THE CONSTANTS AND PARAMETERS OF NUCLEAR STRUCTURE AND NUCLEAR REACTIONS

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INTERACTIVE INFORMATION SYSTEM ON THE NUCLEAR PHYSICS PROPERTIES OF NUCLIDES AND RADIOACTIVE DECAY CHAINS

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INTERACTIVE INFORMATION SYSTEM ON THE NUCLEAR PHYSICS PROPERTIES OF NUCLIDES AND RADIOACTIVE DECAY CHAINS. A brief review is given of a computerized information system on the nuclear physics properties of nuclides and radioactive decay chains. The main difference between the system presented here and those already in existence is that these evaluated databases of nuclear physics constants are linked to a set of programs, thus enabling analysis of a wide range of problems regarding various nuclear physics applications.

An interactive information system (IIS) has been set up containing comprehensive information on the characteristics of more than 2600 nuclides of the 107 chemical elements known to exist in 1999. The following characteristics are given for each nuclide: charge, mass, spin and parity of the ground state, mass excess, half-life, modes of decay and branching ratios, average decay energies of gamma-, electron-, positron- and alpha-radiation, and also the spectra of gamma and X-rays and of beta and alpha particles.

The main difference between this IIS and the widely available charts of the nuclides, Refs 1-4, reference books, Refs 5 and 6, and computerized systems, Refs 7 and 8, is that these evaluated databases of nuclear physics constants are linked to a set of programs, making it possible to:

- automatically plot and visualize radioactive decay chains;
- calculate the number of nuclei, activity and radiation energy release, for both the chain as a whole and the individual nuclides in it;

- calculate the spectra of gamma- and X-rays, and of alpha and beta particles in the chain, at a given moment in time;

- identify radionuclides from the measured gamma spectra; identification can be carried out using the half-life, gamma-radiation energy or a combination of these two parameters;

- analyse the spectra of individual radionuclides and radioactive chains, which allows radiation spectral lines to be selected for a given energy and intensity threshold.

The calculations can be made both for a "free" decay chain (for a given number of nuclei in the parent radionuclide chain at the starting point) and also when the number of nuclei in the parent radionuclide is supplemented from outside at a given rate ("target activation"). This algorithm enables the calculational capabilities of the IIS to be used in a wide range of tasks requiring analysis of radioactive decay chain parameters because it does not depend on the way in which the nuclei of the parent radionuclide are supplemented.

A user-friendly feature of the IIS is that, apart from information on nuclides and their decay chains, it also contains additional information: it gives the main physics properties of all the elements in the periodic table; fundamental physics constants; or relations between the various measurement units; information from the SI system for all the units used in nuclear physics and its applications; the binding energies for all the nuclides; and the energies and thresholds of any nuclear reaction can be calculated.

All the information in the IIS is presented in numerical or graphical form. The IIS also has a comprehensive HELP function.

1. The main principles underlying radioactive decay chain analysis and calculation

In setting up the IIS on the nuclear physics properties of radioactive chains we had to solve several important problems relating to the automatic plotting of decay chain diagrams, the calculation of radiation spectra and the determination of absorbed doses. Conventional numerical methods for computing the differential equation systems which describe radioactive decay chains, make huge demands on machine resources, even when high-speed computers are used. We describe below a calculation method which has made it possible to set up a fast-acting program package for analysing radioactive chains, and calculating the radiation spectrum and energy release.

To set up the information system we analysed all the up-to-date material and, after initial analysis, took the main data from Refs 5, 7 and 8.

1.1. Plotting radioactive decay chains

Hereafter, in accordance with the conventional notation system, we will identify nuclides by the signature Z-A-I, where Z is the nuclear charge of the nuclide, A is the mass

number and I is the isomer classification ('g' is ground state, 'm','m1' is first excited state and 'm2' the second, etc.).

In general terms, plotting a radioactive decay chain is formulated as follows: finding, for a given radionuclide (Z-A-I)0, all the nuclides which are formed during its decay process.

We note that, from the mathematical point of view, the decay chain is represented by a directed graph. The component nuclides of the chain are the vertices of the graph and the edges correspond to the mutual transformations of the nuclides in the decay process. A feature of this graph is that it always has one initial vertex, representing the parent radionuclide of the chain, and one or more pendant vertices, representing the stable nuclides into which the given chain decays. The edges of the graph run in the direction from the parent to the daughter nuclide and never form cycles. In this graph there is always at least one edge sequence from the initial vertex to any of the pendant vertices.

The task of plotting a decay chain is thus one of plotting this directed graph.

The algorithm for solving this task is recursive. Let us assume that we need to plot the decay chain of the nuclide Z-A-I. Each step of the recursion includes the following points:

1) Nuclide Z-A-I is added to the list of vertices already plotted; if the nuclide under investigation is stable or is already in the list, plotting is complete and we return to the previous step of the recursion;

2) If nuclide Z-A-I is unstable then, a list of daughter nuclides (for the given nuclide) is drawn up from data on its decay modes;

3) Points 1-3 are carried out for each nuclide in the list of daughter nuclides obtained in point 2.

As a result of plotting we obtain a list of graph vertices and lists of daughter nuclides for every vertex. Thus the directed graph corresponding to the decay chain of nuclide Z-A-I is fully determined. It should be noted that for point 2 of the algorithm information is needed on how to identify the daughter nuclide $(Z\text{-}A\text{-}I)_d$ from the known parent nuclide $(Z\text{-}A\text{-}I)_m$ and its decay mode. This information can be obtained from nuclear physics data on radionuclide decay.

1.2. Visualization of decay chains

The next task when plotting and calculating radioactive decay chains is placing the graph describing the chain on a plane for representation on a computer screen or printing it out in the usual way.

A fact that should be used when doing this is that a system of co-ordinates is already given for the component nuclides of the chain. This is the three-dimensional co-ordinate system (Z,A,I). Visualizing the chain is thus reduced to displaying the three-dimensional arrangement of the nuclides in the chain on a plane (X,Y).

Let us consider the algorithm below as a solution to this problem. It makes use of the fact that the range of values along the I-axis in the three-dimensional space (Z,A,I) is limited (according to available data) to three values:

(a) I='g' or is absent;
(b) I='m','m1';
(c) I='m2'.

Let us place a grid (Z,A) on the plane and arrange the nuclides in the chain in the cells. It may be that there will be two or three isomers in some of the cells. (As already mentioned, isomers are characterized by the same Z-A and their differences are shown in the I classification). In this case we will increase the width of the column and the line height of the (Z,A) grid containing the relevant cell to accommodate the number of nuclide-isomers. We will place the isomers in the cell diagonally from the bottom right to the top left-hand side starting with the lowest isomeric state number. Figure 1 gives an example of the ^{136}Te decay chain arrangement.

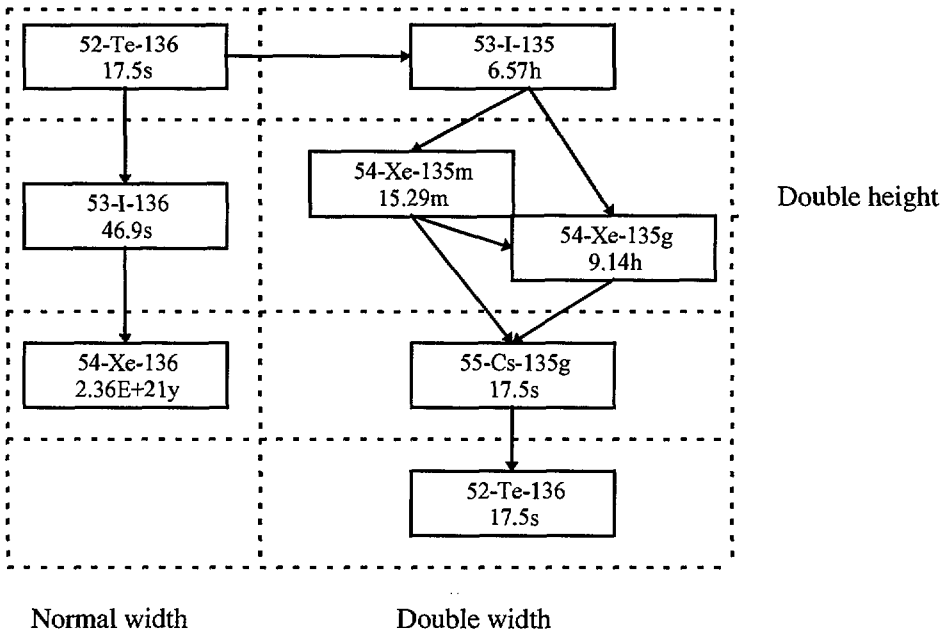

Fig. 1. Example of the Te^{136} decay chain arrangement.

1.3. Calculating the number of nuclei and the activity of the nuclides in the chain

The decay of a radioactive chain is described in general terms by the following system of differential equations:

$$\begin{cases} \dfrac{dx_1}{dt} = -S_1 x_1 \\ \dfrac{dx_n}{dt} = \sum_{i \neq n} S_n^i x_i - S_n x_n, n = 2..N \end{cases} \tag{1}$$

where N is the total number of nuclides in the chain; x_n is the number of nuclei of the nth nuclide; and S_n is the decay rate of the nth nuclide. In the case of radioactive decay, $S_n = \lambda_n$ is the decay constant of the nuclide n; S^i_n is the rate at which nuclide n is formed from nuclide i. If nuclide n is not the daughter for i, then $S^i_n = 0$, if n is the only daughter nuclide of i, then $S^i_n = \lambda_i$, otherwise $S^i_n = \lambda_i k^i_n$, where k^i_n is the branching coefficient for decay of the parent nuclide i, leading to nuclide n.

The initial conditions for this system are written as:

$$\begin{cases} x_1(0) = x_{1,0} \\ x_n(0) = 0, n = 2..N \end{cases} \tag{2}$$

We note that for a linear chain the equation system (1) becomes:

$$\begin{cases} \dfrac{dx_1}{dt} = -S_1 x_1 \\ \dfrac{dx_n}{dt} = \sum_{i < n} S_n^i x_i - S_n x_n, n = 2..N \end{cases} \tag{3}$$

and has the analytical solution:

$$\begin{cases} x_1(t) = x_{1,0} \cdot \exp(-S_i t) \\ x_n(t) = x_{1,0} S_2^1 ... S_n^{n-1} \cdot \sum_{j=1}^{n} \dfrac{\exp(-S_j t)}{\prod_{k \neq j}^{n} (S_k - S_j)}, n = 2..N \end{cases} \tag{4}$$

This can be used in solving the general task of (1) and (2).

Assuming that we need to calculate the number of nuclei of a certain radionuclide X in the chain, then we must first identify all the possible edge sequences for getting from the parent nuclide of the chain to nuclide X. Thus, we will select all the possible linear chains leading to the formation of X. Next, for every linear chain obtained, we calculate the number of nuclei in nuclide X using the formulas in (4) and adding the results together. This method makes use of the fact that different modes of nuclide decay occur independently.

We note the singularity of solution (4) if there are two nuclides with identical decay rates (S_k=S_j) in the chain. The simplest way out of this situation is to change one of the decay rates by a negligible amount within its error margin (by say 0.01%).

Let us now consider the calculation for a chain where the number of nuclei of the parent nuclide is supplemented from outside during the decay process. A typical case in point is

where a nuclide (target) is irradiated by a particle flux and, as a result of a nuclear reaction, the parent nuclide of a chain is formed.

In this case, the equation for the first nuclide in the system (1) changes to:

$$\frac{dx_1}{dt} = \hat{S}_0 - S_1 x_1, \tag{5}$$

where $\hat{S}_0$ is the rate at which the parent radionuclide of the chain is formed when the target is activated.

The following algorithm can be used to solve this: let us add an additional abstract nuclide to the existing decay chain such that it becomes the parent nuclide of the chain. Let us set the following decay conditions for the newly introduced nuclide:

$$S_0 \to 0, x_0(0) \to \infty, S_0 x_0(0) = \hat{S}_0, \tag{6}$$

where S_0 is the decay rate of the added nuclide; and x_0 is the number of nuclei of the added nuclide.

Then the system of equations describing the task with activation is similar to the system for the task in (1) and (2), and the formulas in (4) can be used to solve it. The solution is written as:

$$x_n = \hat{S}_0 S_2^1 \ldots S_n^{n-1} \sum_{i=0}^{n} \frac{\exp(-S_i t)}{\prod_{k \neq i}^{n} (S_k - S_i)}, n = 1..N \tag{7}$$

This result has singularity for stable nuclides in the chain, since in this case the product in the denominator for the two terms vanishes. (S_0=0 by definition, S_n=0, since the nuclide is stable).

Once the uncertainty has been resolved, the solution to the task for a stable nuclide in the chain is written as:

$$x_n = \hat{S}_0 S_2^1 \ldots S_n^{n-1} \left[\sum_{i=1}^{n-1} \frac{\exp(-S_i t)}{\prod_{k \neq i, k=1}^{n-1} (S_k - S_i)} + \frac{t}{\prod_{k=1}^{n-1} S_k} - \frac{\sum_{k=1 l \neq k, l=1}^{n-1} \prod^{n-1} S_l}{\prod_{k=1}^{n-1} (S_k)^2} \right] \tag{8}$$

If it is necessary to find a solution when activation has continued for a given time T_A, but then the chain has decayed without activation, the solution may be obtained from the following algorithm: up to the moment $t=T_A$ the problem is solved using activation (1, 5) and (2), and after the moment $t=T_A$ the problem is solved with (1), (without an additional nuclide), with the initial conditions obtained at the moment $t=T_A$ and for the time $\tau=t-T_A$.

It is obvious that when the target is activated under non-zero initial conditions, in (1, 5), we may obtain the solution as the sum of solutions to the task with activation and zero initial conditions and the task without activation with the given initial conditions.

From the number of nuclei calculated for a nuclide it is easy to obtain its activity by using the well-known expression:

$$A(t) = \lambda x(t), \tag{9}$$

where λ is the nuclide decay constant.

When the initial conditions for nuclides *2...N* are not zero, the solution may be obtained from the following algorithm: for each nuclide in the initial chain a decay chain is plotted (which is a part of the initial chain) and tasks (1) and (2) are solved. The results obtained are summed.

1.4. Calculating radiation energy release

If the average energies per radionuclide decay are known we can calculate the energy release *D* from radionuclide decay using the following formula:

$$D(t) = K\overline{E}\int_0^t \lambda x(\tau)d\tau, \tag{10}$$

where x is the time dependence of the number of nuclei of the radionuclide; λ is the decay constant of the radionuclide; $\overline{E}$ is the average energy, released by a single radionuclide decay; and K is the coefficient, depending on the choice of measurement units. If $\overline{E}$ is in MeV and D is in Gy, then $K = 1.602 \cdot 10^{-13}$.

Henceforth, for reasons of simplicity, we shall consider the task of calculating the energy release of ionizing radiation from the decay of one nuclide in the chain. The radiation energy release from the decay of all the nuclides in the chain can be obtained by adding the energy release of the separate nuclides.

It is obvious that an analytical expression for the radiation energy release can be obtained by integrating the expressions for $x(\tau)$. In particular, for a simple decay chain (1) with initial conditions (2), the radiation energy release from decay of the nth nuclide is:

$$D_n(t) = K\overline{E}\lambda_n\int_0^t x_n(\tau)d\tau = K\overline{E}_n\lambda_n \cdot x_{1,0}S_2^1...S_n^{n-1}\sum_{i=1}^{n}\frac{1-\exp(-S_i t)}{S_i\prod_{k=1,k\neq i}^{n}(S_k - S_i)} \tag{11}$$

The problem of obtaining the radiation energy release values for non-zero initial conditions is solved in a similar way as for the number of nuclei: the total energy release is obtained by summation of the values obtained by solving independent tasks with initial conditions (2) for each nuclide in the chain.

Target activation is a rather more complex matter. In this case, the analytical expression for energy release takes the form:

$$D_n(t) = K\overline{E}\lambda_n \int_0^t x_n(\tau)d\tau = K\overline{E}_n\lambda_n \cdot \hat{S}_0 S_2^1 ... S_n^{n-1} \sum_{i=0}^{n} \frac{1-\exp(-S_i t)}{S_i \prod_{k=0,k\neq i}^{n}(S_k - S_i)} \quad (12)$$

This formula has a singularity, – uncertainty of the form $\frac{0}{0}$.

To exclude the uncertainty it is sufficient to expand the exponent containing S_0 in series up to terms of the first order of an infinitesimal and replace S_0 with zero:

$$D_n(t) = K\overline{E}_n\lambda_n \cdot \hat{S}_0 S_2^1 ... S_n^{n-1} \left[\frac{t}{\prod_{k=1}^{n}(S_k)} + \sum_{i=1}^{n} \frac{1-\exp(-S_i t)}{S_i \prod_{k=0,k\neq i}^{n}(S_k - S_i)} \right] \quad (13)$$

The power of radiation energy release is a simpler matter:

$$\dot{D} = \frac{dD}{dt} \quad (14)$$

From formula (10) and determination of activity $A(t)$, we obtain:

$$\dot{D}(t) = K\overline{E}\lambda x(t) = K\overline{E}A(t) \quad (15)$$

Obviously, the radiation energy release may be obtained for tasks using any combination of activation and simple decay with different initial conditions by "joining together" the tasks described above.

1.5. Calculating the radiation spectrum of the chain

The radiation spectrum of the chain is the sum of the radiation spectra of its component nuclides. On that basis, let us consider the question of obtaining the spectrum of an individual nuclide in the chain at an arbitrary point in time. If we know the spectral energies of the nuclide radiation and their intensity we can obtain the spectrum by using the following formula:

$$B_k(E_k, t) = \lambda x(t) \frac{I_k(E_k)}{100}, \quad (16)$$

where E_k is the energy of the kth line of radiation (or particle); $I_k(E_k)$ is the radiation intensity of the kth line (in %); $B_k(E_k t)$ is the number of quanta (or particles) emitted with an energy of E_k at the time of t per second.

It should be pointed out that a large computer memory is needed to calculate radiation spectra because of the large volume of data on the spectral lines of nuclide radiation and the need to calculate the spectrum of complex chains with a large number of nuclides. In order to reduce the volume of information in the proposed information system, radiation lines with an intensity of less than 1% of the maximum line intensity in the given spectrum are sifted out.

2. Identifying gamma-emitting nuclides

We know that we can identify radioactive nuclides from the energy, gamma radiation intensity and half-life values. Despite the fact that modern nuclear spectrometry methods enable us to determine with a rather high level of accuracy the above-mentioned parameters for the radiation under investigation, identification of the nuclides (especially for complex mixtures) is a very laborious task. The main difficulty lies in interpreting the experimental results with respect to known nuclear decay characteristics which, in the available reference literature, are dispersed, firstly, over the nuclides (more than two thousand nuclides) and, secondly, over the gamma-transition energies (the known number of gamma-lines exceeds 60 000).

The proposed computerized information system has the following algorithm for identifying gamma-emitting nuclides. All the radionuclides are broken down into 13 groups according to their half-lives. In each half-life range the gamma lines of the radionuclides present are broken down into energy groups. Having selected the half-life range of interest and, subsequently, the gamma-radiation energy range, the user of the computerized system obtains a table of energies and intensities of the selected gamma-lines with an indication of where every line belongs. The data given in the table may be represented graphically. Moreover, the data in the table can be used to identify radiation lines which exceed the given intensity threshold and/or to select radiation lines in a narrower energy and half-life range.

Conclusion

The IIS is intended for use by a wide range of specialists at different levels (students, post-graduate students, engineers and scientists), who need reliable information (as of 1999) on stable and radioactive nuclides. The main difference between the IIS described above and existing charts of the nuclides, reference books and computerized systems is that the evaluated databases of nuclear physics constants are linked with a set of programs enabling analysis of a wide range of problems in various nuclear physics applications. This work is supported by a grant from the Russian Fund for Fundamental Research (project No. 00-07-90376).

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