DETERMINATION AND DECLARATION OF CRITICAL NUCLIDE INVENTORIES IN BELGIAN NPP RADWASTE STREAMS

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ABSTRACT

The Nuclear Power Plants managed by ELECTRABEL are located at the Doel (4 units) and the Tihange (3 units) sites and have a total capacity of 5700 MWe. All units are of the PWR type.

Taking into account the need for retrievability and reliability of all requested waste data, the operator ELECTRABEL has subcontracted a complete study to the engineering company TRACTEBEL ENERGY ENGINEERING (TEE) in order to elaborate a computer code for the determination of critical nuclides in the different waste streams. This program should guarantee retrievability and reliability of all information related to the waste packages produced at the NPP.

Two computer codes, LLWAA and DECL, have therefore been developed by TEE.

The first code (LLWAA : Low Level Waste Activity Assessment code), enables to predict the global inventories and/or the scaling factors of the critical nuclides in the conditioned and in the non-conditioned waste generated by the operation of a PWR.

This code is site-specific as it takes into account the plant design characteristics and operating conditions. A version for BWR plants is under development.

The second code 'DECL' , deals mainly with the complete database management of each waste package produced in order to guarantee full retrievability.

LLWAA and DECL are implemented as an integrated software package called 'DECLARE' at the sites of Doel and Tihange.

Furthermore the LLWAA-code has been extended for the determination of the critical nuclides activities in ashes produced by incineration (LLWAA-Ashes) and for the assessment of the critical nuclides activities deposited on equipment of the nuclear auxiliary systems (LLWAA-Decom).

LLWAA

LLWAA (**L**ow **L**evel **W**aste **A**ctivity **A**ssessment) is a full featured user friendly computer program, enabling the waste producer to determine the so-called critical nuclides inventories in Nuclear Power Plant radwaste streams and to declare the isotopic content of each waste package.

<u>Program Functions</u>

The **LLWAA code** calculates the radionuclides inventories in the process and technological NPP waste on the basis of characteristics and operating conditions of the Nuclear Power Plant.

LLWAA output deals with :

- the activities of the so-called critical nuclides in the Reactor Coolant and the Auxiliary Systems,
- the activity inventories of those nuclides in the on-site conditioned process waste streams,
- the activity inventories of those nuclides in the non-conditioned technological waste streams,
- the scaling factors in the non-conditioned and on-site conditioned waste streams

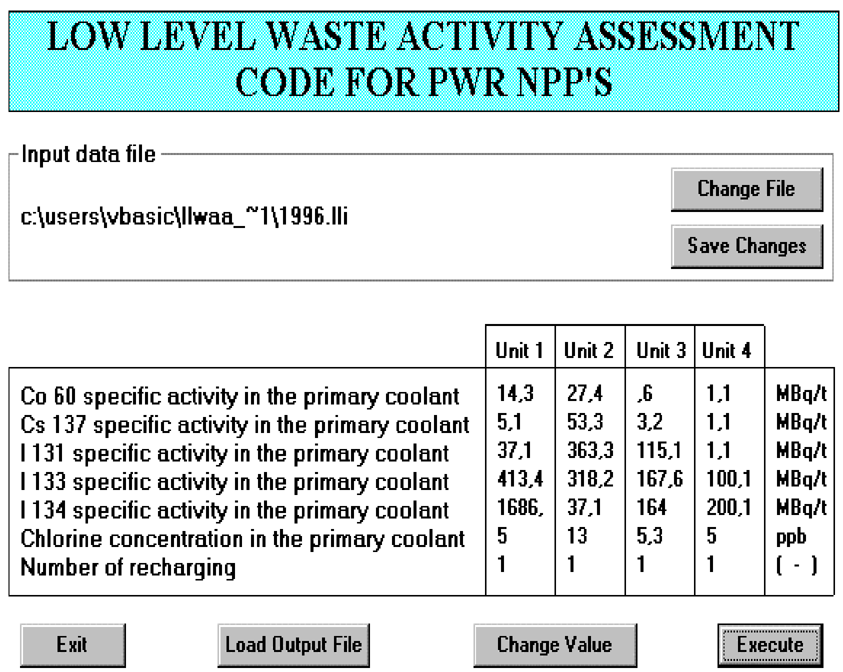

Figure 1 : Input window for the plant operating conditions.

This code is site-specific as it takes into account the design characteristics and operating conditions of the different units of the site with a centralized waste treatment facility. The calculation of the activity inventories for each waste package requires the proper modeling of the Reactor Coolant System (RCS), the Nuclear Auxiliary Systems (NAS) and the Liquid Waste Processing System (LWPS).

(*) *scale factor : ratio between the activity inventory of a specific nuclide and the activity inventory of a critical nuclide.*

1.1 Code parameters and code output

Basically, the **code parameters** are classified into three categories :

- Category 1 : containing all parameters related to the operating conditions which can change or be modified from one cycle to another. The values of these parameters are the main input data of the code and are based on the routine reactor coolant measurement results (^{60}Co, ^{134}Cs, ^{137}Cs, ^{131}I, ^{133}I, ^{134}I activities and Cl^- concentration). The type of fuel (MOX or U235 enriched fuel), fuel cycle length or rated plant power are additional input values.

- Category 2 : parameters pertaining to the plant basic characteristics and operating procedures; among those we find RCS, NAS and LWPS flowrates, boron content in the RCS and evaporators,..

- Category 3 : core activity inventories (calculated by means of the ORIGEN2 Code), chemistry of the primary coolant, equipment materials and surface areas, equipment corrosion rates, DF-factors of the ion-exchange resins and filters, physical-chemical forms of the activity in the RCS (fraction of activity present in soluble and in insoluble forms) during normal operation and refueling outage, diffusion coefficients through fuel cladding defects.

The **code output** consists of :

- The specific activities (or activity inventories) of the critical nuclides in the RCS, ion-exchange resins, filters, evaporator concentrates and miscellaneous non-conditioned waste (combustible waste, non combustible waste, compactable waste, ventilation filters, …) ;

- The scaling factors of the critical nuclides in the same waste streams and waste packages; Table I gives an example of the code output.

IER - MIX

	MIX
	MBq/m3
C 14	1,13e+03
Cl 36	4,67e+00
Mn 54	2,19e+05
Fe 55	7,53e+06
Co 58	1,25e+06
Ni 59	1,41e+07
Co 60	4,40e+06
Ni 63	7,20e+06
Nb 94	6,99e+03
Sr 90	1,20e+04
Tc 99	3,30e+02
I 129	4,91e+00
Cs134	5,13e+05
Cs135	3,54e+00
Cs137	8,07e+05
U 234	7,81e-02
U 235	2,61e-03
Np237	7,60e-03
U 238	1,46e-02
Pu238	3,72e+01
Pu239	1,29e+01
Pu240	1,18e+01
Pu241	2,49e+03
Am241	1,40e+01
Pu242	2,28e-02
Am243	1,80e+00
Cm244	2,24e+01

Table I : Typical activity inventory given by the code for a mix of spent ion-exchange resins.

Code Validation

The predicted values were compared to the values derived from specific measurements performed by the Nuclear Research Centre of Mol (SCK/CEN). There is a good agreement between predicted and measured values for most of the critical nuclides.

The code is qualified and accepted by ONDRAF/NIRAS, the Belgian National Agency for Radioactive Waste Management.

OTHER FEATURES

DECLARE

The LLWAA-code (Figure 2) can be coupled with DECLARE software (cfr. Figure 3); this latter :

- dealing with the complete database management of the data related to each produced waste package (physical form, weight, volume, specific weight, contact dose rate, specific activities of the key nuclides $^{60}Co/^{137}Cs$),
- issuing the declaration form requested for the waste evacuation (evacuation campaign number, identification number of the waste package, conditioner, installation used, type of waste treated, all characteristics on the waste package, dose rate, list of critical nuclides, results of γ-spectrometric analyses, residual surface contamination, …).

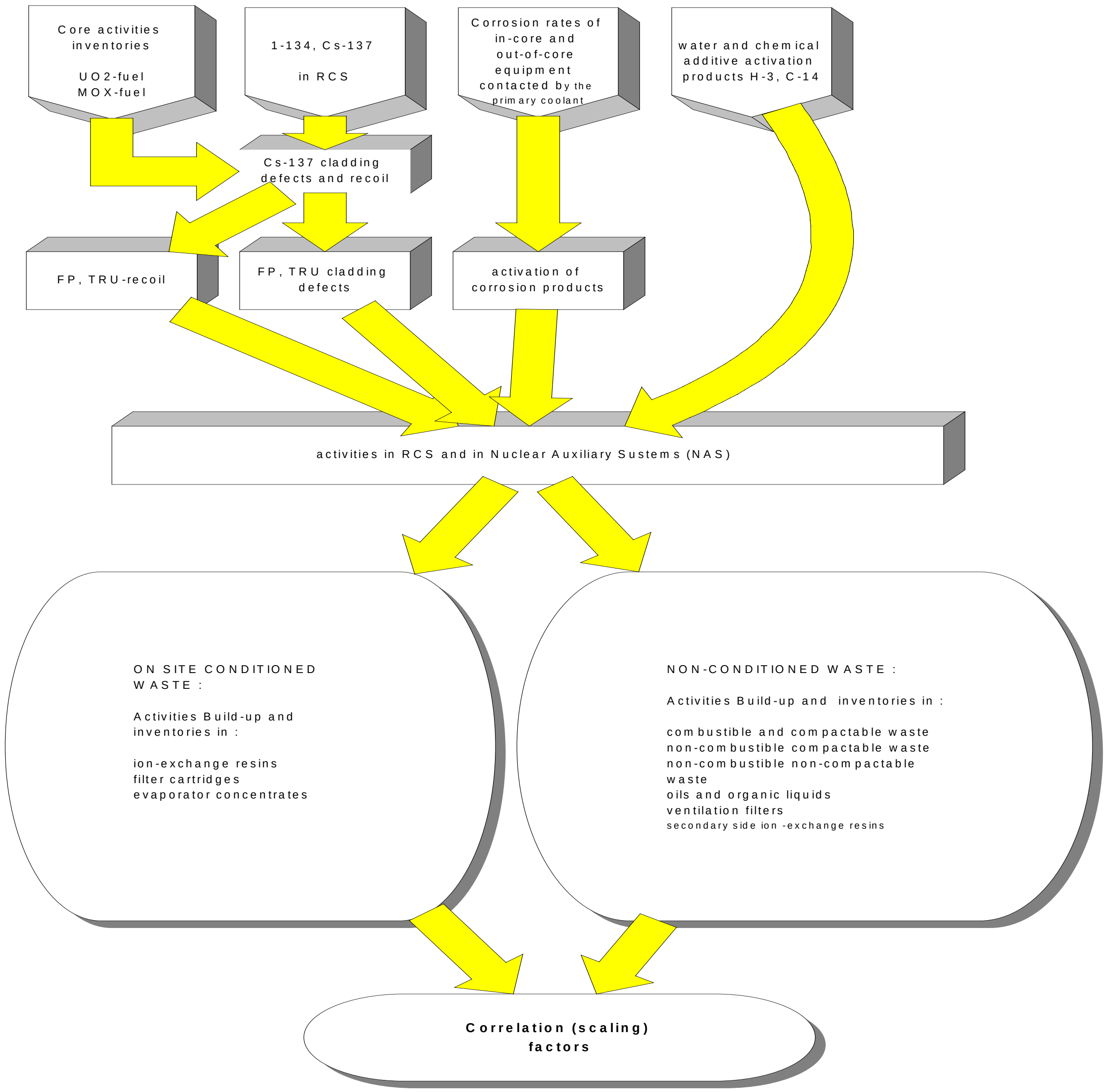

Figure 2 : Structure of the LLWAA- code

Methodology

Apart from the physical data of the waste packages which are 'constant', the activity inventories to be mentioned on the official forms for demand of transfer to the Belgoprocess site are based upon :

- for the on site conditioned waste : gammaspectrometric measurements of ^{60}Co and/or ^{137}Cs in the waste packages as these are considered as the key-nuclides. These measurements, together with the scaling factors computed by the LLWAA-code (automatically), are entered in the DECL-code which calculates the absolute inventories of the critical nuclides. Should the ^{60}Co and/or ^{137}Cs activity not be detectable in the conditioned waste, the DECL-code uses the LLWAA computed activities of these nuclides. Dose rate measurements are also indicated.

- for the non-conditioned waste forms : the nuclide inventories are calculated by the LLWAA-code on basis of dose rate measurements.

OTHER DEVELOPMENTS

These other developments are all based on the LLWAA-code calculating the required specific activities as an input for following codes : LLWAA-Ashes and LLWAA-Decom.

The interrelationship of these codes is given in Figure 3.

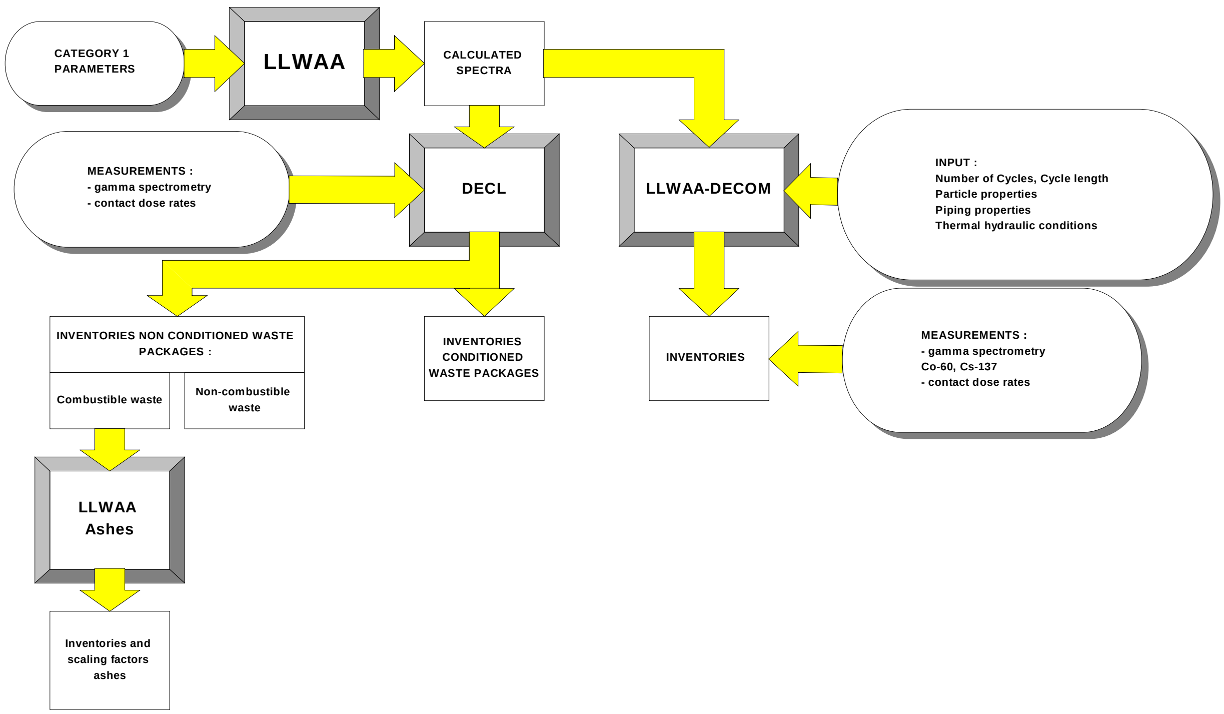

Figure 3 : Interrelationship between the different codes.

<u>LLWAA-ASHES-code</u>

Objectives of the code

The LLWAA-ASHES code calculates the specific activities and the scaling factors of the critical nuclides in the ashes produced by the incineration of the combustible waste generated by nuclear installations. This code has been validated against measurements for the major critical nuclides (^{14}C, ^{54}Mn, ^{58}Co, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{94}Nb, ^{3}H, ^{90}Sr, ^{134}Cs, ^{137}Cs, U- and Pu-isotopes). The structure of the code is given in figure 3.

Code parameters

This code takes into account

- the characteristics of the combustible waste : basic materials, specific weight and specific activities of the critical nuclides (calculated by LLWAA-code)

- and the operating conditions of the incinerator : temperature, waste mass reduction factors and critical nuclides volatility.

Code output

The **code output** consists of :

- the specific activities (or activity inventories) of the critical nuclides in the ashes and fly-ashes;
- the scaling factors of the critical nuclides in the ashes and fly-ashes.

Methodology

The code takes into account all related incineration process parameters; for each type of waste to be incinerated a mass balance has to be established. The solid or liquid waste entering the incinerator will leave the equipment under different chemical and/or physical forms i.e. a solid part (ashes and fly-ashes) and a gaseous part (flue gas).

The activity of each isotope, present in the final product (ash) is related to :

- the corresponding activity of the isotope in the combustible waste, entering the incinerator (this activity can be calculated by the LLWAA-code or is simply a measured value)
- the mass reduction factor which is dependent on the type of waste to be incinerated and the operating conditions of the incinerator (most important parameter is the combustion temperature)
- the activity partitioning factor between the ash and volatile materials ; for a given type of waste this factor depends on the considered isotope.

Concerning the volatility, the codes takes into account the different specific behavior mechanisms of certain isotopes during incineration; four main volatility groups are considered (very volatile: Cs, Te, I, C; average : Co, Mn, Nb, Ni, Tc; low : Sr, Ba and very low volatility : U, TRU).

Scaling (correlation) factors should address only isotopes having the same generation mechanism and behaviour in RCS. Therefore the reference isotope in the ashes for fission products is ^{137}Cs, for activation products ^{60}Co is the reference.

The use of these calculated scaling factors and gammaspectrometric measurements of ^{60}Co and/or ^{137}Cs on the waste packages, containing the ashes, will lead to calculates the absolute inventories of the critical nuclides. Should the ^{60}Co and/or ^{137}Cs activity not be detectable in the incinerated waste, the code uses the LLWAA-values.

Code validation

The code has been validated against measurements, performed by Belgoprocess, for the major critical nuclides (C^{14}, Mn^{54} , Co^{58} , Ni^{59} , Ni^{63} , Nb^{94} , Sr^{90} , $Cs^{134,}$, Cs^{137} , U and Pu-isotopes). Generally there is a good agreement between the calculated and measured values. Table II compares the scaling factors of the critical nuclides in the non-conditioned combustible waste and in the ashes.

Table II : Comparison of the scaling factors in the non-conditioned waste and in the ashes.

	Combustible waste	Ashes		Combustible waste	Ashes
C^{14}/Co^{60}	7.06E-03	1.31E-04	Cs^{135}/Cs^{137}	3.46E-06	3.46E-06
Cl^{36}/Co^{60}	5.14E-05	1.44E-07	U^{234}/Cs^{137}	4.62E-06	2.96E-05
Mn^{54}/Co^{60}	8.05E-01	4.39E-02	U^{235}/Cs^{137}	1.36E-07	8.71E-07
Fe^{55}/Co^{60}	1.56E+00	1.08E+00	Np^{237}/Cs^{137}	5.02E-09	3.21E-06
Co^{58}/Co^{60}	5.93E-01	5.93E-01	U^{238}/Cs^{137}	1.07E-06	6.84E-06
Ni^{59}/Co^{60}	8.01E-03	8.01E-03	Pu^{238}/Cs^{137}	2.85E-05	1.82E-02
Ni^{63}/Co^{60}	8.74E-01	8.74E-01	$Pu^{239}+^{240}/Cs^{137}$	1.78E-05	1.14E-02
Nb^{94}/Co^{60}	1.04E-03	2.52E-04	Pu^{241}/Cs^{137}	1.92E-03	1.23E+00
Sr^{90}/Cs^{137}	6.76E-03	4.26E+00	Am^{241}/Cs^{137}	1.23E-05	7.87E-03
Tc^{99}/Cs^{137}	3.89E-04	1.52E-01	Pu^{242}/Cs^{137}	2.03E-08	1.30E-05
I^{129}/Cs^{137}	3.34E-06	3.34E-06	Am^{243}/Cs^{137}	1.84E-06	1.18E-03
Cs^{134}/Cs^{137}	9.05E-01	9.05E-01	Cm^{244}/Cs^{137}	1.97E-05	1.26E-02

LLWAA-DECOM-code

The costs of the future decommissioning of the NPP's appears to be highly dependent on the equipment contamination (pipework, valves, heat exchangers,…). Therefore a new software has been developed to assess the critical nuclides activities deposited on the equipment of the nuclear auxiliary circuits. This software takes into account the contamination in the streams of the systems (calculated by LLWAA), the operating conditions (fluid velocity, pH, temperature), the corrosion products characteristics (particulate diameter distribution) and the nuclides deposition/release rates on the equipment. The main structure of this code is given below.

The main goal of the program is to estimate, by using correlation factors, the deposited activity on the piping, to be dismantled, in contact with radioactive liquid or gaseous fluids. Scaling (correlation factors) should address only isotopes having the same generation mechanism and behaviour in the reactor coolant system. For fission products the reference isotope is ^{137}Cs, for activation products ^{60}Co is the reference.

Code input

A number of parameters are considered :

- characteristics of the equipment to be dismantled (piping diameter, pipe rugosity,..)
- operating conditions (temperature, average fluid velocity, pH, number of cycles, cycle life,…)
- the corrosion products characteristics (particle density, particle diameter distribution;…)
- physical and chemical characteristics of the isotopes (decay rate, decay energy, photon conversion factors, …)
- specific activities of the considered isotopes (calculated by the LLWAA-code for the considered nuclear auxiliary circuit)

Table III shows an example of the input table of the code.

NPP'S informations
Number of operation cycles 5
Cycle life (months) 16

Particle properties
Particle density (kg/m3) 2000

Piping properties
Pipe diameter (m) 0.1023
Pipe rugosity (mm) 0.0015
Detector - pipe (mm) 3

Thermal hydraulical conditions
Absolute temperature (°K) 319.1
Average fluid velocity (m/s) 0.766
pH(T) 7

Run
Print
Exit
Modif

Isotope data

	Cv (Tot.) (Bq/m3)	frspr	Lambda i (1/s)	hE (mSv/h)/(Bq/cm2)
Fe-59	8.00E6	0.8	1.80E-7	2.24E-5
Mn-54	1.01E7	0.8	2.57E-8	1.74E-5
Fe-55	1.05E7	0.8	8.16E-9	0
Co-58	1.24E8	0.8	1.13E-7	1.94E-5
Ni-59	4.68E4	0.8	2.93E-13	0
Co-60	6.40E6	0.8	4.16E-9	2.32E-5
Ni-63	5.16E6	0.8	2.20E-10	0
Nb-94	6.02E3	0.8	1.08E-12	2.72E-5
Sr-90	7.81E3	0.9995	7.68E-10	0
Tc-99	3.05E2	0.9995	1.03E-13	0
I-129	2.68E0	0.9995	1.40E-15	1.18E-5
Cs-134	1.14E6	0.9995	1.07E-8	2.70E-5
Cs-135	3.71E0	0.9995	9.56E-15	0
Cs-137	8.00E5	0.9995	7.30E-10	1.36E-5
U-234	1.27E-1	0.7	9.02E-14	0
U-235	4.34E-3	0.7	3.13E-17	3.79E-6
Np-237	1.03E-2	0.7	1.03E-14	5.82E-6
U-238	2.75E-2	0.7	4.92E-18	0
Pu-238	1.66E-2	0.7	2.51E-10	0
Pu-239	4.28E1	0.7	9.11E-13	0
Pu-240	5.17E1	0.7	3.37E-12	0
Pu-241	1.15E4	0.7	1.53E-9	0
Am-241	1.04E2	0.7	5.08E-11	5.82E-6
Pu-242	9.95E-2	0.7	5.85E-14	0
Am-243	8.01E0	0.7	2.98E-12	2.65E-6
Cm-244	1.17E1	0.7	1.22E-9	0

Code output

The code output consists of :

- the Reynolds number,
- the friction factor
- the particle deposition and release rate
- the particle velocity due to Brownian movement
- the particle velocity due to the fluid movement
- correlation or scaling factors

Table IV shows an example of output data of the DECOM-code.

Reynolds num.
1.22E+05

friction fact.
4.34E-03

Gamma
9.01E-05

Vb (m/s)
3.64E-03

Vf (m/s)
2.17E-05

Results Box1
New Input
Print
Exit

Correlation Factors

	Cs
Fe-59/Co-60	9.48E-02
Mn-54/Co-60	6.47E-01
Fe-55/Co-60	1.31E+00
Co-58/Co-60	2.28E+00
Ni-59/Co-60	9.62E-03
Ni-63/Co-60	1.04E+00
Nb-94/Co-60	1.24E-03
Sr-90/Cs-137	9.73E-03
Tc-99/Cs-137	4.02E-04
I-129/Cs-137	3.53E-06
Cs-134/Cs-137	8.03E-01
Cs-135/Cs-137	4.88E-06
U-234/Cs-137	1.00E-04
U-235/Cs-137	3.42E-06
Np-237/Cs-137	8.13E-06
U-238/Cs-137	2.17E-05
Pu-238/Cs-137	1.28E-05
Pu-239/Cs-137	3.38E-02
Pu-240/Cs-137	4.08E-02
Pu-241/Cs-137	8.15E+00
Am-241/Cs-137	8.18E-02
Pu-242/Cs-137	7.86E-05
Am-243/Cs-137	6.33E-03
Cm-244/Cs-137	8.48E-03

Methodology

The program calculates, using different modelisation equations, the Brownian diffusion velocity, the entrainment velocity, the deposition and the release coefficient. The latter coefficients are also based on probabilistic settling of particles and transport phenomena.

The evolution of the deposition rate depends mainly on :

- the cycle life time and the number of cycles,
- the initial activity of the deposited materials at the start of the cycle,
- the specific activity of the considered isotope in the equipment (depends on the considered circuit); the value taken into account is the average value based on the cycle length.

In order to validate this code, a dose rate model has been developed and is coupled with the calculation of the deposition based on the principle of consistency between calculated and measured dose rates values because direct measurements of deposited activities presents a number of (practical) difficulties.

Factors affecting the deposition rate are mainly the particle diameter distribution, temperature of the fluid and the flow pattern (turbulent, laminar,..).

The predicted values of the scaling factors are compared to the derived values from specific dose rate measurements performed during NPP shutdown .

Code validation

There is a general good agreement between predicted and measured contact dose rates. The LLWAA-DECOM code allows the calculation of the scaling factors within the range excellent to good; this depends on the considered isotope and the type of equipment. There is also a good consistency between the measured and predicted values of the deposited activities on the portions of the circuits when they were measured, namely, in the frame of the steam generator replacement programs. This concerns mainly the nuclear auxiliary circuits directly connected to the primary system.

Some precautions have to be taken in order to obtain valid measurements :

- the measuring point should be representative for the equipment to be considered;
- 'dead' zones or drain tie-ins should be avoided for measurements (risk of 'hot spots')
- back-scattering effects of other equipment in the neighbourhood of the equipment to be measured should also be avoided
- exact definition of the distance between equipment and detector is required.

MAIN FIELD OF INTEREST

Above mentioned codes are written using Microsoft Visual Basic language and full featured computer programs for use in radiological characterisation of materials, packages and shipments. These programs operate under Windows^{TM} environment. They were developed to provide a practice oriented and easy to operate system for retrieval of all information related to the waste packages.

The software packages LLWAA and DECL have been integrated in the existing computer networks of both NPP sites in Belgium.

The LLWAA-DECOM code is being used for the decommissioning studies of the Belgian NPP's and the LLWAA-ASHES code is used for the characterisation of incinerator ashes.