ORIP_XXI COMPUTER PROGRAMS FOR ISOTOPE TRANSMUTATION SIMULATIONS

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ABSTRACT

ORIP_XXI software suite is developed for the study of radioactive and stable isotope transmutation chains, i.e. nuclide networks with possible feedbacks. Using the programs from this suite it is possible to estimate various quantitative characteristics of a nuclide transmutation chain, both undergoing irradiation in neutron fluxes and in case of spontaneous radioactive decays. The main programs of *ORIP_XXI* software are: the electronic nuclide chart *NKE*, the program for finding chains of nuclear transmutations *ChainFinder*, and the program for full simulation of nuclide production in a nuclear reactor *ChainSolver*. Only the open-source subprograms and licensed software products were used in the coding. *NKE*, *ChainFinder*, and *ChainSolver* codes are accessible via NEA OECD Computer Program Service. All programs use a common data file, which contains nuclear constants and decay data for more than 2800 nuclides with atomic weights from 1 up to 293 (nuclear charge from 1 up to 118) and characteristics of chemical elements. The file also includes data on fission product yields for thermal and fast neutron induced fission of 22 heavy isotopes. *ChainSolver* allows users to take into account neutron flux depression and self-shielding factors. Self-shielding factor calculations in the intermediate resonance approximation are based on additional data from the resolved resonance parameters file. All data are obtained from freely available public nuclear data libraries. The data may be edited as necessary for carrying out isotope concentration calculations. After calculations are finished, various characteristics are calculated based on the estimated isotope densities: isotope masses, element masses, isotope activities, isotope parts, mass yields, depression coefficients, self-shielding factors, and fission energy deposition estimates at all time steps.

KEYWORDS: transmutation, neutron, fission, nuclide, isotope

1. INTRODUCTION

The theory and application of nuclear transmutations is of considerable interest in a number of scientific fields. Radioactive isotopes are now widely used in medicine, industries, general and applied sciences. Main products of "State Scientific Centre of Russia - Research Institute of Atomic Reactors" include radionuclides produced in research nuclear reactors and radiation sources using radioactive isotopes. The research reactors, which have very high neutron fluxes as well as the ability to vary neutron spectra in a wide range, are unique tools for producing highly

radioactive nuclides. Currently available non-commercial software for nuclear transmutation prediction is outdated and does not support modification: neither of the source code nor of the physical data used for transmutation simulations. *ORIP_XXI* program complex [1] allows to analyze, plan and predict successfully radionuclide production by irradiation in a high-flux nuclear reactor, as well as to carry out calculations necessary for storage, transportation, and recycling of radionuclides (in the case of zero neutron flux). The programs' simplicity, robustness, and scalability are due to the use of object-oriented programming. Though programs include extensive data set, users can edit the data necessary for the calculations. The programs can be used to specify widely used isotope production modes in greater detail, to choose new promising (e.g. for a medical application) nuclides, and to study opportunities for new isotope production in nuclear reactors. The programs can also be used to determine the sources of the inevitable impurities in the irradiated material. On the basis of this information it is possible to optimize (for example, to minimize impurities) production process, choosing the irradiation mode, schedule and the initial isotope structure. The case of no neutron flux is vitally important when investigating storage, processing, transportation, recycling of fissionable and radioactive materials, and transmutation of long-lived waste including the irradiated nuclear fuel.

2. PROGRAMS OF THE COMPLEX

ORIP_XXI program complex includes the following components: the electronic nuclide chart *NKE*; the nuclide chain search program *ChainFinder*; the transmutation simulation program *ChainSolver*; the data file with characteristics of decays, emitted radiations, cross sections of neutron reactions, decay channel percentages, neutron induced fission yields; the data file with resonance parameters; and service utilities. The main programs, their use, tasks, resources, and features are described below in detail.

2.1. Electronic Nuclide Chart *NKE*

NKE program is developed to visually represent basic nuclear physical data (half-lives, neutron reaction cross sections, radioactive decay data, fission yields, etc.) and to search for the nuclides satisfying some criteria. Only freely available information sources were used in creation of the data file: libraries of nuclear-physical constants and available publications [2-8] (see also the *NKE* help file). The main program screen (Fig.1) is an electronic nuclide chart ('Karlsruher Nuklidkarte'). The friendly user interface allows to specify exactly what information should be displayed on the nuclide chart. It is possible to move to any nuclide quickly. On a double-click in a nuclide cell the general information about the nuclide is shown. The user also can request the detailed information for any isomeric state. The displayed data include the information about neutron reactions for chemical elements; properties of the stable/ground/metastable nuclide states (the half-life, decay channel percentages); the information about $\alpha-$, $\beta-$, $\gamma-$ decay particles, emitted electrons and positrons, the estimate of ionizing gamma-constant $K\gamma$, in units of $R \cdot cm^2 \cdot hour^{-1} \cdot mCi^{-1}$ ([9]); yields of fission products for various fissionable nuclei in fast and thermal neutron spectra; thermal cross-sections and resonance integrals (capture and fission); cross-sections of (n,p), (n,α), (n,2n), (n,n'), and (n,γ) threshold reactions averaged on a fission neutron spectrum. The latest program versions enable the user to load data from another file as

well. The default data file is ORIP_XXI.oob, the new one is TORI_22.oob. It contains information on radiation from [10] (neutron reaction cross-sections and fission yields are the same as in the default data file).

Figure 1. *NKE* main window (with the legend).

The program can perform complex searches, which allows for the analysis of all the data available in *ORIP_XXI* software suite data file. The dialogue shown below (Fig.2) allows users to define the search criteria. It is possible to combine several filter conditions by either 'AND' or 'OR' operators. See Fig.2 for a filter example and the corresponding output. The shown filter finds isotopes that are sources of β-particles but the radioactive decay of which is not accompanied by gamma radiation. Such nuclides appear to be promising for medical applications.
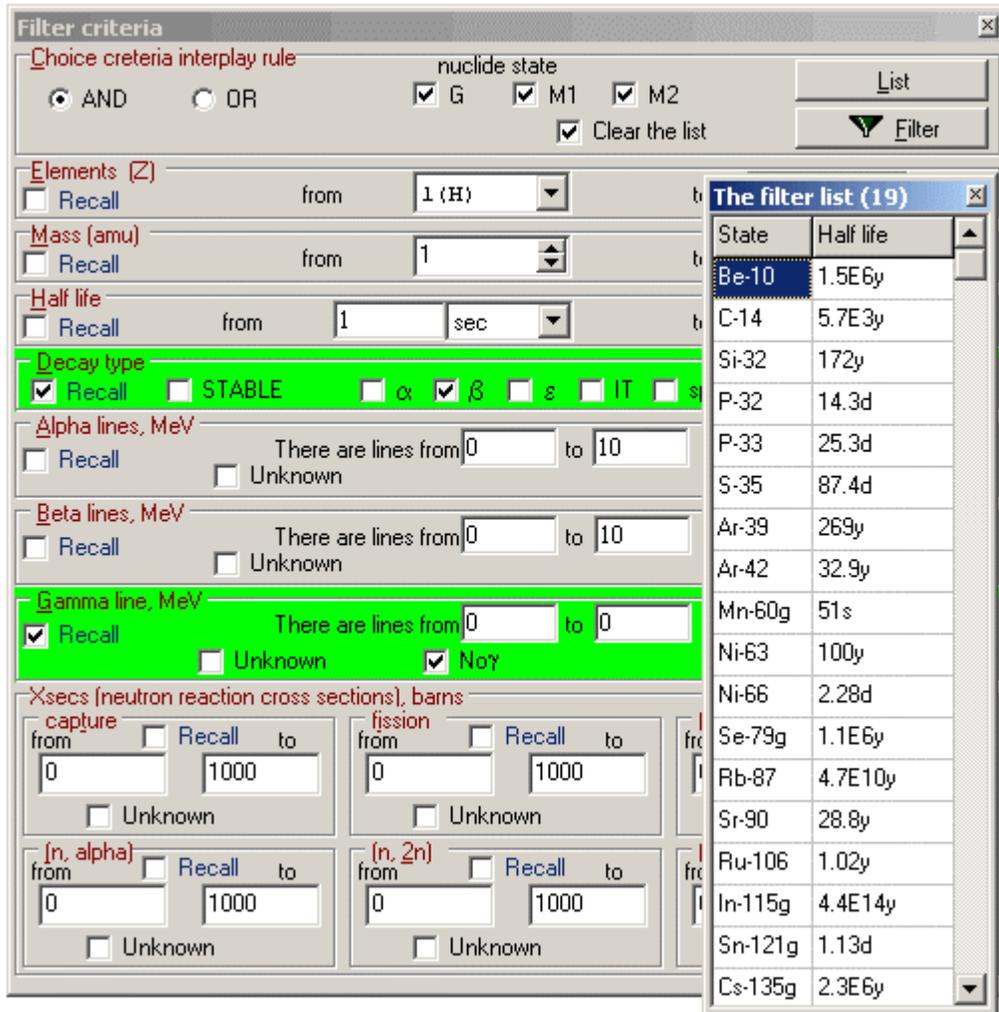
Figure 2. The search dialog of *NKE* program (and the result list).

2.2. Chain Search Program *ChainFinder*

The main function of *ChainFinder* program (Fig.3) is searching for nuclide chains which may be realized in nuclear reactors. Though the primary production chains are well known, sometimes it may be necessary to obtain new nuclides with unknown production networks. The program also allows to find out the reason for appearance of unexpected isotopes in irradiated material. *ChainFinder* automatically builds possible transmutation chains, given the start and finish states. The chain search process takes data on decay "subbranchings" available in the data file into account. For example, β-decay of ^{95}Zr nuclide produces the descendant ^{95}Nb isotope in both ground and metastable states. Thus, the data used includes all the data in the data file and not only those displayed by *NKE*. *ChainFinder* allows the user either to take into account or not, as desired, decay reactions, thermal neutron captures, neutron induced fission, and fast neutrons threshold reactions. This capability is useful, e.g., when trying to find out what transmutations are possible during storage and transportation of non-fissionable nuclides, when only decay

reactions can happen. In cases where fissionable isotopes are absent, excluding fission reactions from search significantly reduces chain search time. Similarly, if fast neutrons reactions are negligible (for example, in a thermal reactor), excluding threshold reactions simplifies the analysis.

The pop-up menu allows to transfer the list of state names in the chain(s) into the Windows clipboard, or to save a chain into a file. A saved chain may be loaded later into *ChainSolver* program for carrying out nuclide production calculations. An interesting example of the program application: Consider the need to find out how ^{234}U is accumulated in nuclear reactor fuel. For this you would use ^{235}U or ^{238}U as the starting state. During the program testing it turned out that *ChainFinder* finds a lot of non-trivial networks. Some of these have no practical interest because they have very low probabilities. But at the moment the program shows all the found chains because transmutation rates can change drastically during irradiation depending on conditions and no acceptable "price" function has yet been found.
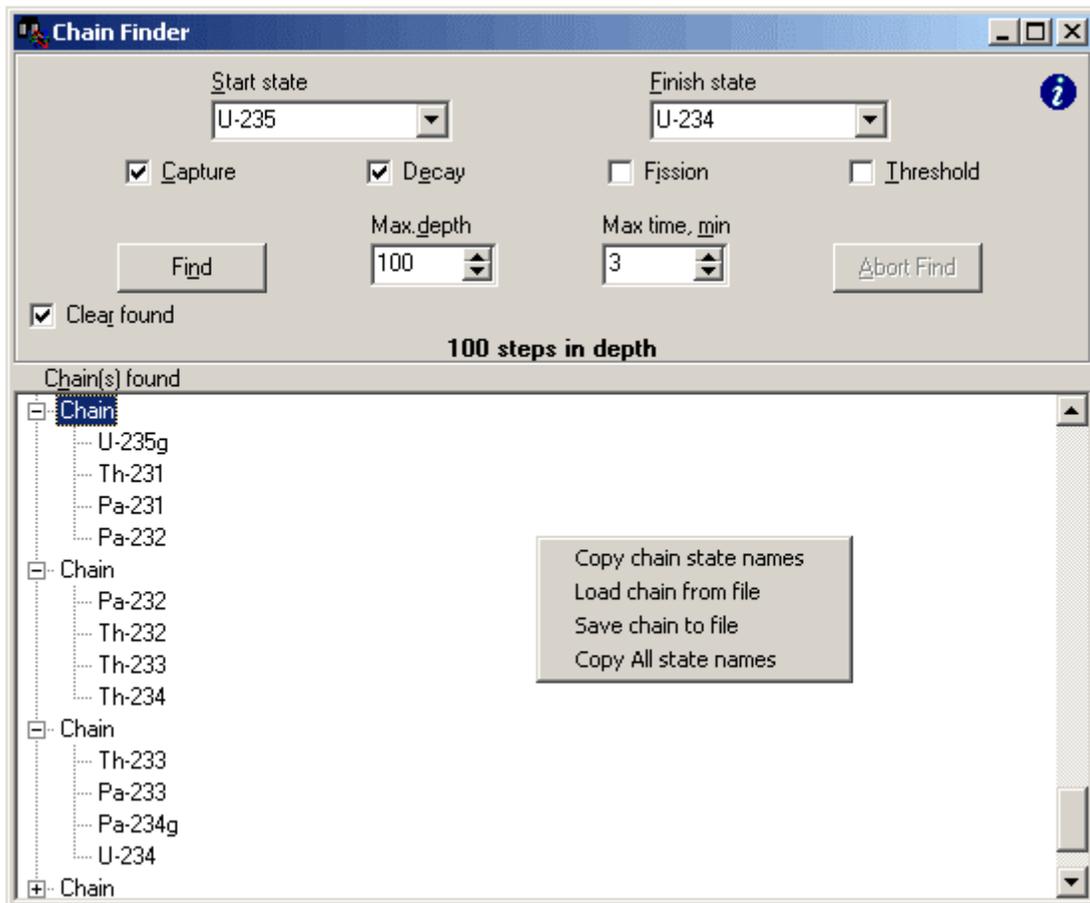
Figure 3. *ChainFinder* **window (with pop-up menu).**

2.3. Transmutation Simulation Program *ChainSolver*

ChainSolver program is developed for numerical simulation of nuclear transmutation at the final stage before the irradiation or before shipping isotopes. The neutron capture resonance self-shielding, thermal neutron flux depression and the schedule of the irradiation (the reactor schedule and rearrangement of the irradiated target in various positions in one or different reactors) can be taken into account. Doing these calculations is extremely tedious since at each irradiation time step the specific neutron fluxes, reaction cross-sections, and the initial isotopic composition all have to be taken into consideration. The calculations allow to choose the irradiation mode more precisely and to estimate the output of the products as well as the inevitable impurities. The main approximation used in transmutation simulations is the following: changes of the neutron spectra specified by the user due to the changes of the composition of the irradiated material are assumed to be negligible.

The program solves the following ordinary differential equations system describing a network of isotope states undergoing both spontaneous decay and neutron induced nuclear transmutations:

$$\frac{dN_i}{dt} = -N_i \cdot \mu_i + \sum_{j \neq i} k_{ji} \cdot N_j$$

N_i - nuclear density of i-th isotope state, μ_i- constant of i-th state withdrawal: $\mu_i = \lambda_i + \sum \sigma_g \Phi_g k_g$, where λ_i - the half-life, σ_g - the burnout cross-section, Φ_g – neutron flux, k_g – self-shielding/depression blocking factors, g – neutron energy group identifier (g=1..3), $\sum$ - sum over energy groups; k_{ji} - constant of transformation from j-th isotope to i-th (taking into account spontaneous transformations and neutron reactions, similar to the formula for μ_i);
i (j) - unique isotope state identifier in the network.

The program interface uses a Delphi visual component, which enables to display several pages on one main form. Each page contains a group of parameters, which are necessary for numerical evaluation of nuclear transmutations and derived quantities. The 'Chain' tab of *ChainSolver* main window (Fig.4) allows the user to create a transmutation chain and to edit it. Editing here means adding states and/or links and changing their numerical characteristics. A link is an arrow corresponding to the transformation between two isotope states, possibly two states of the same nuclide. There are some numerical values associated with the link, which define the transformation rates. A nuclide network to be simulated can be created in several ways. The chain can be loaded from a file created by *ChainFinder*. The chain also can be created by starting with an empty chain and adding nuclide states. To add a nuclide state type its name (e.g. U-235 or U-235g, U-235m or U-235m1, Hf-177m2) and press the 'Add' button. If the 'Build Links' checkbox is checked, adding a state automatically adds the links between the states that are already in the chain and the new one. The types of links created are controlled by the other checkboxes: 'Decay' – spontaneous radioactive decays, 'Capture' - captures of thermal and resonance neutrons, 'Fission' – neutron induced fissions, 'Threshold' - threshold neutron reactions for fast neutrons. The isotope production due to spontaneous fission is considered negligible and so the corresponding links are not created. If spontaneous fission must be taken into account, the

necessary links may be added manually (or the links for neutron induced fission transformations between these nuclide states may be edited, if those are in the chain already). Any links between isotopes are constructed on the basis of the information available in the data file. The bottom part of the 'Chain' tab shows the detailed information on the selected link or state and can be edited. For example, if nuclides ^{14}N and ^{14}C are added to the network, the link corresponding to the fast neutron (n, p) transmutation between them is automatically created. However, the information on the same reaction induced by thermal neutrons is absent from the data file, so it has to be added manually. For this purpose it is enough to add values of the thermal cross-section and the resonance integral of the reaction to the corresponding rows of the table. The latest program versions create this link automatically, as well as some other links for known (n, p) and (n, α) reactions.
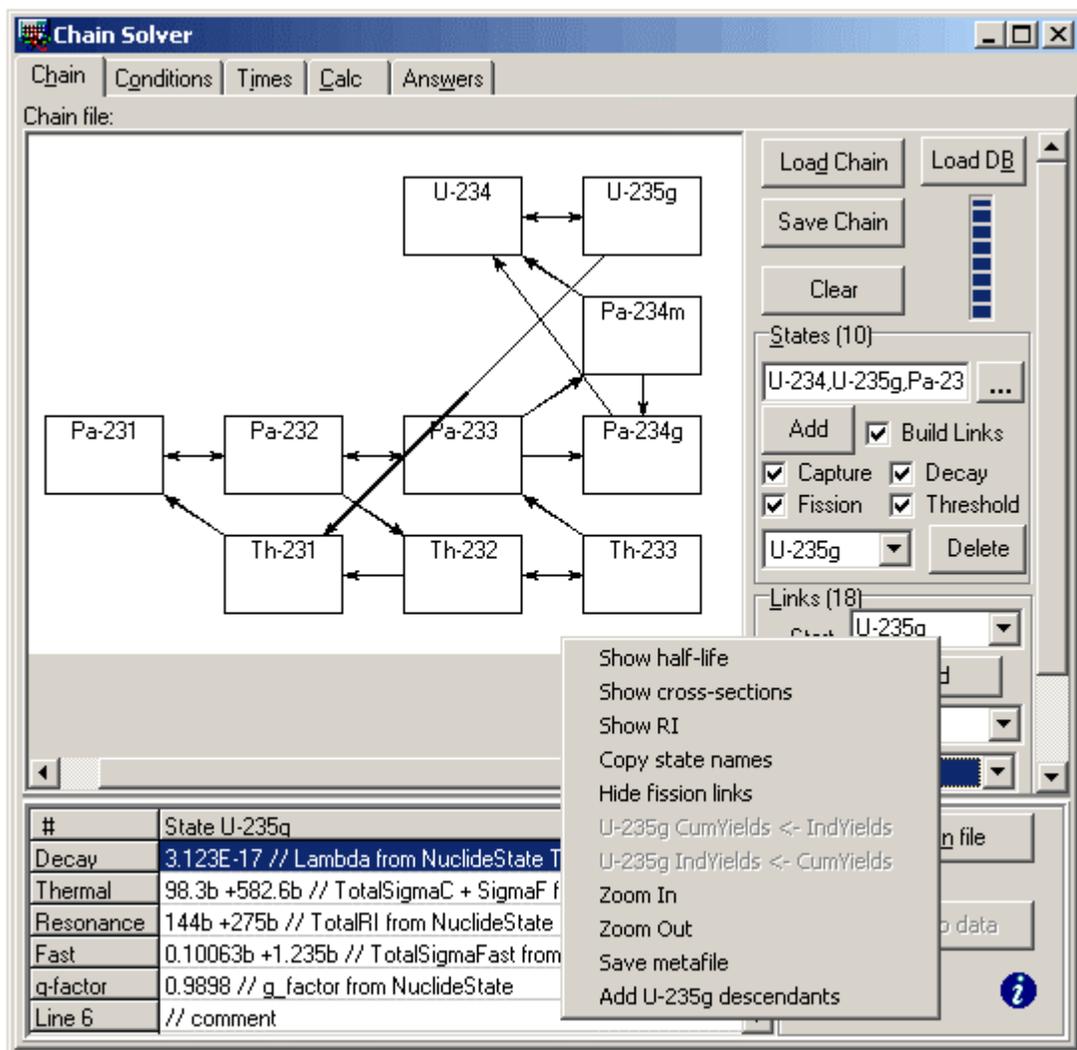

Figure 4. 'Chain' tab of *ChainSolver* program.

'Conditions' tab (Fig.5) is where the conditions of irradiation are set: the initial composition, required accuracy, and the data on thermal flux depression and resonance self-shielding. Initial mass parts of the nuclide states, weights of elements, a lump of a sample (for specific mass yield calculations), and the absolute and relative tolerances (*atol* and *rtol* respectively). The local tolerance for i-th nuclide concentration y(i) on each time step is determined by the formula $TOL(i) = rtol(i) \cdot abs(y(i)) + atol(i)$. The accuracy requirements are assumed to be satisfied when the difference between two consecutive approximations does not exceed the local tolerance. Calculations taking into account resonance self-shielding (at the moment using intermediate resonance approximation [11]) are extremely tedious and need a lot of additional parameters. Since resonance self-shielding factors calculations are rather long, the factor table may be saved to a disk file in order to load and use it in the program later. To calculate the table the user sets the resonance absorber lattice type ('No' - a single block in an environment of external moderator, 'Sq' - a rectangular lattice, 'Hex' - a hexagonal prism lattice, 'Simple' - a single block in indefinite external moderator). The block volume, outer moderator parameters, and mixed moderator parameters (nuclear weight, neutron scattering cross-section and nuclear density) are set. "Mixed moderator" is usually the target matrix material.
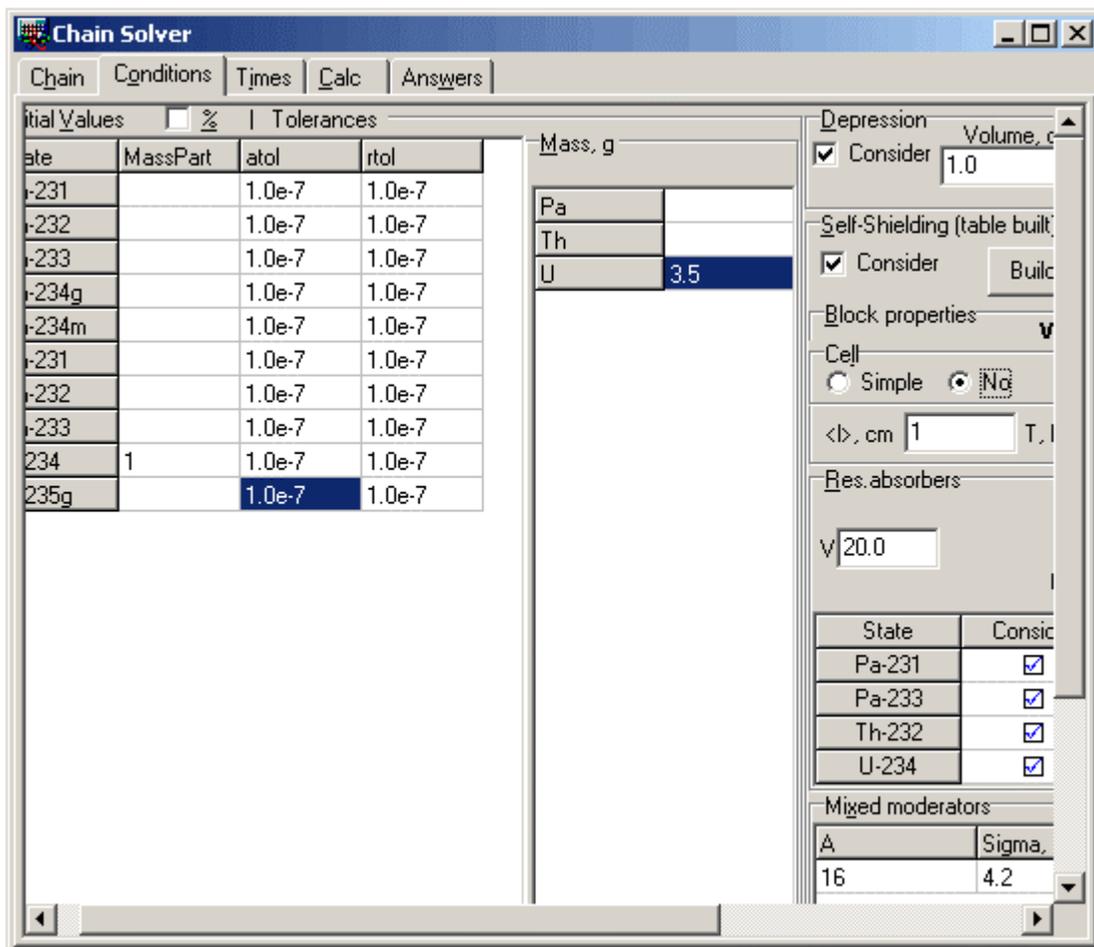

Figure 5. 'Conditions' page of *ChainSolver* program.

The 'Times' page (Fig.6) is where the user sets the irradiation schedule. In the multi-line editor paired values are set: time interval (in days) and the corresponding power level. In one-line editors the following values are assigned: 'T n.g' - neutron gas temperature (in Kelvin degrees) for the account of neutron reaction cross-section dependences from the thermal neutron spectrum characteristics; 'Th', 'Res', 'Fast' - the flux of thermal, resonance and fast neutrons respectively for nominal reactor power. 'Nominal' is the nominal power level. During calculations the neutron flux for a time interval is defined as a product of flux value for nominal power and the ratio of the actual power level (entered for the time interval) to the nominal. The program allows to simulate multi-stage irradiation processes when the irradiated material is placed in different positions in the reactor at different stages, or even in different reactors and/or endurance pools with different neutron spectra.
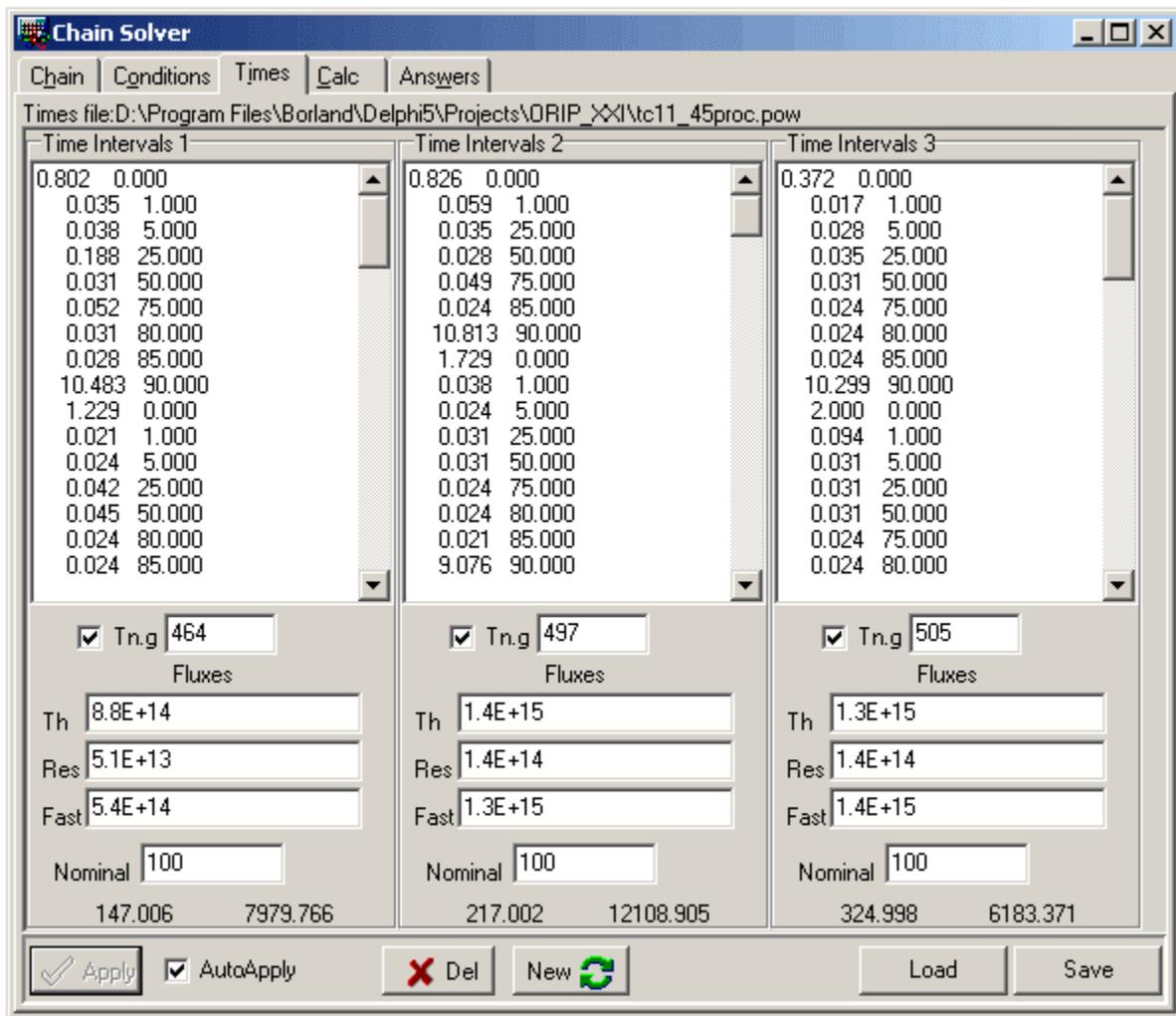

Figure 6. 'Times' page of _ChainSolver_ program.

The 'Calc' page gives the interface to various solution methods for the system of ordinary differential equations (ODE) arising from the transmutation network. The ODE coefficients depend on time, both because neutron flux depends on the irradiation schedule and because changes of nuclear densities result in changes of thermal flux depression and resonance self-shielding factors. Four ODE solving subroutines - VODE, LSODA, RADAU, and MEBDF are available. All subroutines can be called from dynamically loaded libraries (DLL). Communication between DLLs and the main program responsible for the user interface and realization of objects is accomplished by means of disk files. For the program to be functional on systems where disk storage is absent or the user has no write access, LSODA and VODE subroutines were source-converted and coded in Delphi Pascal programming language. Two Jacobian calculation methods are used: the matrix can be calculated in the main program or Jacobian can be generated internally in the ODE solving subroutine from results of intermediate calculations. It is well known that there is no single ODE system numerical solution method, which is "excellent" for any system of equations (see e.g. [12]). VODE [13,14] uses variable-coefficient Adams-Moulton and backward differentiation formulas (BDF) methods in Nordsieck form. A change in step size and/or order that is decided upon at the end of one successful step is not implemented until the start of the next step, so that interpolations performed between steps use the more correct data. LSODA (Livermore Solver for ODE, Automatic, by Linda Petzold and Alan Hindmarsh) uses automatic method switching for stiff and non-stiff problems. It uses Adams methods (predictor-corrector) in the non-stiff case, and BDF methods (the Gear methods) in the stiff case. The user does not have to determine whether the problem is stiff or not, and the solver will automatically choose the appropriate method. LSODA always starts with the non-stiff method. RADAU [15] uses implicit Runge-Kutta method of order 5 for problems of the form My'=f(x,y) with possibly singular matrix M with dense output (collocation solution). MEBDF subroutine [16] uses modified extended backward differentiation formulas of moderate to high order with better stability. Stability is not the only issue, however: the formulas also are computationally efficient. The subroutines for solving the ODE systems have passed series of tests, both common ones for initial values problems [17] as well as specific tests for simple transmutation networks by means of the special utility for linear transmutation chains having an analytical solution [18]. The typical calculation time doesn't exceed several minutes for practical problems.

After calculations are finished, the 'Answers' tab shows various characteristics obtained from calculated isotope densities: isotope masses, element masses, activities (see below), isotope specific activities per element gram, isotope parts, mass yields, depression coefficients, self-shielding factors, and fission energy deposition estimates for all time steps. Displayed activities are: α- is alpha decay; β- is beta-minus decay; EC – electron capture plus beta-plus decay; and IT - isomeric transition, typically to the ground state. For energy deposition calculations it is assumed that only neutron-induced fission contributes to energy deposition significantly, at 200 MeV per fission (spontaneous decays and transport of particles are ignored). The energy deposition estimate is essential when irradiating weight quantities of actinides and trans-uranium elements, because heating due to fissions of starting or intermediate isotopes can lead to significant difficulties. The program allows users to choose activity units to be used (Ci or Bq). The results can be saved to a Microsoft Excel spreadsheet file.

3. CONCLUSIONS

Since year 2000 *ORIP_XXI* freeware programs have been successfully used by Division of Radionuclide Sources and Preparations of State Scientific Centre of Russia - Research Institute of Atomic Reactors to visualize and analyze data on neutron properties of nuclides and elements, to automatically construct transmutation networks, and to calculate the production of any member of a network in the process of spontaneous decay and/or induced neutron transformation in a nuclear reactor. *ChainSolver* has been extensively tested against the analytical and experimental results for a number of problems. Some of interesting problems which were solved with the programs are production of super heavy isotopes of einsteinium and fermium, investigations in opportunities of ruthenium radioactive isotope production as fission products from trans-uranium targets, fission product yields estimates in irradiated fuel from viewpoint of safety, where transmutation network includes near hundred isotope states. *ORIP_XXI* includes *NKE*, the electronic nuclide chart, *ChainFinder*, the program for finding transmutation chains, and *ChainSolver*, the program for simulating these chains. *NKE* program allows the user to obtain detailed information on radioactive and neutron-physical properties of isotopes and elements as well as to produce nuclide sampling based on the analysis of the entire data set on all isotopes. *ChainFinder* program enables the user to automatically construct transmutation chains for any given start/finish isotope pair. The program uses a subroutine of search for links which allows the user to either take into account or not, as desired, separate transmutation channels: spontaneous radioactive decay, thermal and resonance neutron capture, neutron induced fission, and threshold reactions due to fast neutrons. Finally, *ChainSolver* program for isotope transmutation simulations allows users to create and to edit an isotope transmutation network. During network construction nuclide transformation rates (including neutron induced fissions) are initialized by values from the data file and may be edited. Users can add both nuclide states and links between them, and change numerical values to be used in the subsequent calculation. *ChainSolver* also allows to set initial conditions and reactor schedule. The program can take spatial dependence of neutron reaction rates, namely thermal flux depression and resonance self-shielding factors, into account. On the basis of the calculated nuclear densities, the program figures out various characteristics of the irradiated material: isotope weights, specific outputs, activities, factors of self-shielding and depression, energy deposition caused by neutron-induced fission. Numerical subroutines for the transmutation system solving were coded as dynamically loaded libraries (DLL) with GNU Fortran compiler, due to the fact that there is available professionally written and well-tested mathematical software in Fortran language for solving ordinary differential equation systems. Communication between DLL and the main program responsible for the user interface and realization of objects is accomplished by means of disk files. For programs to be functional on systems where disk storage is absent or the user has no write access two solving subroutines were coded in Delphi Pascal programming language. Thus, only open-source subroutines, licensed software products, and published data were used. The *ORIP_XXI* freeware enables to carry out calculations on materials activation by neutron radiation, to estimate the induced activity, and to determine changes of chemical and isotope structure. This allows to predict changes in material properties as a result of an irradiation and/or storage. The programs are useful in optimization of radionuclide production, nuclear physics experiments, general and applied sciences, and industries.

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