CALCULATION OF THE ACTIVITY INVENTORY FOR THE TRIGA REACTOR AT THE MEDICAL UNIVERSITY OF HANNOVER (MHH) IN PREPARATION FOR DISMANTLING THE FACILITY

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ABSTRACT

It is planned to dismantle the TRIGA reactor facility at the Medical University of Hannover (MHH).

Radioactive waste resulting from this dismantling will be disposed of externally, any remaining materials as well as the building structures will then be measured to ensure there is no residual activity. In preparation for this and to plan the techniques which will be used to dismantle the reactor, calculations were made in order to determine the amount of activity and the dose rates for the reactor tank and its inside components as well as for the biological shield and its radial beam tube.

The activation of all irradiated components was determined on the basis of both the density of the neutron flux at the location of its installation when the power level of the facility was at operating level and the composition of its materials. The neutron flux was calculated by means of a two-dimensional neutron transport model (S_N program DORT). In addition, it was determined using Monte Carlo calculations with MCNP code at non-symmetrical locations. The application ORIGEN-2 was used for calculating the activity. The calculations were made for all spectral areas, taking into consideration both the history of the power levels and the average density of the flux.

Neutron and photon transport calculations are based on the neutron cross sections from the JEF-2.2 and the ENDF/B-VI libraries. The ORIGEN-2 code was used for calculating the activity of components and the corresponding photon sources. The ORIGEN library was prepared for every spectral zone using average flux spectra from DORT or MCNP.

Measurements of activation and determination of impurities were made from several probes taken from the biological shield and the components. Calculations for both activity and dose rates of irradiated components were compared with the measurements.

INTRODUCTION

Since 1973 a research reactor of type TRIGA I had been in operation at the MHH Clinic for Nuclear Medicine. Until 1996 the reactor was used at a maximum power level of 250 kW mainly to produce radio-pharmaceuticals and for activation analyses. In 1997 the reactor facility was shut down and in 1999 all of the spent TRIGA fuel elements were returned to the United States /1,2/.

In order to determine the present radiological condition of the facility, samples were taken from the reactor components /3/ and both the activation and dose rates were calculated.

The main activated components are the core support, the graphite elements, the graphite reflector with lower and upper grid plate, the irradiation carousel, the reactor tank, the radial beam tube, the central irradiation tube, the filter equipment and the surrounding baryt concrete of the biological shield. The activity distribution and the photon dose for these components must be calculated or measured before dismantling.

The calculations provide a complete picture of the activity of the area near the core from the center of the core to the biological shield. Thus all the main data required for planning the dismantling of the reactor are available.

Details of the values calculated are as follows:

- average and maximum specific activity in the components of the reactor
- dose rates of components and in the areas affected by dismantling
- dismantling thresholds in the reactor tank and biological shield

The first step in getting these values is to calculate the neutron flux distribution for the operational periods by solving a neutron transport equation. The flux densities can then be used to calculate the activity and photon source distribution.

A second transport calculation will be made for photons emitted from the components to get the dose at the surface and at a distance of 1 meter. For validation the calculated quantities will be compared with the results from the samples taken and the radiological measurements.

Thus the information necessary for preparing to dismantle the TRIGA-reactor is available with regard to the amount and treatment of radioactive waste and other residual materials, so that concepts can be developed for waste disposal, the dismantling procedure and final measurements to ensure there is no residual activity.

METHOD OF CALCULATION

The calculation of the activation and dose rate is a rather complex procedure since the geometry of the reactor and its components as well as the tank and the surrounding concrete have to be taken into account. The principle geometry of the TRIGA reactor is cylindrical and therefore easy to describe. But there are some non-symmetrical parts, i.e. the radial beam tube, the filter equipment, graphite elements, control rods and instrumentation tubes as well. These non-symmetrical parts require three-dimensional treatment. Furthermore, the neutron spectra and the neutron flux densities must be known in order to calculate the activation rate and the components' photon sources correctly.

In order to solve the neutron and photon transport equation

- 2D S_N method (program DORT /4/) and
- 3D Monte Carlo Method (program MCNP /5/) were used;

for calculating the activity after irradiation and determination of photon sources

- ORIGEN-2 /6/ was used.

Calculation of the flux density in symmetric parts was done by the S_N method since the solution is very detailed and easier to obtain, also for deeply shielded zones. The Monte Carlo method was used for the nonsymmetrical parts, especially for the materials surrounding the radial beam tube. The scheme of calculation is shown in the flow chart at Fig.1.

CROSS SECTION LIBRARIES

The cross sections for the transport and activity calculations were taken from different libraries, mainly processed at the Institute for Nuclear Technology and Energy Systems (IKE). The library is based mainly on ENDF/B-VI /7/, and was processed by NJOY /8/ at IKE. By means of the code TRANSX /9/ the cross sections were self-shielded and transformed into a data format suitable for the S_N transport code. Since the 292 energy groups were difficult to handle in a 2D code (storage and computing time) the data were condensed into 69 groups via a spectrum calculated by the 1D S_N code ANISN /10/. Therefore the 2D S_N calculations were performed with these 69 energy groups condensed from a multi-group library (in so-called MATXS format) with 165 groups above 3 eV and 127 groups below 3 eV.

The 1D calculation was performed with 292 groups for a radial traverse through the center of the core up to the outer zone of the biological shield for 25 different material and spectral zones. All cross sections for the 2D model were condensed by one of the zone spectra.
The core was homogenized for this calculation. The axially averaged power density from the 3D MCNP calculation was used as the spatial source distribution. The energy source distribution was the U-235 fission spectrum.

2D MODEL

The 2D model covered the complete reactor from 100 cm below the bottom of the tank up to the top of the reactor tank and radially up to the wall of the irradiation room where the radial beam tube ends. There was no attempt to simulate the radial beam tube by means of a ring zone since this part was calculated separately by the 3D model. The core was homogenized for the rotational 2D model as well as the grid plates and some support structures. The central irradiation tube was also homogenized to avoid extreme small meshes.
The whole region for which the 2D transport calculation was made by the DORT code was subdivided into material and spectral zones. From every spectral zone the spectrum was used for the calculation of average activation of zone by ORIGEN-2. 105 different zones were used. The total number of meshes was 160 in a radial direction and 351 in an axial direction.
The neutron source for the 2D model was taken from the averaged distribution of the 3D calculation by MCNP in spatial space and the fission spectrum of U-235 in energy space. This resulted in the flux densities in the fine meshes.

3D MODEL

The 3D model for non-symmetrical parts can be seen at Fig. 2 (vertical and horizontal cross section, respectively). All main parts of the components were modeled explicitly.
The most important non-symmetrical parts were the radial beam tube, the radiation filter equipment, the irradiation carousel, the graphite elements, control rods and the instrumentation tubes.
The graphite elements, the control rods, the irradiation carousel and the instrumentation tubes were realistically modeled.
The cross sections for the MCNP calculations were based on JEF-2.2 and partly ENDF/B-VI. The library was generated at IKE by the NJOY processing system /8/.

The 3D calculation was performed for the first core of TRIGA fuel elements. To get the power distribution a k_{eff} calculation with source iteration was chosen. The result was a power distribution for every fuel assembly and 292 group zone spectra in predefined zones of interest (e. g. in zones of the surrounding concrete of the radial beam tube, sub-zones of the filter equipment etc.). Due to the statistical character of the MCNP results the zones chosen could not be too small, therefore only about 100 different zones of interest were defined.

ACTIVITY AND PHOTON SOURCE CALCULATIONS

The main task was to calculate the activity, after which the photon sources for dose rate calculations were done for single components and the tank without components or with lowered water level.
The program chosen for activity calculations was ORIGEN-2, which can simulate the buildup and decay of nuclides during irradiation. Since most of the reactions leading to activation were thermal neutron capture, the reactions observed from ORIGEN-2 for activation describe the physics sufficiently accurately. The only exception was made for the Li-6 (n,t) reaction: this reaction was simulated as (n,alpha). Since ORIGEN-2 requires a well-weighted one group cross section library, a separate library for each spectral zone of the 2D or 3D model was generated using the 69 group or 292 group spectra, respectively. The basic library for the activation cross section was mainly taken from FENDL-2.0 /11/, which was prepared at IKE for 292 groups and all available and in ORIGEN-2 observed isotopes.

For every spectral zone of the 2D and 3D model the ORIGEN-2 calculations were performed taking into consideration the power history of the TRIGA reactor during the operational period from 1973 to 1996. The total operational time was 9335 days and the total energy production was 2.188 GWh.

The power history simulated in ORIGEN-2 was reduced to a monthly power period followed by a decay period from the first month of operation to the last one. During operation time the power was generated in continuous mode. At the beginning of operation the power was 250 kW, since November 1990 the power had subsequently been reduced to a maximum of 100 kW. This was taken into account for every ORIGEN-2 calculation. The following calculations were performed for all spectral zones

- Generation of the ORIGEN-2 library (condensation of activation cross sections from 292 groups to one group via corresponding zone spectrum)
- ORIGEN-2 calculations for normalized neutron flux densities according to average total zone flux and power history for material specification in this zone
- Corresponding ORIGEN-2 calculations with modified flux density for interpolations of ORIGEN-2 results according to relations between average zone flux and fine mesh fluxes in this zone (2D model)
- Interpolation of ORIGEN-2 results (activities and photon sources) according to fine mesh fluxes (2D model) and fluxes in sub-zones (3D model)
- Extraction of activities for isotopes of interest and photon sources from output of previous step for all fine meshes (2D model) and for zones or sub-zones for the MCNP 3D model

The results of these calculations were the distribution of activities and photon sources for every fine mesh of the 2D model or zone of the MCNP 3D model.

One of the main reasons for these calculations was the specification of material composition for activation calculation. Since the activity of the irradiated material was dependent on impurities like Co, Fe, Eu or Li, the results of activity calculation were strongly dependent on the initial inventory of such impurities. The specification of activated material was therefore partly taken from measurements performed at the VKTA-Rossendorf and MHH laboratories.

DOSE RATE CALCULATIONS

The dose rates were calculated using the photon sources from the previous calculations and performing photon transport calculations using the 2D and 3D model. The cross section libraries for these calculations were taken from libraries for neutron transport calculations since these libraries also contain the photon production data and the photon transport cross sections as well. Since the fuel elements had been removed from the core, only the photon sources of the remaining structures had to be taken into consideration.
For the task of determinating the dose rates of a single component at the surface and at a distance of 1 meter in the air, only the photon sources of the component were used for the 2D model , and materials not belonging to the component were treated as air. The advantage in this was that the main part of DORT input for neutron transport calculations could be left unchanged. This was also the case for calculating the dose rate at the working platform at the top of the reactor tank when the water level was lowered and for the empty reactor tank where only the photon sources from tank and concrete were taken into consideration. After performing the different photon transport calculations the dose rates were calculated using the fluxes from 42 photon group transport calculations and integrated via the dose conversion factors according to ICRP-74 /12/.

The dose rate calculation for the 3D model based on photon source spectra was calculated by the procedure described. Here a special input source had to be defined for every component, due to the more general options for geometry specification in MCNP. The geometry part could remain unchanged. Zones of complete the 3D geometry model which should be removed for single component calculations were set to a void zone or replaced by water.

VALIDATION

The validation of the described method for activity and photon source calculation was performed by means of comparison of measured and calculated activities and dose rates as well. There were samples taken from the outer area of the upper grid plate (Al). The samples were analysed for both chemical composition and activity. The measured and calculated activities of these samples are listed in Table I.
Since the sample positions were at a symmetric location the fluxes of both the 2D and 3D model could be used for calculation. The measured and calculated values agree well inside the estimated uncertainties for calculation and measurement. Only the initial value of Eu had to be adjusted.

Table I. Impurities, calculated and measured activities for AlMg2F18 samples (shavings) from outer area of the upper grid plate.

Impurity	mg/kg	Isotope	Calculated activity Bq/g	Measured activity Bq/g
Fe	5.0	Fe-55	24,000	29,000
Co	2.2	Co-60	8,970	8,100
Ni	0.01	Ni-63	35	730
Zn	800	Zn-65	40	40
Cs	0.03	Cs-134	9.5	<10
Eu	0.0019	Eu-152	228	190
Eu	0.0019	Eu-154	17	<20

Further comparisons were made for the concrete from radial drilling samples near the tank wall immediately next to the radial beam tube. The activation of Co-60 and Ba-133 is compared along the axial position of samples. The comparison is shown in Figure. 3 (upper graph). The measured and calculated activities agree sufficiently well. For these samples the chemical composition of Co and Ba was obviously correctly specified. The 3D results were used for the calculation, since the drillings were in a non-symmetric position. The axial maximum of the activity does not lie in the expected position of the axial core center but rather about 36 cm lower. The reason for this was the cylinder of the filter equipment filled with He (about 36 cm below core mid-plane) between the core and the tank at the location of the radial beam tube (see also Fig.2). The neutrons from the core could pass through the He zone and activate the tank and concrete at adjacent positions. Due to the realistic 3D model of the filter equipment the calculation could reproduce the measurement very well.

For the validation of dose rate calculations the dose rate at the surface of the central irradiation tube was measured and calculated (due to the central position by flux densities of the 2D model). The results are shown in Fig. 3 (lower graph). The measured and calculated values agree for all axial positions. Further validations were made for dose rates at the surface of the stainless steel screw on the upper grid plate.

RESULTS

The results of the transport and activity calculations give an overview of the regions which contain activated materials.

Table II shows the calculated specific and total activities of the important components and materials.

Table II. Materials and activity of components

Component	Material	Specific activity [kBq/g]	Total activity [MBq]
Upper grid plate	AlMg3F18	39.0	195
Lower grid plate	AlMg3F18	52.7	422
Steel components (screws etc.)	Stainless steel	17,370.0	3,474
Graphite elements	Graphite, Al	93.4	3,267
Central irradiation tube	AlMg3F18	182.8	366
Control rods	B_4C, Al	115.0	460
Instrumentation tubes	AlMg3F18	5.0	100
Irradiation carousel	Stainless steel, Al	15,000.0	33,000
Filter equipment	Al, Pb, He, graphite	4.1	1,446
Graphite reflector	Graphite, Al	20.2	16,154
Reactor tank with radial beam tube	AlMg3F18	0.09	116
Biological shield (2D model)	Baryt concrete	0.01	30
Biological shield (3D model)	Baryt concrete	1.1	8,043

The maximum total activity of the activated components and materials of the TRIGA reactor is about 70 GBq. This value includes the measured activity of the reinforcement irons of the biological shield.

The main parts of the total activity are the irradiation carousel, the stainless steel components, the graphite reflector and the activated concrete of the biological shield. The total mass of the stainless steel components are only about 200 g, the irradiation carousel consists of about 2.2 kg stainless steel, the graphite reflector has less specific activity, but a mass of 800 kg and the activated concrete also has a low specific activity, but a heavy mass of 10,000 kg.

From the calculations the nuclide vectors were determined on the basis of the nuclide specific activities of the materials. The results are summarized in Table III. The main nuclides of the activated materials are Fe-55 for the aluminum components, Co-60 for the stainless steel, H-3 for the graphite and Ba-133 for the baryt concrete.

Table III. Nuclide vectors for the relevant materials.

Material	Nuclide	Portion [%]
Aluminum	Fe-55	71
	Co-60	27
	Ni-63	1
	Eu-152	1
Stainless steel	Fe-55	23
	Co-60	62
	Ni-63	15
Graphite	H-3	65
	C-14	11
	Co-60	6
	Eu-152	16
	Eu-154	2
Baryt concrete	H-3	15
	Fe-55	2
	Co-60	2
	Ba-133	64
	Eu-152	16
	Eu-154	1

The calculations confirm the main assumptions that only the core support is activated and the reactor tank, radial beam tube and the biological shield are components with low activation.

Fig. 4 shows an example of activation of the tank wall and the surrounding concrete. Due to the radial beam tube not only the tank wall but also the concrete of the biological shield is non-symmetrically activated . Therefore, the 3D model was used for the activity calculations.

The upper picture of Fig.4 shows the Co-60 activity of the Al tank wall in axial and azimuthal direction. The azimuth angle is measured from the center of the radial beam tube. The axial maximum lies at the location of the radial beam tube about 36

cm below the center of the core, in an opposite position (at 180 degrees) at about the height of the center of the core. The reason is the position of the He container of the filter equipment. The boundary within the material has to be dismantled (0,1 Bq/g for Co-60) can be seen in the picture.
The lower pictures of Fig. 4 show the specific activity of Ba-133 in the baryt concrete. The right one is calculated using the 2D model and shows the symmetric region of the activation in the baryt concrete. The boundary within the material has to be dismantled (1 Bq/g for Ba-133) as can be seen in the picture. The baryt concrete must be dismantled in a height of 160 cm and in a depth of maximal 35 cm.
The left picture shows the area around the radial beam tube calculated with the 3D model. The shift of the maximal activation of 36 cm below the center of the core can be seen. Compared to the symmetrical part in the right graph the amount of concrete which must be dismantled is much higher.
The complete results of the calculations allow the decision to be made as to which material has to be treated as radioactive disposal or normal disposal. The total mass of radioactive waste is about 5,000 kg, from that 2,200 kg are metals, 800 kg graphite and 2,000 kg baryt concrete.

Table IV shows examples of the results of the dose rate calculations. The reactor facility can be dismantled by hand using the usual tools. With mobile and temporary shielding the radiation protection for the working staff can be guaranteed. The individual and collective radiation exposure can be minimized.

Table IV. Dose rates at the surface and at a distance of 1 meter for some components.

Material / Location	**Dose rate in [μSv/h]**	
	At the surface	**At a distance of 1 meter**
Empty reactor tank, 1 m over bottom	5.0	2.8
Lower grid plate	6,000.0	80.0
Graphite reflector	9,500.0	900.0
Irradiation carousel	2,000.0	4.1
Central irradiation tube	600.0	3.0

SUMMARY

In preparation for dismantling the TRIGA reactor at the MHH two- and three-dimensional calculations were made in order to determine the neutron and photon fluxes inside and outside the reactor tank. With this data and the composition of the materials, activations and dose rates of the components were determined. The calculations agree well with the results of measurements.

The calculations and measurements provide a complete picture of the activity of the area near the core from the center of the core to the biological shield. Thus all the main data required for planning the dismantling techniques of the reactor and the radiation protection measures are available.

It is possible to use manual dismantling techniques and to dismantle the reactor tank and the biological shield partial.

The radiation exposure for the working stuff and pollution into environment can be minimized.

The amount of radioactive waste is low with a total mass of about 5,000 kg and a total activity of about 55 GBq. The waste will be given to an external disposal. For all the other components of the reactor facility it is possible to fulfil the requirements of the German Radiation Protection Law for unrestricted waste disposal or use of materials.

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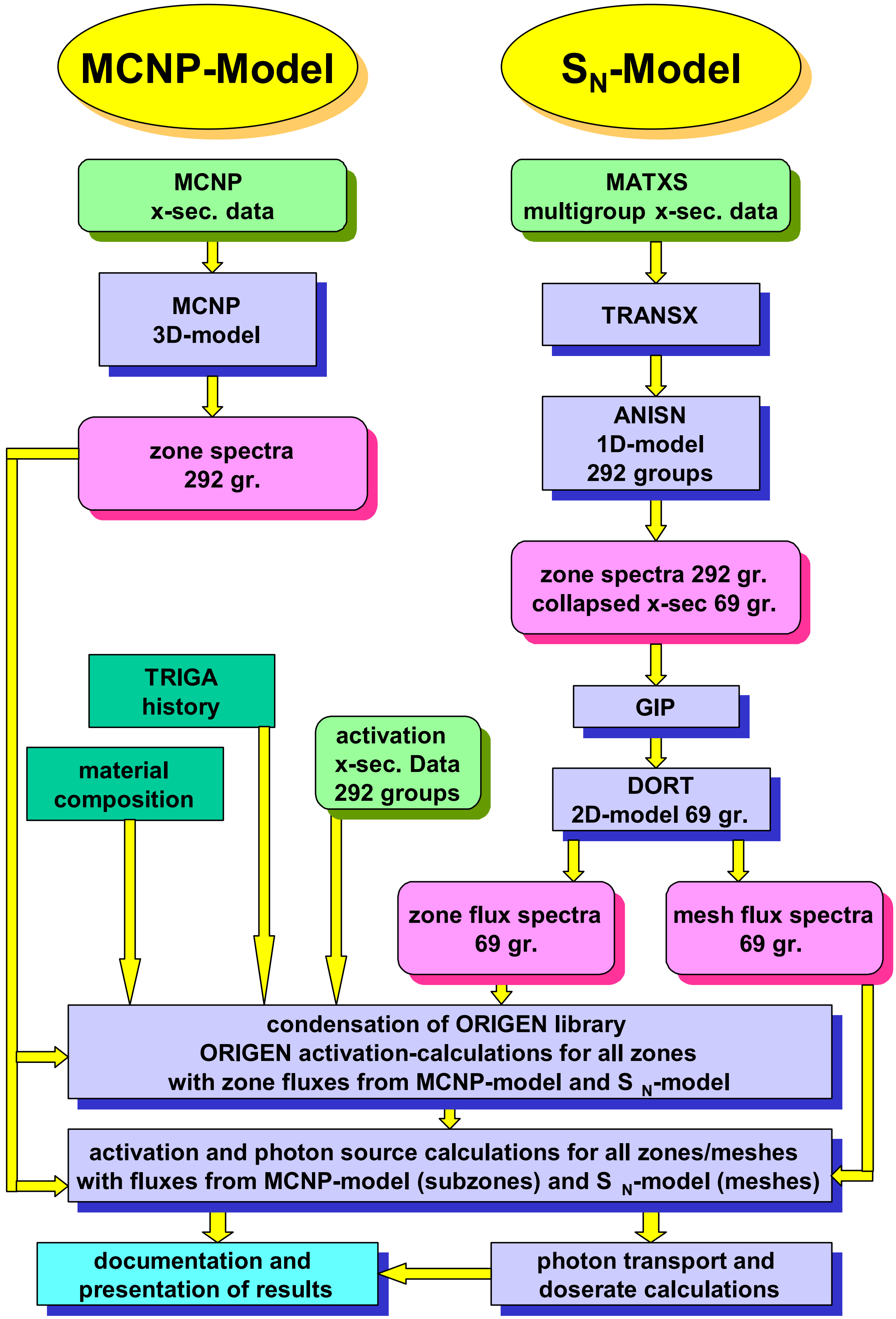

Fig. 1. Chart of calculation models, used computer codes and data flow

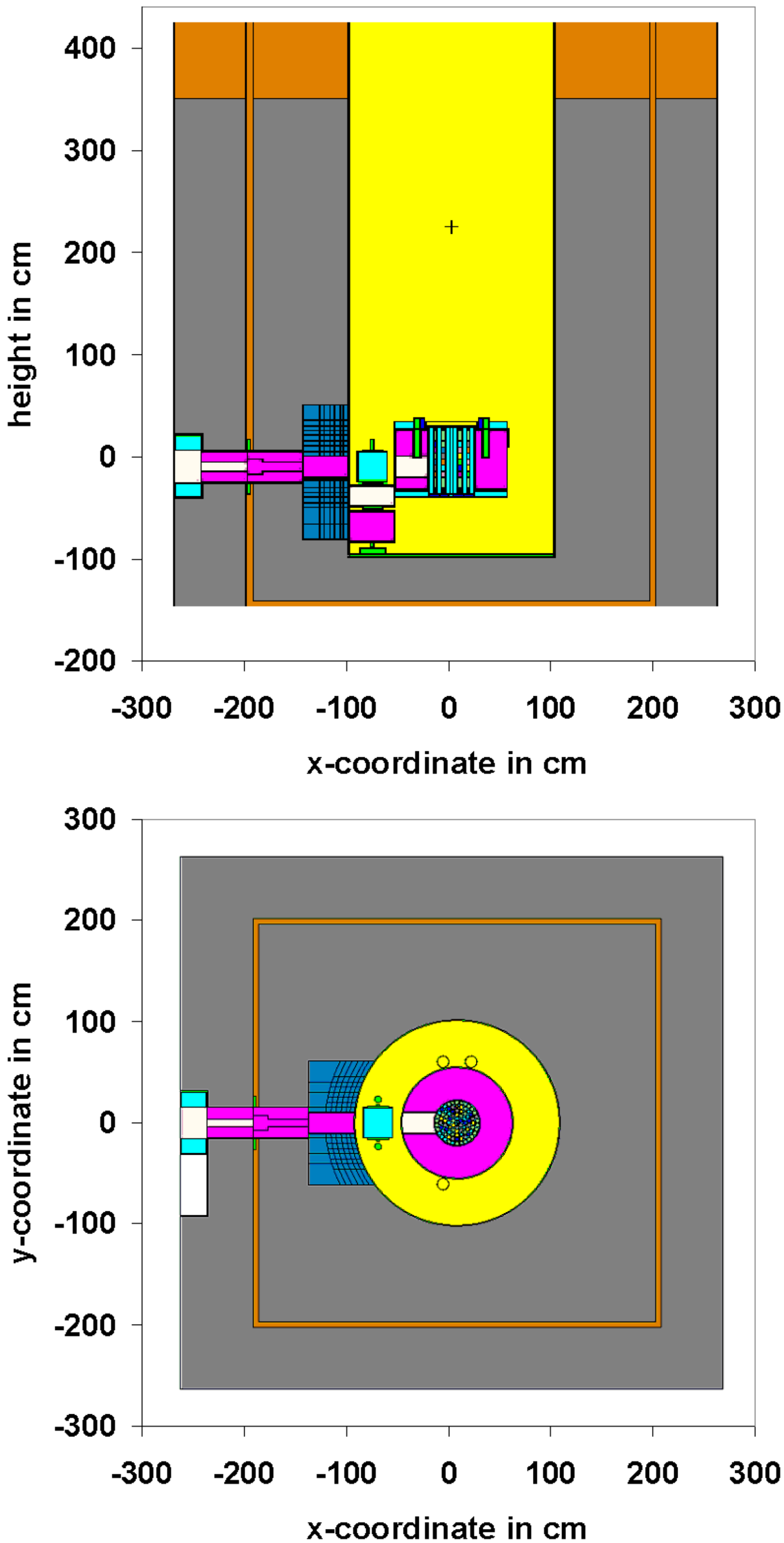

Fig. 2. 3D-model of the TRIGA geometry for MCNP-calculations. (top: vertical cross-section; bottom: horizontal cross-section in height of middle core)

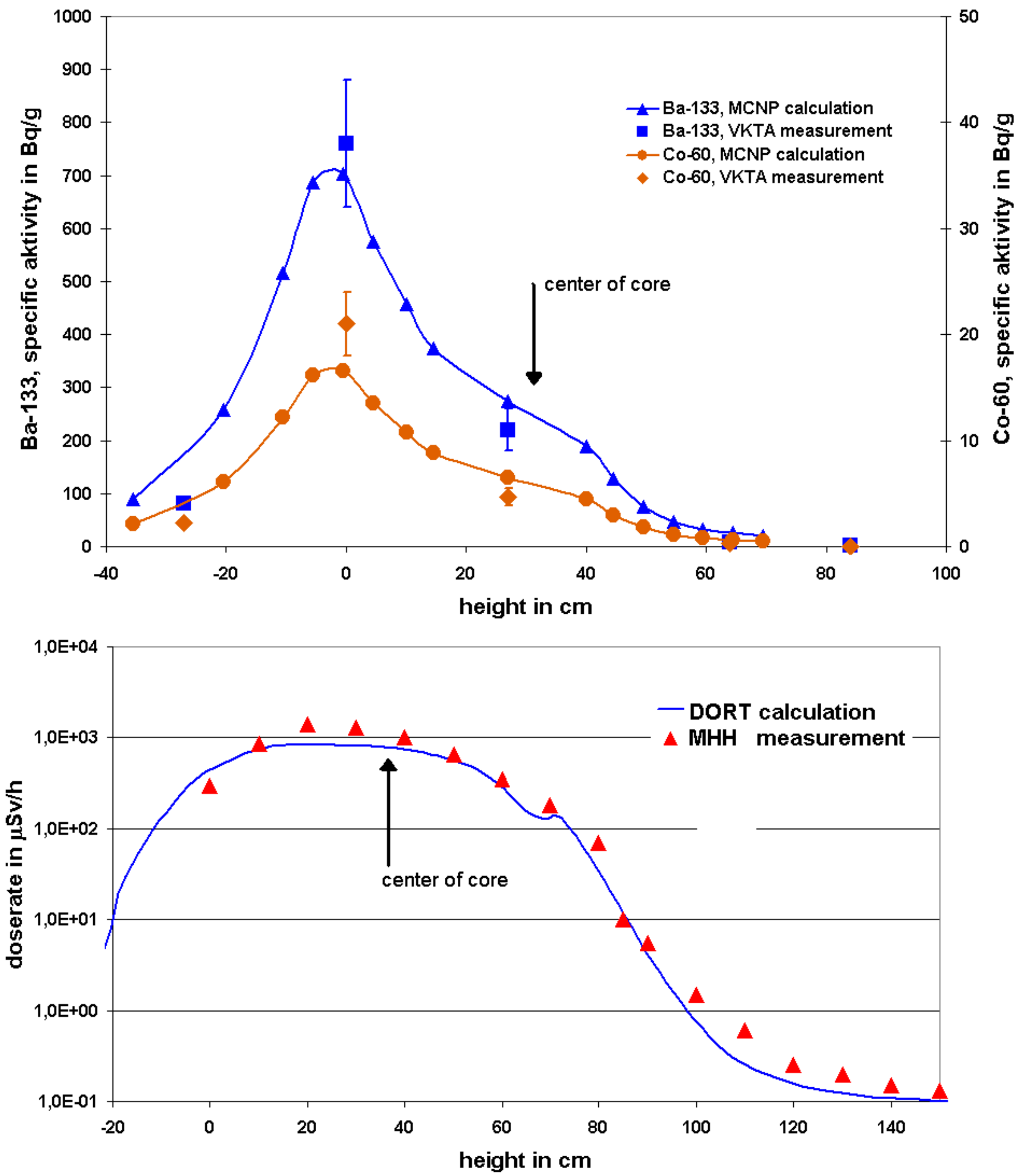

Fig. 3. Comparison of calculation and measurements. (top: Co-60 and Ba-133 activation products in heavy concrete - radial drillings; bottom: surface dose rate for the central irradiation tube)

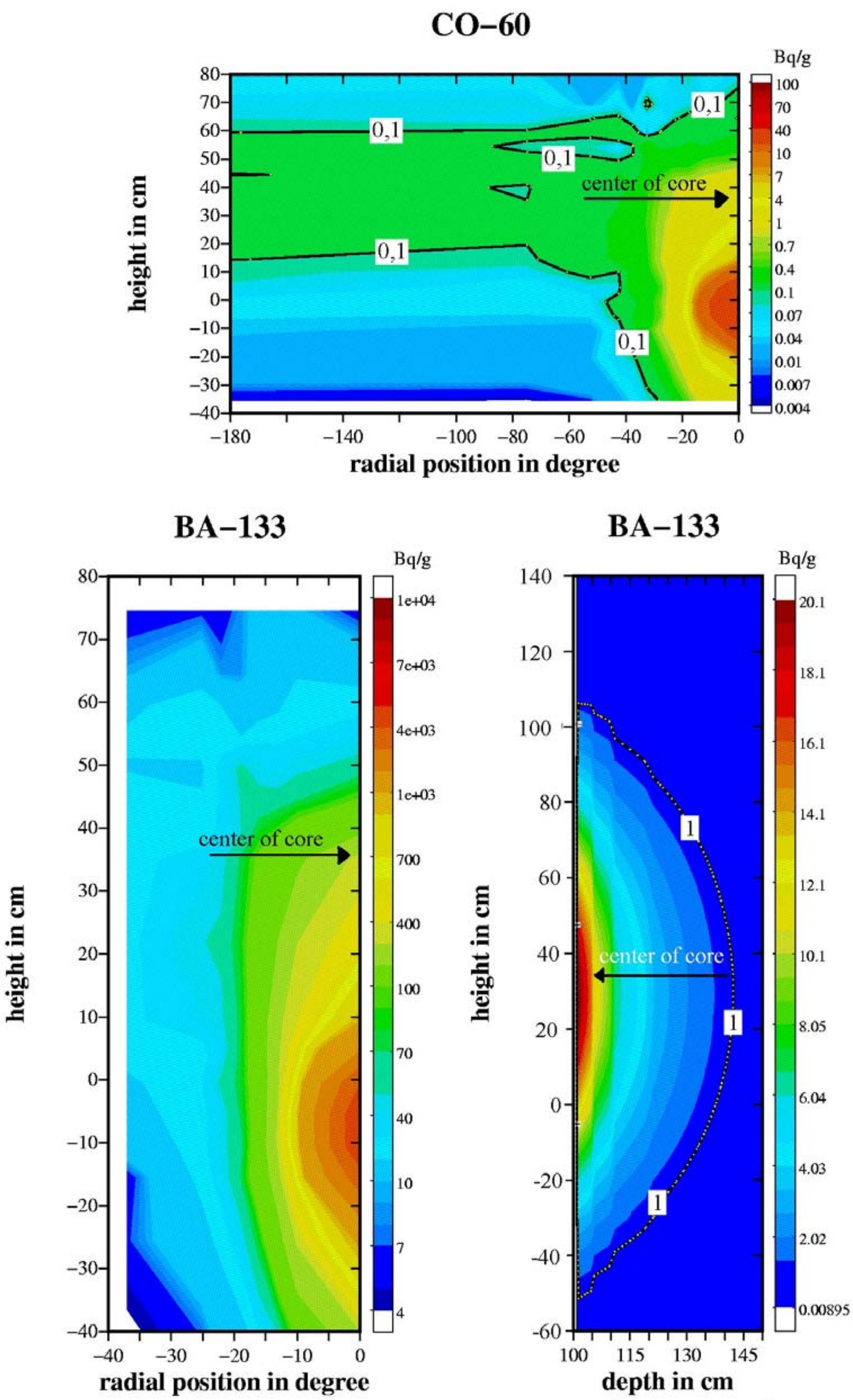

Fig. 4. Top: Co-60 activation product in the aluminium tank with boundary for dismantling, Bottom: right: Ba-133 activation in the symmetric region of the biological shield with boundary for dismantling, left: Ba-133 activation products in the top layer of heavy concrete near the radial beam tube.