Validation of Numerical Simulations of Activation by Neutron Flux - 15038

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ABSTRACT

The knowledge of the radionuclide content of radioactive waste is of utmost importance for safety and waste management reasons. Numerical simulations are used at EDF-CIDEN to anticipate the dismantling and the radioactive waste management. The activation scheme by neutron flux developed at EDF-CIDEN comprises four steps. First of all, a 3 dimensional description of the reactor, the chemical composition of each structure, and the neutron sources at nominal power rating conditions are used as the input data to simulate the neutron propagation and to generate a 3 dimensional multi energy group neutron flux distribution in nominal power rating conditions. Secondly, this output data is used with the chemical compositions (including impurities) and the history of the operating conditions to calculate the radiological inventory for each structure. Thirdly, according to the radioactive inventory of each component or sub-component, a waste classification can be made ; the classification criteria are based on the levels of radioactivity and radiotoxicity of 143 radionuclides. Finally, comparisons between the calculated radioactive inventories and the results of radio-chemical analyses can be made. These comparisons are made with regards to the activation of "standard" chemical elements (meaning radionuclides being perfectly modeled with no uncertainty about the chemical concentrations). The results, linked with "standard" chemical elements, can be used to validate the calculation scheme, including the neutron propagation, the neutron activation and all the other associated simplified assumptions (geometry, history of irradiation, etc). For steel, activation of "Fe" is retained, and for stainless steel, activation of "Fe" and "Ni" are retained. Overall, the feedback from Chooz A (Pressurized Water Reactor) shows that numerical simulations produce a slight overestimation of "Fe-55" and "Ni-63". The results linked with minor chemical elements or impurities allowed us to validate the use of the average measured compositions.

INTRODUCTION

The knowledge of the radionuclide content of radioactive waste is of utmost importance for safety and waste management reasons.

State of Dismantling in France

As EDF is the only operator of the French power plants, with regard to the French regulatory, EDF is responsible for dismantling its plants.

About 186,000 t of radioactive waste will be produced from the dismantlement of the 9 EDF first generation power plants, which have been shut down for over 20 years.

There are 9 first generation plants which have been shut down for more than 20 years. Four types of reactors can be noted. So there are 6 Natural Uranium Gas Cooled Reactors, 1 Pressurized Water Reactor (Chooz A), 1 Heavy Water Reactor and1 Fast Breeder Reactor.

Currently, in France ANDRA operates 2 deposit sites, one for "Very Low Activity waste" and another for "Short Life waste". However, 2 new sites will be required, one for graphite, and the other for fission products, actinides and "Long life waste".

The main issue of the radioactive waste management is a correct classification of the waste according to the levels of activity and radio-toxicity.

Radioactive Waste Management in France

Basically, in France, 5 classes of nuclear waste are defined. Each refers to a particular level of activity and radio-toxicity. The different classes from the least to the most penalizing are respectively the "Very Low Activity waste", "Low Activity Short Life waste", "Low Activity Long Life waste", "Intermediate Activity Long life waste", and "High Activity Long Life waste".

The "High Activity waste" class concerns the fission products and the actinides, thus it is not included in the dismantling waste management.

Usually "Long Life waste" is used to talk about Intermediate "Activity - Long Life waste" and "Short Life waste" is used to talk about "Low Activity - Short Life waste". This will be done hereafter.

In France the distinction between the "Long Life" and the "Short Life" waste is based on a list of activity limits for 40 radionuclides. If one of these 40 radionuclide limits is crossed, the waste is classified in the "Long Life waste" category. If none of these limits is crossed, a weighted average activity level is used to separate "Very Low Activity" and "Low Activity Short Life" waste. The weighted average activity value is obtained by taking into account the level of activity balanced with the radio-toxicity of 143 radionuclides. The "Low Activity Long Life waste" concerns principally the graphite.

Precise knowledge of the radioactive inventory is crucial in order to prepare the management of irradiated waste. This can be obtained with numerical simulations, which involve the calculation of 143 radionuclide activities for each structure or sub-structure of interest.

DESCRIPTION

Numerical simulations are used at EDF-CIDEN to anticipate the dismantling and the radioactive waste management. An overall validation of the numerical simulation scheme can be done by using the activation of "standard" chemical elements.

Numerical Simulation Scheme of Activation by Neutron Flux

Figure 1 presents the numerical simulation scheme used by EDF-CIDEN to calculate the activation by neutron flux.

The activation calculations are carried out in 4 steps :

- 3 dimensional multigroup flux calculations in a 3 dimensional geometry : The mapping of the neutron flux is obtained on the basis of a neutron propagation calculation. The tool used (MCNP reference [1] or TRIPOLI reference [2]) solves the Boltzmann equation with no approximation by a Monte Carlo method. So, random series of numbers are used to simulate the life of millions of neutrons. The code follows each neutron individually, from its birth to its disappearance by leakage (when the neutron leaves the geometry modeled), by absorption, or by fission. Basically, each neutron interaction is associated with several random numbers. The input data cover the microscopic cross-sections, the geometry, the isotopic composition with no impurities and the computed neutron sources resulting in the neutrons emitted by the fuel assemblies. The neutron flux map is calculated at the nominal power rating conditions, and each flux is computed in 315 groups of energy.

- Activation calculations of the structures : The radioactive inventories are calculated for the different structures of interest. The tool used (PEPIN-DARWIN reference [3]) solves a Bateman

system of isotopic evolution equations by the Runge Kutta method. So the system of differential equation is solved in successive iterations using approximate solutions for each step. The input data covers the 3-dimensional neutron flux map calculated in step one, the microscopic cross sections, the radioactive decay series associated to the radioactive half lives, the isotopic compositions with impurities, and the history of irradiation resulting in the daily power production. The output data is the radioactive inventory of each component or sub-component of interest limited to a list of 143 radionuclides.

- Waste classification : According to the radioactive inventory of each component or sub-component and the waste classification criteria, a waste classification can be made. Basically the criteria are based on the levels of radioactivity and radiotoxicity of 143 radionuclides : a weighted average activity value is calculated to classify the ""Very Low Activity waste", and 40 radionuclide limits are used to separate the "Short Life waste" from the "Long Life waste".

- Comparisons between calculated results and measured results : For this, we need samples, results of radio-chemical analyses, and results of calculation linked with the analyses. The comparisons are based on calculation measurement ratios (i.e. "C/M"). A value greater than 1 corresponds to an overestimated calculation, and a value less than 1 corresponds to an underestimated calculation. Depending of the results, the input data may be redefined to make a new simulation.
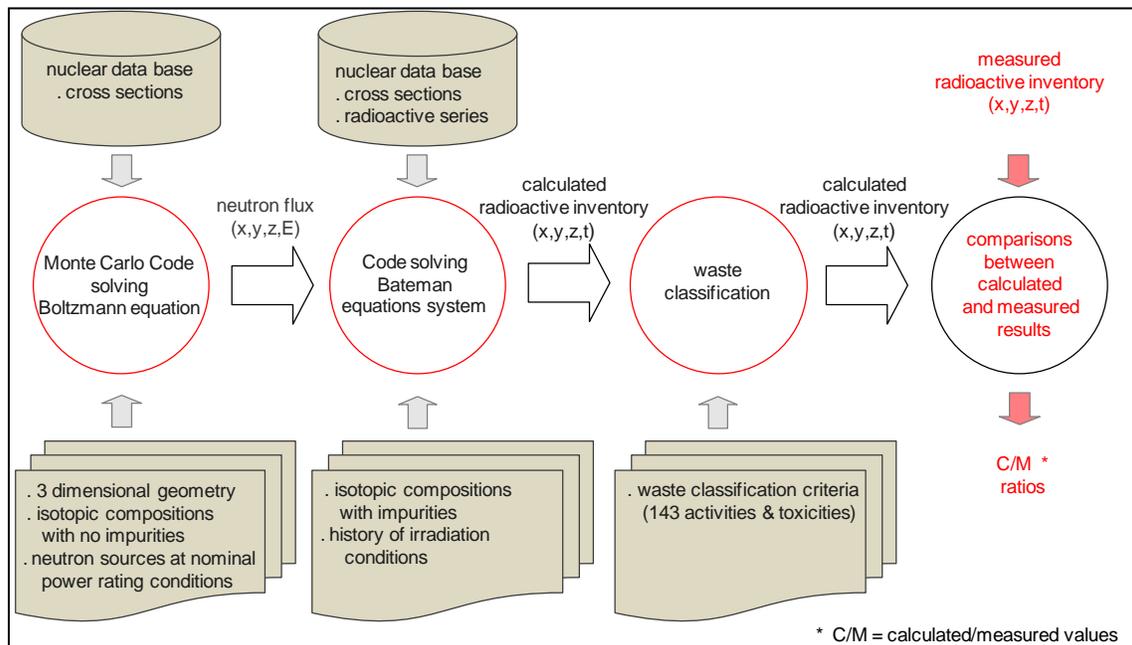
FIGURE 1
Numerical simulation scheme used to calculate the activation by neutron flux

Principal Hypothesis of the Simulations

A 3-dimensional geometry was taken into account in the Monte Carlo code to compute the mapping of the neutron flux : the peripheral fuel assemblies were described pin by pin, whereas the internal fuel assemblies were homogenized. All the internals were fully described 3-dimensionaly, including the different plates, the

control rods, the guide tubes, and the in-core instrumentation. The concrete vessel wall was modeled up to 1 meter deep.

The neutron sources calculated with a core code were given pin per pin with an axial distribution. The control rods are extracted from the core.Three temperatures were defined for the primary water (core inlet, fuel assembly zone, core outlet).

The isotopic compositions are reduced to the major chemical elements to calculate the neutron flux because impurities or traces do not affect the neutron propagation. However, for the activation, it is necessary to use the complete chemical composition including impurities and traces. In fact, these minor impurities could directly impact the waste classification.

The flux mapping is calculated at the nominal power rating conditions while the isotopic evolution uses the history of irradiation. However this history was simplified to a limited number of steps, corresponding to monthly steps.

Figure 2 illustrates the result of the 3D neutron flux mapping in a limited number of tallies (~ 150).
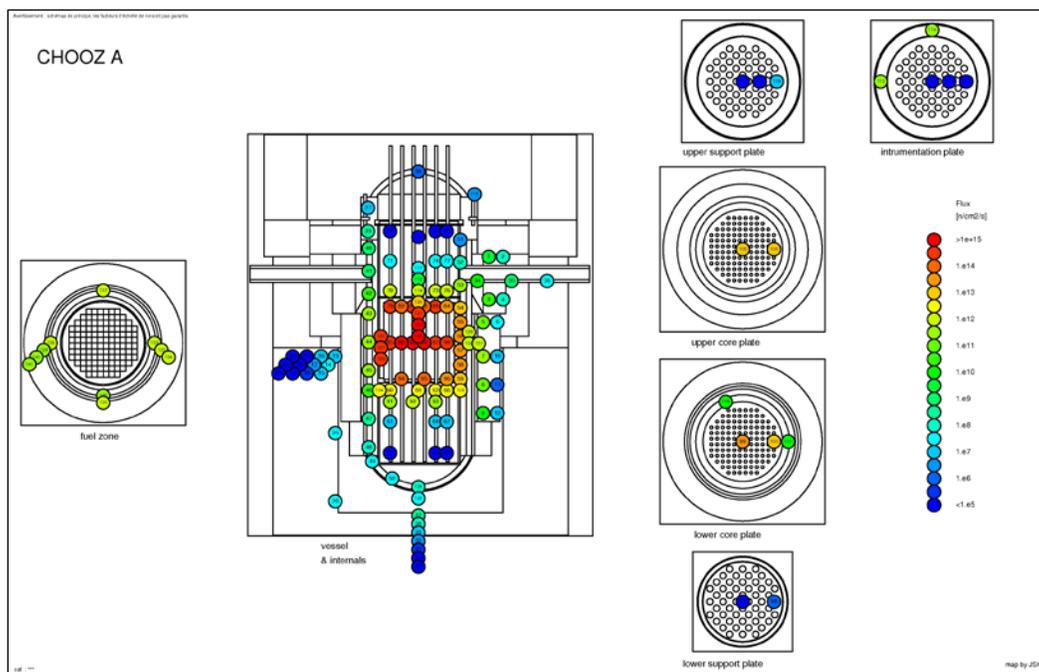
FIGURE 2
3D neutron flux mapping by numerical simulations (Chooz A)

Waste Classification of the Chooz A Structures by Numerical Simulations

The neutron propagation was computed 3 dimensionally in continuous energy scale. The flux was then homogenized in 315 groups of energy in a limited number of tallies, approximately 150 (cf. figure 2). For each of these cells, the radioactive inventory was calculated. Using these radioactive inventories, alongside the waste classification criteria, it was possible to determine the waste classifications.

Figure 3 shows the result of the classification of the Chooz A irradiated waste. As expected, the "Long Life wastes" are located near the fuel assemblies.

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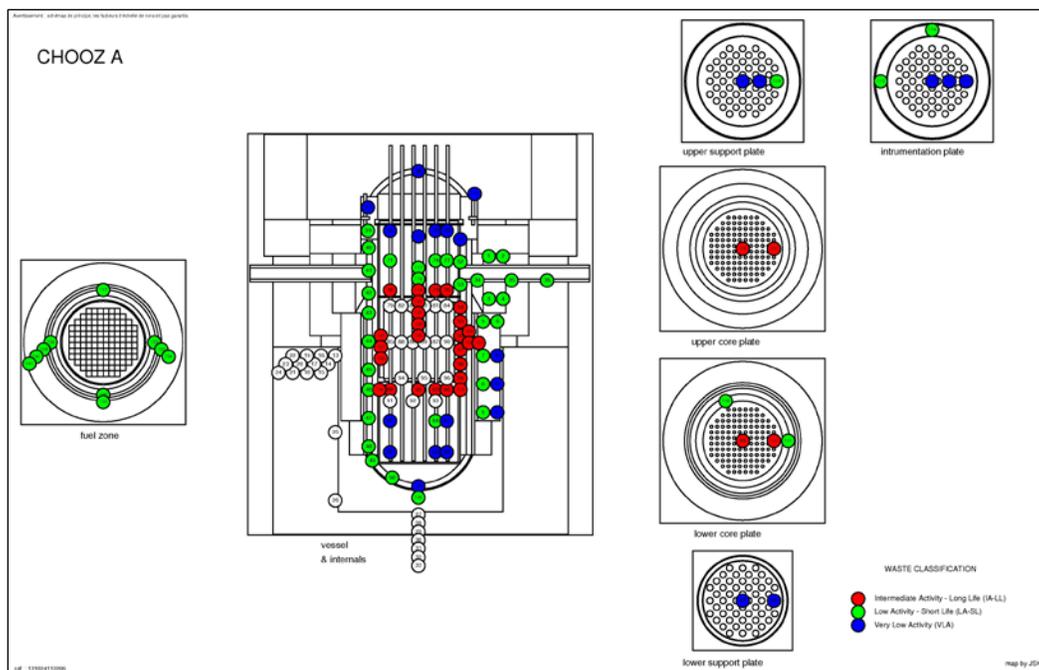
FIGURE 3
Classification of the Chooz A irradiated waste by numerical simulations

Activation of "Standard" Chemical Elements

An overall validation of the numerical simulation scheme can be made by using the activation of "standard" chemical elements. Certain criteria must be satisfied in order to qualify a chemical element as "standard" : high concentration, activation into large cross section range, low concentration uncertainties, production of radionuclide having significant half life (greater than several months) and no difficulties measuring the radionuclide produced.

For steel, activation of "Fe" was retained, and for stainless steel, activation of "Fe" and "Ni" were retained. Table 1 gives the main specifications of these "standard" chemical elements, the activation reactions associated, and the radionuclides produced.

standard chemical element	material	average chemical concentration	activation reaction	produced radionuclide	energy band cross section	half live of produced radionuclide
Fe	steal	~ 98%	$^{54}Fe(n,\gamma)^{55}Fe$	Fe-55	10μeV/20MeV	2.7 years
	stainless steal	~ 68%				
Ni		~ 11%	$^{62}Ni(n,\gamma)^{63}Ni$	Ni-63	10μeV/20MeV	100 years

TABLE 1
Main specifications of "standard" chemical elements and radionuclides associated

DISCUSSION

To make comparisons between calculated and measured values, we used calculation measurement ratios. The reasons for using "C/M" ratios are that each radionuclide has the same weight and each tally also has the same weight. So the comparisons are not affected by the level of the radioactivity or the level of the flux.

The comparison between calculated results and measured results concerning the activation reactions of "standard" chemical elements allows us to quantify the quality of the numerical simulations of the activation by neutron flux. The "C/M" ratios give information with regards to the hypotheses of the 3 dimensional geometry, and the simplified history of irradiation conditions.

Results Concerning Activation of "Standard" Chemical Elements

Figure 4 presents the "C/M" ratios linked with the activation of the "standard" chemical elements of the Pressurized Water Reactor Chooz A.

The analysis of this figure shows that, first of all, regardless of the "standard" chemical element, the numerical simulation produces a slight overestimation of the radioactive inventories. Secondly, these overestimations are linked with the structure considered. Thirdly, the overestimations increase with an increasing fuel distance (i.e. decreasing neutron flux).

The reasons behind these overestimations are directly linked to the measurement uncertainties, the nuclear data uncertainties, the Monte Carlo statistical uncertainties and the hypotheses retained for the irradiation conditions history (like the position of the control rods, the temperature of the primary water, the average monthly power level, etc).

Nevertheless, these overestimations can be qualified as reasonable since the waste classification is not a consequence of these overestimations.
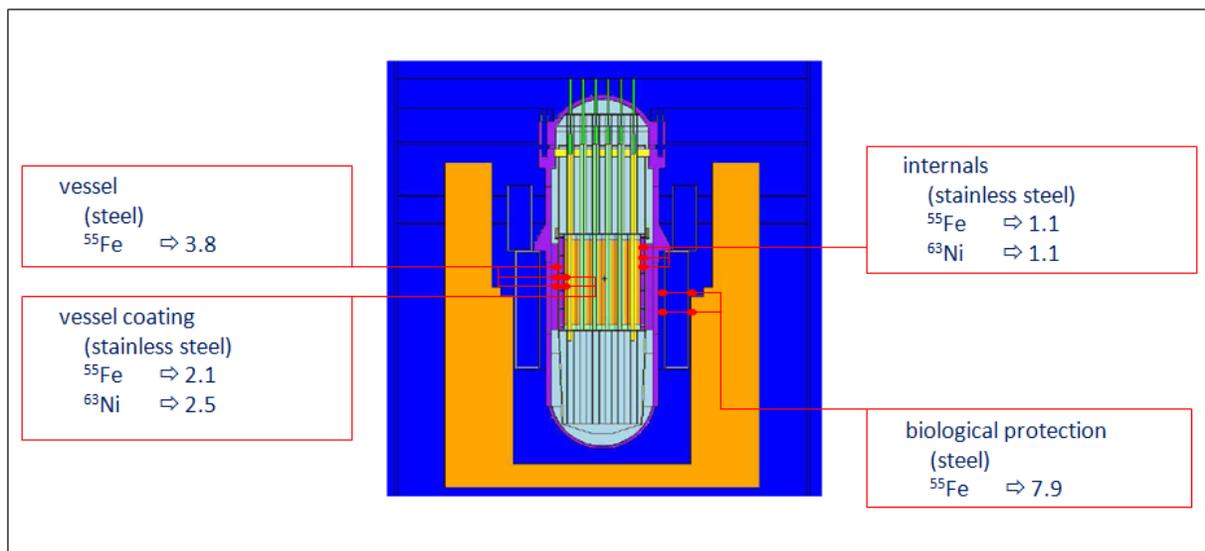
FIGURE 4
"calculation/measurement" ratios linked with the "standard" chemical elements (Chooz A)

Results Concerning Activation of Impurities and Minor Chemical Elements

Average measured concentrations were used to calculate the radioactive inventories due to the activation of impurities and minor chemical elements.

Figure 5 presents the "C/M" ratios linked with the activation of impurities and minor chemical elements of Chooz A.

The analysis of this figure shows that, first of all, regardless of the minor chemical element, the numerical simulation produces an overestimation of the radioactive inventories. Secondly, most of these overestimations are equivalent than those noticed for the "standard" chemical elements. Thirdly it can be noted that for some radionuclides such as "C-14", or tritium, the "C/M" ratios depend on the component considered, and they are significantly different than those observed for the standard chemical elements.

In fact different parents can have the same daughter. Thus "C-14" is the result of the activation of Nitrogen and Carbon. The same can be said for the Tritium which results from different nuclear activation reaction.

Basically the "C/M" variabilities for the minor chemical elements are directly linked to the variability of the concentrations of the minor chemical elements or impurities. But using average concentrations measured on different samples is a good option.

These overestimations are acceptable because they do not change the waste classification. In fact, the sorting between the "Long Life waste" and the "Short Life waste" is principally due to the level of "Ni-63" inventory.
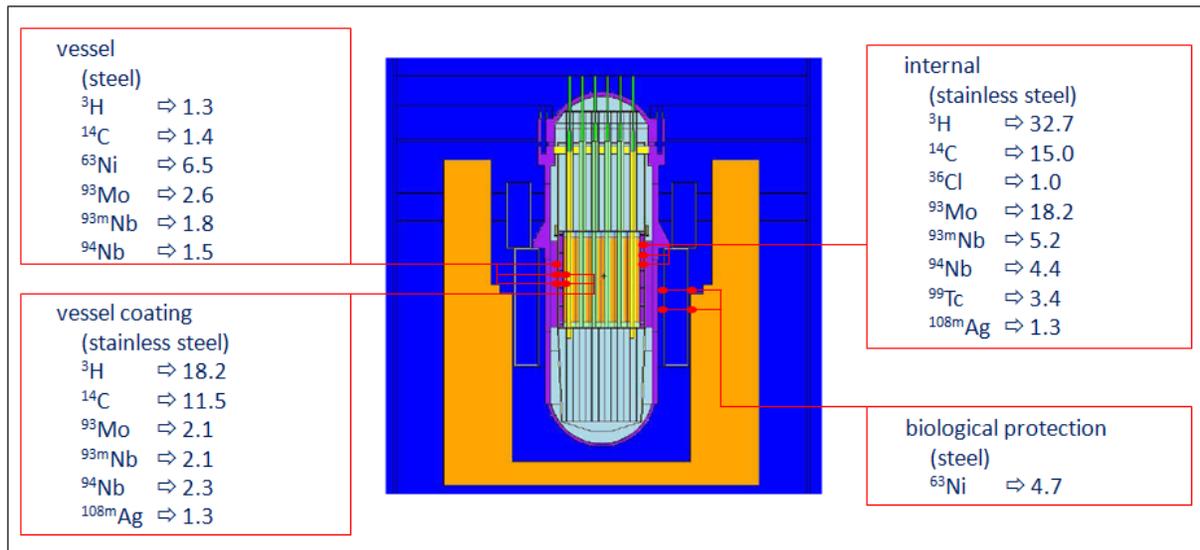
FIGURE 5
"calculation/measurement" ratios linked with the minor chemical elements (Chooz A)

Next Step

The next step is to repeat the same procedure on the other types of reactors and to define "standard" chemical elements of other types of material (concrete, control rods absorber, etc).

The final goal is to obtain the same overestimation levels for "standard" chemical elements and minor chemical elements regardless of reactor size and design.

Initial "C/M" ratios concerning Bugey 1 (Natural Uranium Gas Cooled Reactor) are encouraging because they lead to overestimations equivalent to those observed for Chooz A.

CONCLUSION

The knowledge of the radionuclide content of a radioactive waste is of utmost importance for safety and waste management reasons. Numerical simulations are used at EDF-CIDEN to anticipate the dismantling and the radioactive waste management.

This paper proposes checking the calculated activation by neutron flux of "standard" chemical elements. For this, "C/M" ratios are used to compare calculated values with measured values of "Fe-55" and "Ni-63". The numerical simulations produce overestimations, which are directly linked to the hypotheses retained for the calculations. This check validates the main hypothesis of the numerical simulations and the calculation scheme used to build the radioactive inventory.

Regardless of the minor chemical elements, the numerical simulations produce an overestimation of the radioactive inventories. These overestimations, slightly greater than those noticed for the "standard" chemical elements, are linked to the variability of the concentrations of the minor chemical elements.

The overestimation levels of the numerical simulations can be qualified as reasonable since they do not change the waste classification.

The next step is to repeat the same procedure on the other types of reactors and other types of material.

REFERENCES

[1]   Nuclear Energy Agency - Computer Program Services - CCC-0701 MCNP4C2

[2]   Nuclear Energy Agency - Computer Program Services - NEA-1716 TRIPOLI-4 VERS. 8.1

[3]   Nuclear Energy Agency - Computer Program Services - NEA-1339 PEPIN

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