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The use of fission product nuclear data by D. Zappe, V. Schuricht, F. W. Krüger and M. Gegusch

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1. Introduction

The solution of various problems in the field of radiation protection, nuclear safety and dosimetry requires the knowledge of the properties of radiation sources and fields caused by fission products. From 1970 to 1976 in the VEB Kombinat Kraftwerkss-lagenbau by order of the National Board on Atomic Safety and Radiation Protection and at the Technical University Dresden extensive investigations were performed for that purpose. In the following a review about methods and results of these investigations and about their applications are given. Furthermore experiences about handling of fission product nuclear data and requirements for improvement of these data are discussed.

2. Investigations of the properties of fission product mixtures

In the frame of the investigations various computer codes were developed. They serve for assessment and designing of radiation protection provisions in and around nuclear installations, for evaluation of radiation exposure and for calculation of measuring effects of radiation detectors and dosimeters.

With the aid of the computer code AKTIVIST III it is possible to calculate several properties of fission product mixtures on the basis of constant power /1,2,3/. The input data, such are type of fission, neutron energy, neutron flux density and irradiation and decay time, allow to calculate the following quantities of

fission products: mass (kg/NW), activity (Bq/W or Ci/W), beta decay energy (W/W), heat after irradiation of the fuel elements - the afterheat - (W/W), for individual nuclides and elements of the fission product mixture in relation to the thermal power of the reactor, and the gamma-ray spectra for groups of elements, divided into 31 and 5 groups of energy between 0 and 5.25 MeV.

The computer code AKTIVIST III is based on a data set containing the following fission product data /9,10,24/: - independent fission yields for the fission of 235 U and 239 Pu

by thermal neutrons, of ²³⁵U, ²³⁸U and ²³⁹Pu by fast neutrons

with the yield for the direct formation of a nuclide and the cumulative yield for two parallel precursors,

- effective cross sections for thermal neutrons (calculated from the σ_{2200} cross section and the resonance integral) and for rast neutrons (calculated from 26-group cross sections and the neutron spectrum),
- decay constant of the nuclide and of two precursors,
- transition probabilities in the case of multiple decays from the and into the corresponding nuclide,
- beta- and gamma-decay energies and
- gamma-emission probabilities for 31 energy groups.

With the aid of this code for a large interval of input parameters chemical composition and decay properties of the nuclide mixtures formed by thermal and fast neutron fission were calculated /1, 2/. Furthermore a comparison of the activity of the fission products, of the afterheat and of the gamma spectra was carried out for these two types of fission /3/.

An additional computer code /4/ also allows the calculation of the activity of individual fission products and of various fission product mixtures, the determination of energy spectra of fission product mixtures of different composition (i.e. of different decay time and type of fission) and the calculation of other quantities for prompt fission, derived from the mentioned quantities or functions. For the calculations the following fission product nuclear data were used:

- data on decay chains from MARTYNENKO /5/, GUSEV /6/ and DAVIES /7,8/,
- fractional independend yields from DAVIES /7,8/ and MEEK /9/,
- gamma spectra of the individual nuclides from LEDERER /10/, DA- VIES /7,8/ and CROCKER /11/ as well as
- beta decay properties from several authors.

This code allows the numerical calculation of complex (branched) decay chains basing on the analytical solution of the differential equation for linear decay chains. By the choice of various initial conditions or by weighing the results for individual radionuclides, respectively, it is possible to realize by calculation different conditions of formation and also other alterations of the composition of the fission product mixtures (e.g. in consequence of the escape of inert gases etc.).

The development of a rather universally utilizable organization system for the supply of nuclear data from several references for defined calculation jobs will be finished in the near future /12/. With the aid of this system it will be possible to carry out also comparisons of large amounts of data in respect to the influence of uncertainties of nuclear data.

Pission product mixtures are high intensive radiation sources which must be shielded in nuclear installations and in transport and storage bins. The laying out of such shieldings requires the knowledge of gamma spectra. For small and intermediate decay times relatively small energy groups are necessary. With increasing decay time the mean energy strongly decreases. After several years, finally, and for fission by thermal neutrons only the gamma radiation of ¹³⁷Cs (0.67 MeV) is predominant. For fission by fast neutrons the spectrum is, owing to the major portion of ¹³⁴Cs and ¹⁵⁴Eu. harder. The estimation of transport bins for irradiated fuel requires to take into account besides the gamma radiation also the neutrons, emerging from the spontaneous fission of transuranium elements and from (og, n) reactions. Furthermore gamma spectra were used for comparisons with the results of other authors (e.g. CROCKER /13/, MARTYNENKO /5/, GUSEV /14/; see /15,16,21/) and for the calculation of the exposure rate /17/and of radiation fields /18/ as well as for the investigation of impulse height distributions /19,20/ for radiation protection and dosimetry purposes.

The knowledge of the calculated afterheat is necessary for assessments in the case of transport of irradiated fuel, for the storage of fuel elements and for calculation of the temperature of stored radioactive wastes. These calculations are also important for the assessment of the reactor performance in accident situations.

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The composition of activity and the chemical properties of the fission products must be known for estimating contamination processes and for affecting decontamination operations. These informations are also of importance for the activity releases under accident conditions, for the hazards emerging in such cases and for the accident control. Therefore, specifications about the activity of individual chemical fractions (e.g. inert gases or halogens) are used for accident analyses and for selection of the mest effective arrangements for decontamination.

3. Discussion of results

The gamma spectra of fission product mixtures from different fission reactions (various fissionable materials and energy of neutrons) only slightly differ in the case of prompt fission. For the most part these differences are smaller than the differences of gamma spectra, emerging in the same fission reaction, but given by different authors. Therefore, for radiation protection purposes it is sufficient to assume a "standard" mixture of fission products and to take into account significant alterations as correotions for this standard mixture. This is true especially for predictions about the exposure rate, for the attenuation of radiation by shields etc. Differences arising by use of various models and nuclear data give deviations e.g. in the exposure rate behind a shield out of normal concrete to a maximum of 10 % /21/. Detailed investigations, e.g. for the determination of impulse height distributions or other characteristics of radiation detectors or dosimeters, require the use of individual gamma spectra of mixtures. To that end a completion of the gamma spectra of individual nuclides (absolute yield of a transition) as well as a more precise informations about half-lifes, branching probabilities and fission yields of several nuclides are necessary (see also /22/).

Furthermore investigations of chemical composition and of decay properties of fission product mixtures show that only for fission by fast neutrons and for detailed examinations it is necessary to take into account the influence of (n, 2n) reactions because the

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threshold energy of these reactions is far above the probable energy of neutrons. (n,p) and (n, α) reactions can be neglected. Bremsstrahlung arising from beta-active fission products exceeds for mixtures of fission products and fuel for large decay times in several energy intervals the primary gamma radiation or causes a smoothing of the spectra. Especially the contributions of 90 Y, 106m Ru and 144 Pr must be considered. The shielding of burned-up fuel material is hardly influenced by bremsstrahlung because the contribution to exposure rate behind the shield in general don't reach 10 f.

In .iew of the described investigations the following demands on fission product nuclear data appear to be realistic:

- For decay times from several months to several years ^{7,24}Cs gives an important portion to the gamma radiation of fission product mixtures. The data on the production of ¹³⁴Cs at the present time considerably differ and should be given more precisely.
- The decay data for long-living nuclides which are important for waste disposal should be given in a more precise manner and should be completed.
- A more precise knowledge about the beta spectra of fission products is necessary in order to get better information about decay power and about the influence of bremsstrahlung on the gamma spectra. It should be tried, similar to the gamma radiation, to give also for beta radiation the emission in the individual energy groups. To that end several factors (form factor, Coulomb correction factor) should be known more precisely (see also /23/).
- Fission yields for several nuclides with low absolute yields and larger half-lifes (months or years) should be given more precisely.
- Informations about the absolute gamma-ray yield for some nuclides, e.g. ¹³⁹Xe, ¹⁴⁰Xe, ¹⁴²Xe, ¹⁴¹Ba, ¹⁴²Cs and ¹⁴³La, are necessary.
- In most of the references about fission product nuclear data there are no informations about the accuracy of the data which

would only allow an assessment of the accuracy of the results obtained by using these data /15,21/. Such informations would be very useful.

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