Implementation, Development, Validation and First Applications of Different Code Chains for the Investigation of Accelerator-Driven Transmutation

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Abstract. For neutron physics investigations of the transmutation of nuclear waste by accelerator driven systems, a linkage of the intermediate energy Monte Carlo transport code HETC, the deterministic transport program TWODANT, and the Karlsruhe burnup system KARBUS has been developed. The program chain HETC-TWODANT has been validated against HETC-MCNP for an iron assembly and for a subcritical core with $Th^{232} - U^{233}$ fuel. Satisfactory agreement is found for one group cross sections, total neutron flux densities and neutron flux density spectra. As a first application, HETC-TWODANT-KARBUS has been applied to investigate the capabilities of plutonium incineration in a sodium cooled accelerator driven subcritical core with $Th^{232} - Pu$ - fuel and 3000MW of thermal power.

INTRODUCTION

The neutron physics investigations of Accelerator Driven Systems (ADS) have been started at Forschungszentrum Karlsruhe (FZK) in autumn 1991. This work is part of a more general activity that studies the capabilities of transmutation of nuclear waste and transuranics, especially the capabilities of the incineration of Pu in Pressurized Water Reactors (PWR), Fast Breeder Reactors (FBR), and in Accelerator Driven Systems [1].

The first step for establishing a program system for the neutron physics calculations was the implementation of the modules HETC and MORSE of the HERMES system [2], developed at Forschungszentrum Jülich. The program sequence HETC-MCNP-KORIGEN (PROSDOR) [3] has been developed at FZK. With HETC-MORSE and HETC-MCNP-KORIGEN we participated in an international Benchmark on the calculation of thick lead and tungsten targets [4]. The system PROSDOR has also been applied for the calculation of transmutation of actinides and fission products [3].

Recently a linkage between HETC, the deterministic transport program TWODANT [5] and the Karlsruhe burnup system KARBUS [6] has been established as an alternative that needs less computer time and therefore is a good tool for routine and survey calculations. HETC-MCNP will be used for control calculations in special cases. HETC-TWODANT has been tested in comparison with HETC-MCNP for an iron assembly and for a subcritical core with $Th^{232} - U^{233}$ fuel. As a first application, HETC-TWODANT-KARBUS [6] has been used to investigate the capabilities for Pu incineration in an accelerator driven subcritical core with Th-Pu solid fuel and Na-coolant.

THE LINKAGE HETC-TWODANT AND VALIDATION FOR AN IRON ASSEMBLY

The neutron source generated by HETC is fed into TWODANT as a full space-energy distribution, where the kinetic energy is specified by the energy group structure and the spatial distribution is specified by volume elements that are defined by the fine mesh intervals of the TWODANT input. The anisotropy is taken into account approximately; control calculations with MCNP with isotropic and with anisotropic neutron source showed that in our applications, the influence of the anisotropy of the source was small. In order to separate in a first study the influence of different codes from the influence of different nuclear data we started with an iron assembly that was hit by a proton pencil beam of 800 MeV. Nuclear data for iron based on EFF-1 [7] are available as library for MCNP and as group constants for use in TWODANT. The relative deviations between calculations with HETC-TWODANT and HETC-MCNP for one group cross sections are less than 20% except for threshold reactions, for the (n,2n) cross section the relative deviation is about 70 %. For the neutron flux density (Φ) the relative deviation is about 20%. Moreover, by use of different cross section sets it was shown that the deviations, caused by the application of different codes are comparable to the differences that are caused by the use of different nuclear data.

VALIDATION OF HETC-TWODANT FOR AN ACCELERATOR DRIVEN SUBCRITICAL CORE

These comparisons are based on a water moderated cylindrical core (radius=64cm, height=132cm) with $Th^{232}-U^{233}$-fuel. The energy of the proton beam was 1.5 GeV. The calculations have been carried out for the voided state in order to minimize in the comparisons possible difficulties in MCNP-3A in the statistical resonance region. The nuclear data for the Monte Carlo calculations were taken from existing MCNP-3A libraries, based on ENDF/B-IV and ENDL. For the calculations with TWODANT, data from a 69 energy group constant set, mainly based on the Karlsruhe nuclear data library KEDAK-IV have been used. The group constants for U^{233} have been newly calculated from ENDF/B-VI and those for Th^{232} from JEF-2.2. The comparisons have been carried out for calculations with an external neutron source and for criticality calculations. The results for k_{eff}, for the core averaged neutron flux density and for the microscopic one group cross sections of Th^{232} are shown in Table I. The relative deviations of the one group cross sections for Th^{232} are less than 17 %, except for inelastic scattering and for the (n, 2n) -reaction. The difference in the inelastic scattering group cross section may be partly caused by the difference in nuclear data (only continuum inelastic scattering is given for Th^{232} on the MCNP library used, excitation of discrete levels and continuum inelastic scattering on the group constants library). The relative deviations for the (n,2n) reaction are larger. Threshold reactions are very sensitive on the weighting spectra used in the group constant generation and also strongly influenced by the upper energy boundary of the group constant set. This has to be kept in mind if the production of isotopes via (n,2n) reactions is discussed e.g. the generation of U^{232} via the $Th^{232}(n,2n)$ and $Pa^{231}(n,\gamma)$ reactions. Table I also shows that one group cross sections obtained from calculations with external neutron source and those from criticality calculations differ by 4% or less except for the (n,2n)-reaction. The conclusions are similar for the other isotopes of the core (U^{233}, Zr_{nat}, O^{16} and H^1). The differences in Φ are about 40% corresponding to the differences in $\frac{1}{1-k_{eff}}$. (The values for Φ shown in the table are the neutron flux densities per source neutron resulting from the MCNP or TWODANT calculation without further normalization). The proton current necessary to generate $300MW_{th}$ is 8.84 mA as result of TWODANT and 4.89 mA as result of MCNP, where the total fission rate per source neutron obtained from TWODANT is 4.198 and the one resulting from MCNP is 7.589 and the average number of neutrons produced per proton is 40.4 as a result of the HETC-step in both calculation paths. For the total neutron flux density in the $300MW_{th}$ core $1.76 \cdot 10^{15}$ $\frac{n}{cm^2 sec}$ are derived from calculations with TWODANT and $1.60 \cdot 10^{15}$ $\frac{n}{cm^2 sec}$, from calculations with MCNP.

FIRST APPLICATION OF THE PROGRAM CHAIN HETC-TWODANT-KARBUS FOR THE INVESTIGATION OF PU-INCINERATION IN AN ACCELERATOR DRIVEN SUBCRITICAL CORE WITH TH-PU-FUEL

As a first application the program sequence HETC-TWODANT-KARBUS has been used to investigate the capabilities of Pu - incineration in an accelerator driven subcritical core of 3000MW thermal power with $Th^{232} - Pu$ - fuel and sodium coolant. The radius of the core was 110 cm, its height 220 cm, the primary energy of the protons was 1.6 GeV. Burnup calculations have been carried out for $3 \cdot 365$ days. Within each period of 365 days full power operation has been assumed for $3 \cdot 110$ days and zero power for 35 days. The depletion and buildup of isotopes during $3 \cdot 365$ days is shown in Figure 1 for U^{233}, Pu^{239} and Pa^{233}. The dotted and dot-dashed lines show the results, when spectra of MCNP calculations are used instead of TWODANT spectra. The total amount of Pu incinerated within $3 \cdot 365$ days is found to be 1940 kg corresponding to 600 kg/GW(e)a (efficiency assumed: 40%). During the same time 1470 kg of U^{233} are produced corresponding to 450 kg/GW(e)a. Th^{232} is reduced from 19100 kg in the fresh core to 16200 kg at the end of the burnup time discussed. The proton current necessary to obtain 3000 MW_{th} power is 21.5 mA, 18.9 mA, 25.4 mA and 40.8 mA for the fresh core, after 365 days, 730 days and 1065 days of burnup; the total neutron flux density is between $5.97 \ 10^{15}$ and $5.46 \ 10^{15}$ $\frac{n}{cm^2 sec}$, the results for k_{eff} for the different burnup states are: .9785, .9804, .9737 and .9585. The core stays subcritical, when it is voided and also when the temperature is decreased to 300 K. $(\frac{\Delta k}{k})_{void}$ is between $-1.2 \ 10^{-2}$ and $-1.7 \ 10^{-2}$ and $(\frac{\Delta k}{k})_{933 \to 300K}$ is between $8.3 \ 10^{-3}$ and $6.1 \ 10^{-3}$.
Figure 2 shows the volume averaged neutron flux density spectra for the fresh core and after 1095 days of burnup. The Pu^{239} resonance at about 0.3 eV, the Pu^{240} resonance at about 1 eV and the 3 keV sodium resonance are clearly recognized.

CONCLUSION

For neutron physics studies of accelerator-driven transmutation, a code chain HETC-TWODANT-KARBUS has been developed. The linkage HETC-TWODANT has been tested in comparison to HETC-MCNP. For an iron assembly, the relative deviations between results of the two codes are about 20% for the total neutron flux density, and less than 20% for one group cross sections except for threshold reactions. In a second comparison, a subcritical core with Th^{232} - U^{233} - fuel and a hard neutron spectrum has been studied. The relative deviations for the one group cross sections of the individual isotopes were less than 20% except for threshold reactions. The largest relative deviation is found for the inelastic scattering of U^{233} (86%). This large deviation is probably due to differences in the nuclear data bases of the cross section libraries used by the two codes. For the total neutron flux density per source neutron, a relative deviation of about -40% was found, corresponding to a deviation of about -4 % in k_{eff} (relative to MCNP). As a first application of the new code chain, the incineration of Pu in a Th^{232} - Pu core of $3000 MW_{th}$ has been investigated. In this core about 1.9 tons of Pu are incinerated during an operation time of 3 years. During this period about 2.8 tons of U^{233} are produced: about 1.5 tons are contained in the irradiated fuel and about 1.2 tons are in-situ fissioned (about 0.1 tons of the U^{233} are transmuted into U^{234}). The Th^{232} is reduced from about 19 tons in the fresh core to about 16 tons after 3 years, where 0.1 tons of this reduction result from in-situ fission.

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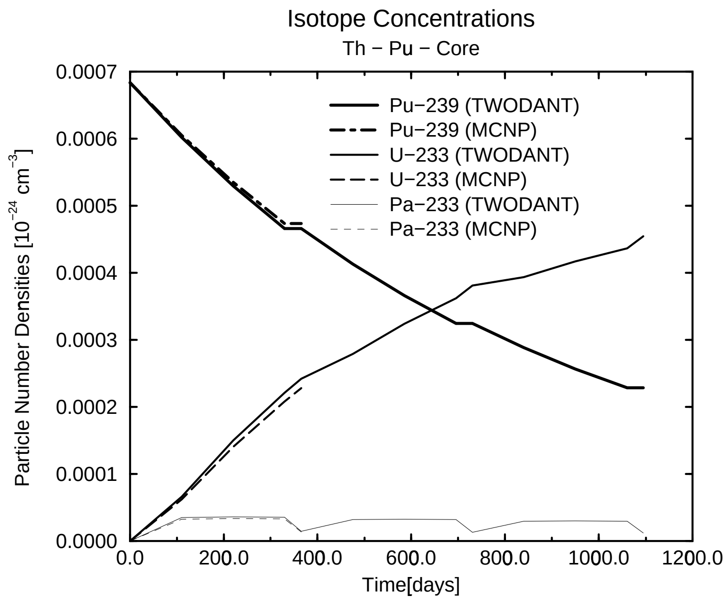

Figure 1: Development of the isotopic concentrations of Pu^{239}, U^{233}, and Pa^{233} during burnup of a sodium cooled Th^{232} - Pu core of $3000 MW_{th}$

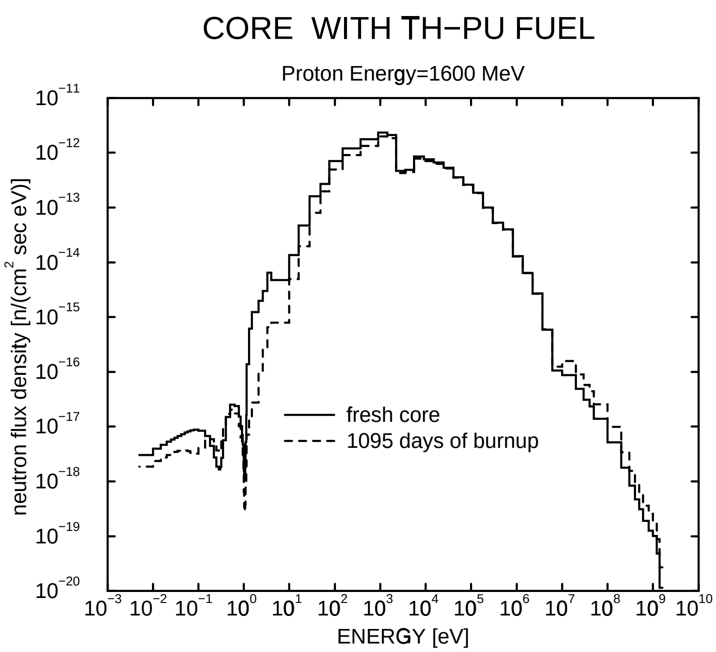

Figure 2: Neutron flux density spectrum for a sodium cooled Th^{232} - Pu core for different burnup states (normalized to $\sum_{g=1}^{G} \Phi_g = 1$, $\Phi_g =$ group flux in energy group g)

Table I : Comparison of k_{eff}, neutron flux densities and microscopic one group cross sections for Th^{232} from calculations with HETC-TWODANT and HETC-MCNP for a subcritical Th^{232} - U^{233} core

	TWODANT 69 groups $S_{16}P_2$ k_{eff}	**TWODANT 69 groups** $S_{16}P_2$ external source	**MCNP-4a** k_{eff}	**MCNP-3a** external source	$\frac{T-M}{M}$% [1)] external source
k_{eff} / M[2)]	0.90345	0.91422 M=7.4853	0.91834 [3)]	0.95033 M=12.442	-3.8 -39.8
$\frac{1}{1-k_{eff}}$	10.358	11.657	12.246	20.134	-42.1
$\Phi[\frac{n}{cm^2 \cdot sec}]$	6.7113E-5	7.8754E-4	6.3555E-5	1.2924E-3	-39.1
microscopic one group cross sections for $Th^{232}[barn]$					
σ_{tot}	9.2046	9.2115	1.0539E+1	1.0539E+1	-12.6
σ_{el}	7.7339	7.7418	9.2476	9.2178	-16.0
σ_{fiss}	1.5341E-2	1.5784E-2	1.5901E-2	1.6442E-2	-4.0
$\sigma_{(n,\gamma)}$	2.4010E-1	2.4043E-1	2.8799E-1	2.8867E-1	-16.7
σ_{inel}	1.2125 [4)]	1.2097 [4)]	9.8499E-1 [5)]	9.7975E-1 [5)]	23.5
$\sigma_{(n,2n)}$	2.7219E-3	3.8139E-3	2.5232E-3	5.4800E-3	-30.4
ν $[neutrons/fission]$	2.3369	2.3605	2.0669	2.1039	12.2

1) T=TWODANT M=MCNP

2) the values of k_{eff} for the calculations with external neutron source have been obtained by the following formula, given in [8]

$$k_{eff} = \frac{M - R^{(0)}(\sigma_{n,xn}) - 1}{M - \frac{1}{\nu}} \text{ , where } M = R(\nu\sigma_{fiss}) - R(\sigma_{fiss}) + R(\sigma_{n,xn}) + 1$$

R : reaction rate, $R^{(0)}$: reaction rate from criticality calculation

3) the estimated relative errors in the MCNP calculations are between 0.002 and 0.03, which, according to [8], means reliable results

4) excitation of discrete states of the residual nucleus and of the continuum

5) continuum inelastic only