (-1%)
	Mock-up 2	621	600 (-3%)	579 (-7%)	553 (-11%)
	Mock-up 3	231	319 (38%)	245 (6%)	248 (7%)
	Mock-up 4	79	22 (-72%)	80 (1%)	51 (-35%)

accuracy for CC_{pred} using the OLS, MLP, and RF methods in passive coincidence counting and active interrogation.

In the passive mode, results indicate that the agreement in the predicted CC in the four mockup drums lies in the [1%–58%], [0%–17%], and [0%–3%] ranges in passive coincidence counting and in the [3%–72%], [1%–12%], and [1%–35%] ranges in active interrogation for the OLS, MLP, and RF methods, respectively. The model based on the OLS method provides poor prediction results in passive coincidence counting, where three of the four drum cases are outside the interval $\pm 2 \sigma_{res}$. This indicates an overfitting of the OLS model and reducing the amount of input parameters, as well as the polynomial order, enables to mitigate this disagreement (while nonetheless degrading the residual error of the fit). On the other hand, the MLP and RF methods forecast all the results inside the interval $\pm 2 \sigma_{res}$. In active neutron interrogation, a better accuracy and consistency of the predictions is also observed for the MLP and RF methods.

D. Model Prediction Extension to Heterogeneous Waste Cases

To assess the validity and limitations of the prediction regression models established with homogenous waste matrices, ORANO La Hague provided two realistic heterogeneous drums case studies, together with their detailed Monte–Carlo simulation model, in the frame of MICADO project [14]. The first realistic case (RC1) consists of a 118 L alpha waste drum divided into five bags containing decontamination wipes. The activity of the plutonium inside the drum is heterogeneous. The matrix is also moderately heterogeneous and exclusively constituted of humid plastic C₃H₆(H₂O) of densities varying from 0.32 to 0.53. The second realistic case (RC2) is a 118 L alpha waste drum containing 50 stainless steel pipes with a low contamination level. The average density is 1.08 g.cm⁻³ and about 87% of the plutonium activity is concentrated in a valve located at the bottom of the drum. Fig. 7 shows a view of the RC1 and RC2 Monte–Carlo models.

The result of the regression models trained with the L104 data set is illustrated in Figs. 8 and 9, in passive neutron coincidence counting and in active neutron interrogation, respectively.

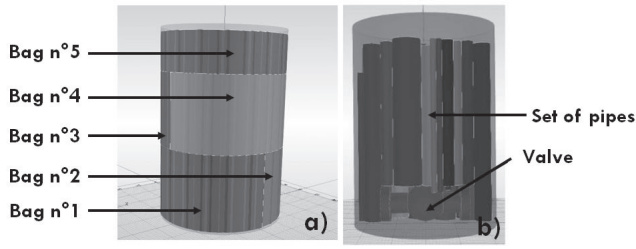


Fig. 7. MCNP models of (a) RC1 and (b) RC2 waste drums. The different colors correspond to different types of material.

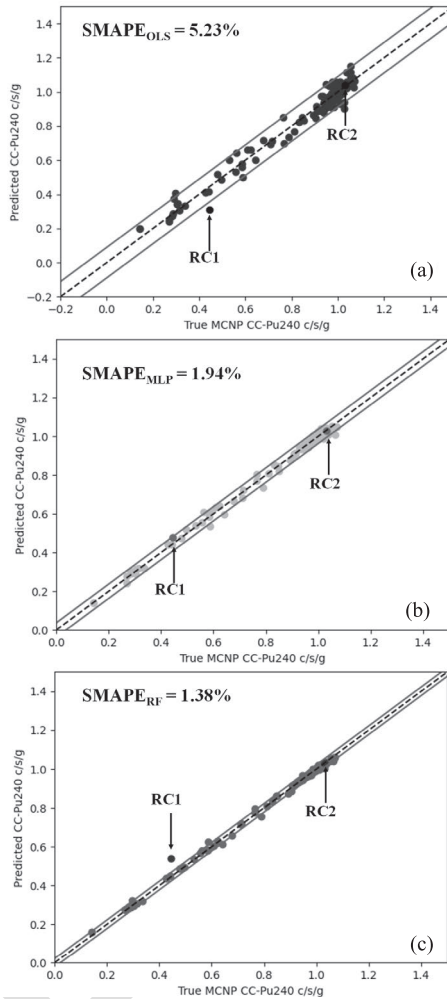


Fig. 8. Predicted CC-240Pu of the realistic drum cases (labeled points) in passive neutron coincidence counting obtained by (a) OLS, (b) MLP, and (c) RF algorithms. The two red lines are the interval $\pm 2 \sigma_{res}$.

348 Table IV compares the accuracy metric results for CC_{pred}
 349 using the OLS, MLP, and RF methods in passive coincidence
 350 counting and active interrogation.

351 As previously mentioned, both drums present certain level
 352 of heterogeneities (matrix and distribution of the plutonium
 353 mass) that was not simulated in the experimental design
 354 calculations. Thus, the regression algorithms have not been
 355 trained with heterogeneous matrices and nuclear material
 356 distributions. This being said, CC_{pred} agrees within [0%–30%]
 357 in passive neutron coincidence counting, regardless of the
 358 regression method, but results are significantly better using

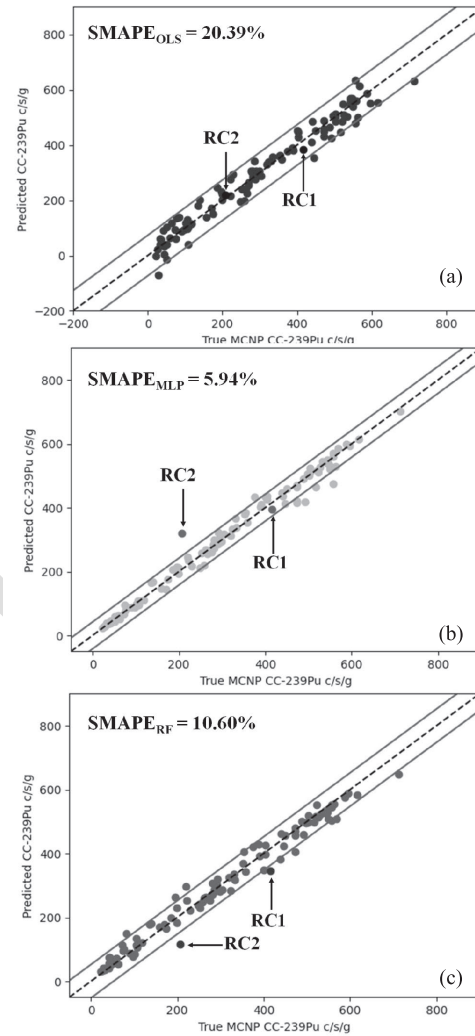


Fig. 9. Predicted CC-239Pu of the realistic drum cases (labeled points) in active neutron interrogation obtained by (a) OLS method, (b) MLP, and (c) RF algorithm. The two red lines are the interval $\pm 2 \sigma_{res}$.

TABLE IV
 COMPARISON METRIC RESULTS OF THE DIFFERENT REGRESSION MODELS
 IN PASSIVE NEUTRON COINCIDENCE COUNTING AND ACTIVE
 NEUTRON INTERROGATION FOR THE
 REALISTIC DRUM CASES

	Mock-up number	CC_{true} (MCN P) [$c \cdot s^{-1} \cdot g^{-1}$]	$CC_{pred,OLS}$ ($c \cdot s^{-1} \cdot g^{-1}$ and relative difference in %)	$CC_{pred,MLP}$ ($c \cdot s^{-1} \cdot g^{-1}$ and relative difference in %)	$CC_{pred,RF}$ ($c \cdot s^{-1} \cdot g^{-1}$ and relative difference in %)
Passive neutron coincidence counting	RC 1	0.445	0.310 (-30%)	0.477 (7%)	0.539 (21%)
	RC 2	1.032	1.039 (1%)	1.027 (0%)	1.033 (0%)
Active neutron coincidence counting	RC 1	416	383 (-8%)	394 (-5%)	345 (17%)
	RC 2	207	220 (6%)	321 (55%)	115 (-44%)

359 MLP and RF than OLS. In active neutron interrogation,
 360 predictions are within [5%–55%] and OLS provides better
 361 predictions of CC for RC2 (6% against 55% and 44% for
 362 the MLP and RF methods, respectively). Beyond the fact that

RC2 shows heterogeneities in matrix composition and nuclear mass distribution, these poor predictions for MLP and RF algorithms are probably due to the average density of RC2, which is out of the boundary limits defined in the experimental design (i.e., 1.08 versus 0.7 g.cm⁻³ maximum in the training set). Overall predicted results of realistic heterogeneous drum cases are thus logically less accurate than the prediction for the previous homogeneous mockup drums.

E. Conclusion and Outlooks

The MICADO neutron measurement system prototype has been designed by MCNP simulation, with the objective to estimate the nuclear material mass in a wide range of radioactive waste drums by passive neutron coincidence counting and active neutron interrogation. The useful neutron signal (namely, the CC in count per second and per gram of ²⁴⁰Pu or ²³⁹Pu, respectively) was calculated by Monte-Carlo simulations for a series of matrix compositions, density, and filling levels defined with an experimental design. The obtained 104 simulation results were used to establish regression models with three different algorithms to reduce the uncertainty on the nuclear mass estimation. These models were based on internal matrix monitors and a neutron transmission measurement. Within the context of this issue, MLP and RF regression techniques show a clear advantage over conventional OLS (linear regression with OLSs), both in passive coincidence counting and active interrogation techniques. This behavior may be attributed to the strong correlation between some of the predictors knowing that one of the conditions to use classical linear regression method is the independence of the predictors. The residual error and the SMAPE on the linear regression of the MLP and RF techniques are lower, and the predicted CC never results in nonphysical negative values. Therefore, the uncertainty on the determination of the nuclear mass can significantly be reduced by comparison to standard OLS method usually employed in our laboratory.

In the next steps, we will investigate more regression methods and we will enlarge the training database with additional Monte-Carlo simulations, including more waste materials and heterogeneous matrices as well. Currently, the use of experimental designs helps to reduce the number of MCNP simulations while providing maximum information for constructing regression models. However, this approach only works if the range of variation for influential parameters (density, filling level, matrix material, etc.) covers the entire range of waste barrels to be measured. When dealing with homogeneous matrices, this approach is relatively robust. However, accounting for matrix heterogeneities and radioactive materials becomes very challenging, even when using

experimental designs driven by MCNP simulations. At present, the idea is to consider extreme cases in terms of heterogeneities. These cases are unlikely to be measured in practice, but it allows to cover a broad range of variations. Despite this, it is still expected that regression algorithms (based on interpolation between different points of the experimental design) will exhibit poorer performance compared to homogeneous cases. In this specific scenario, simulating all the possible heterogeneous configurations to train the models is very complex, if not impossible. Work is ongoing to evaluate the performance for these heterogeneous waste scenarios. Another important prospect is to perform laboratory measurements with mockup drums in DANAIDES casemate of TOTEM nuclear facility, at CEA Cadarache, both to validate our numerical Monte-Carlo calculation models and to test with experimental data the regression models established with simulation data.

ACKNOWLEDGMENT

The work presented in this article reflects only the author's views and the Commission is not liable for any use that may be made of the information contained therein.

REFERENCES

- [1] [Online]. Available: <https://www.micado-project.eu/>
- [2] Q. Ducasse et al., "Design of MICADO advanced passive and active neutron measurement system for radioactive waste drums," *Nucl. Instrum. Methods Phys. Res. A, Accel. Spectrom. Detect. Assoc. Equip.*, vol. 1005, Jul. 2021, Art. no. 165398.
- [3] *MCNP User's Manual Code Version 6.2*, Oct. 2017.
- [4] N. Ensslin, "Principles of neutron coincidence counters," Tech. Rep., LA-UR-90-732, 1991.
- [5] J. T. Caldwell, "The Los Alamos second-generation system for passive and active neutron assays of drum-size containers," Tech. Rep., LA-10774-MS, 1986.
- [6] [Online]. Available: <https://inventaire.andra.fr/>
- [7] *Sodern Tubes and Generators*. Accessed: Sep. 2023. [Online]. Available: <https://sodern.com/en/tubes-neutroniques/>
- [8] R. Sabre, "Taguchi experimental design," in *Engineering Techniques*, Mar. 2007.
- [9] [Online]. Available: <https://support.minitab.com/fr-fr/minitab/18/help-and-how-to/modeling-statistics/doe/supportingtopics/taguchi-designs/catalogue-of-taguchi-designs/#132-21-49>
- [10] L. Lepore et al., "The MICADO integrated gamma station for radioactive waste packages radiological characterization," in *Proc. EPJ Web Conf.*, vol. 288, 2023, p. 07015, doi: 10.1051/epjconf/202328807015.
- [11] G. E. Hinton, "Connectionist learning procedures," *Mach. Learn.*, pp. 555–610, Jan. 1990, doi: 10.1016/B978-0-08-051055-2.50029-8.
- [12] L. Breiman, "Random forests," *Mach. Learn.*, vol. 451, pp. 5–32, Oct. 2001, doi: 10.1023/A:1010933404324.
- [13] F. Pedregosa et al., "Scikit-learn: Machine learning in Python," *J. Mach. Learn. Res.*, vol. 12, pp. 2825–2830, Nov. 2011.
- [14] E. Breuil and A. Bielen, "WP2: Virtual cases & WP8: Combination of measurements and uncertainty assessment," in *Proc. Micado Final Demonstration Conf.*, Jan. 2023.