COMPUTER SIMULATIONS OF THE STAINLESS STEEL COMPOSITION CHANGES INDUCED BY NEUTRON BASED NUCLEAR REACTIONS

Y.V. Rudychev, M.A. Khazhmuradov *, S.I. Prokhorets, D.V. Fedorchenko

National Science Center "Kharkov Institute of Physics and Technology", 61108, Kharkov, Ukraine

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Problem of construction material composition changes induced by neutron fluxes of nuclear reactor VVER-1000 as a result of a nuclear reaction and associated activation processes was considered. The special methodic to simulate this processes using computer simulation methods was developed. Stainless steel H18N10T composition changes during a long irradiation time was simulated.

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1. INTRODUCTION

The life time extension of operational nuclear power plains is a critical problem of Ukrainian energy industries. Solution of this problem is significant low cost operation as compared with new nuclear power plaint production. This is especially important because of limited building power of a new VVER-1000 power unit in Russian Federation. Physical statement and support of life time extension of nuclear power plains is elaborate and complex problem. This required participation of many specialists from different fields of science. One of the possible ways of solution this problem is an investigation of stainless steel X18H10T as a constructional material of reactor vessel and in-vessel component[1]. Within the bounds of this one of the possible methods is making of imitational experiments. These experiments are irradiations of given constructional materials by different kind and energy ions in goal to simulate the same conditions for radiation induced composition changes as under irradiation by nuclear reactor. Unfortunately, because of different nature of neutron and ion irradiation, the misfit between results of imitational experiments and reactor irradiations is observed.

In a given work, using special simulation methods, the problem of the stainless steel composition changes induced by neutron fluxes of nuclear reactor VVER-1000 as a result of a nuclear reaction and associated activation processes is considered. It is necessary to note that during imitational experiments impossible to taking into account such processes because of no neutron fluxes exist in this case. As in our case we have long time neutron irradiation. Therefore the significant initial compositions changes are possible by activation processes of transmutation in given material. Production of hydrogen and helium by $^{56}Fe(n,p)^{56}Mn \rightarrow p + e^- \rightarrow {}^1H$ and $^{56}Fe(n,\alpha)^{53}Cr \rightarrow \alpha + 2e^- \rightarrow {}^4He$ reactions plays significant role in initial composition changes.

Production of given gases combined with radiation damages effects may lead to significant material properties changes.

2. SIMULATION TECHNIQUES

General principles of solution of given tasks are calculation of concentration of the given elements depending on irradiation time. Isotope transmutation and activation processes physically grounded on nuclear reactions induced by external elementary particle source or secondary particles of decayed isotopes. Examples of such reactions are $(\gamma,n),(\gamma,p),(p,n),(n,\alpha)$ and other ones.

In present moment the most of well-known methods of simulations of activation and transmutation processes based on solution of differential equations of concentration changes. Main trouble in given methods consist of define of reactions cross-sections and define of fluxes and energy spectrums of the given particles with was induced by this reactions. Most of modern's objects of investigations are complicated 3D systems with complex heterogeneous structures such as devices, constructive elements and other. Definition of fluxes and spectra of radiation transport problems was initially attempted on the basis of the Boltzmann transport equation. However, this procedure comes up against considerable difficulties when applied to limited geometries as written above, with the result that numerical methods based on the transport equation have only had certain success in simple geometries, mainly for unlimited and semi-infinite media. Hence for this case the special complex methodic was developed. Conceptual scheme of this methodic is represented on Fig. 1.

*Corresponding author. E-mail address: khazhm@kipt.kharkov.ua

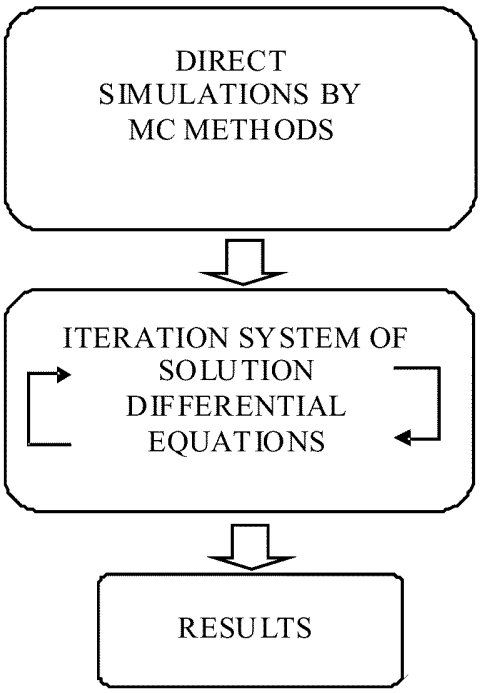

Fig.1. *Conceptual scheme of developed simulation methodic*

In this methodic all radiation transport characteristics were defined directly by Monte Carlo method. After that obtained results were used as a data for applying in iteration system of solution of differential equation set.

As tools for realization of given scheme the several special codes were used. As Monte Carlo simulation tools the widely-used particle transport codes GEANT4 and MCNPX were chosen[2],[3].

These codes provide adequate description of all necessary physical processes and have possibility of implementation of 3D geometry of investigation object with very complicate structure. All results obtained from MC codes prepared by special procedure for using them as initial data for solution of corresponding time-evolving differential equations.

For this task the special code FISPACT was chosen[4]. FISPACT uses external libraries of reaction cross sections and decay data for all relevant nuclides to calculate an inventory of nuclides produced as a result of the irradiation of a starting material with a flux of neutrons. The actual output quantities include the amount (number of atoms and grams), the activity (Bq),α,β and γ-energies (kW),γ - dose-rate (Sv h^{-1}), the potential ingestion and inhalation doses (Sv), the legal transport limit (A2 value), the clearance index and the half-life for each nuclide. Amounts and heat outputs are also given for the elements and the γ-ray spectrum for the material is listed as well as various summed quantities, such as total activity and total dose-rate. At the end of each time interval the dominant nuclides (in terms of activity, heat, dose-rate, potential biological hazards and clearance index) and the pathway data for the production of these nuclides can be shown. The uncertainties in eight total radiological quantities can be calculated and output. As options, data files can be produced for subsequent use by other programs to plot graphs of the total responses as functions of elapsed time and selected blocks of output may be written to external data files.

The core task of FISPACT is the solution of a set of differential equations that describe the amounts of atoms of various nuclides present following the irradiation of a given material in a neutron field. The set of differential equations is given in equation (1).

$$\frac{dN_i}{dt} = -N_i(\lambda_i + \sigma_i\phi) + \sum_{i\neq j} N_j(\lambda_{ij} + \sigma_{ij}\phi) + S_i\,,$$
$$S_i = \sum_k N_k \sigma_k^f \phi Y_{ik}\,, \qquad (1)$$

where N_i is the amount of nuclide i at time t; λ_i is the decay constant of nuclide i $[s^{-1}]$; λ_{ij} is the decay constant of nuclide j producing i $[s^{-1}]$; σ_i is the total cross section for reactions on i $[cm^{-2}]$; ϕ is the neutron flux $[n\ cm^2\ s^{-1}]$; σ_{ij} is the reaction cross section for reactions on j producing i $[cm^2]$; σ_k^f is the fission cross section for reactions on actinide k $[cm^2]$;Y_{ik} is the yield of nuclide i from the fission of nuclide k. S_i is the source of nuclide i from fission. The final term is only required if actinides are included in the initial material.

It is necessary to use an efficient method of solution of the set of equations in (1). Since the total number of nuclides considered is over 1900. The method used in FISPACT is that of Sidell[5]. This method is an extension of the Euler (first order Taylor series) which uses an exponential function of the step length. Equation (2) shows the standard Euler solution and (3) the Sidell solution for the step time h.

$$N_i(h+t) = N_i(t) + h\left.\frac{dN_i}{dt}\right|_t\,, \qquad (2)$$

$$N_i(h+t) = N_i(t) + h\,\frac{(e^{\Lambda_i h}-1)}{\Lambda_i}\left.\frac{dN_i}{dt}\right|_t\,. \qquad (3)$$

The error in using (3) is lower than (2), but for stability of the solution it is still necessary that the time step be related to the reciprocal of the largest eigenvalue. For this reason a restriction is placed on the largest eigenvalue considered (some nuclides are considered in equilibrium). The number of steps in the computational solution of the Sidell method is greater than the Euler method, but not sufficiently to outweigh the advantages. The procedure is to split the irradiation time into two steps, perform the calculation, test the convergence of all the nuclides and, if the test fails then repeat with double the number of time steps. This procedure is continued until sufficient accuracy is achieved. The results at each stage are corrected using the results from the previous stage ('extrapolation') to improve the convergence of the solution.

Experience with this solution method in FISPACT shows that it is rapid to converge and able to give sufficient accuracy. The code implements a maximum number of iteration stages is 10, but if convergence has not been achieved by then it is usually only for a very few 'unimportant' nuclides. The output flags these nuclides, thus enabling the worth of the particular 'non-converged' run to be judged. It was mentioned above that there is a limit on the largest eigenvalue considered in the solution of the equations. This means that physically only nuclides with sufficiently long half-lives are calculated by the above method. The remainder is assumed to be in equilibrium, and thus their values can be written down immediately as shown in equation (4)

$$N_i^{equil} = (\lambda_i + \sigma_i\phi)^{-1} \sum_j N_j(\lambda_{ij} + \sigma_{ij}\phi)\,. \qquad (4)$$

The half-life at which nuclides are considered to be in equilibrium is under the control of the user. This is done by choosing the time interval (code word TIME) and the parameters following the LEVEL code word. FISPACT requires connection to several data libraries before it can be used to calculate inventories. While any libraries in the correct format could be used, the development of FISPACT over the last few years has run in parallel with the development of the European Activation File and this EAF library is the recommended source of cross section data. Therefore in our scheme EAF library was used.

3.THE SIMULATION OF ACTIVATION OF STAINLESS STEEL H18N10T

For first approximation of a given task we decided to simplify scheme of simulation because of complete cycle of computation applying to VVER-1000 reactors by Monte Carlo methods spend a lot computer time. In other hand fluxes and energies have no significant differences compare to averaged values of neutron spectra. Therefore the averaged spectrum for VVER-1000 reactors type was used. The special 69 group's representation of this spectrum called WIMS was used. This method of presentation makes it clear in which energy ranges particular structures have most groups and will therefore give a good representation of the cross sections. The average burn up was 1 MW/kg and an average neutron flux was $2.6693{\times}10^{14}$ n/cm^2 per second. This spectrum is shown on Fig. 2.

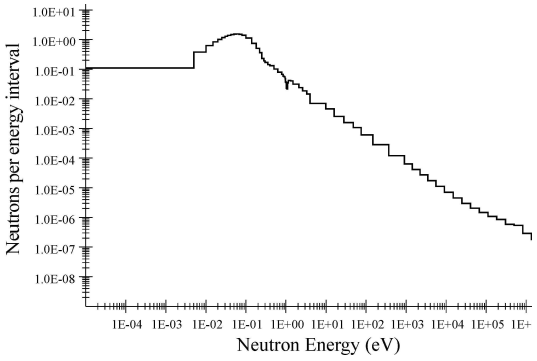

Fig.2. *Neutron spectrum during stainless steel irradiation*

Composition of H18H10T steel was chosen without taking into account a doping material.Initial composition of given steel normalized by 1 kg is shown in Table 1.

Table 1. *Composition of initial stage of H18N10T stainless steel*

Isotope	Number of atoms	Mass, gram
C12	4.96E+22	9.88E-01
C13	5.57E+20	1.20E-02
Ti46	1.04E+22	7.92E-01
Ti47	9.36E+21	7.30E-01
Ti48	9.27E+22	7.38E+00
Ti49	6.81E+21	5.53E-01
Ti50	6.52E+21	5.41E-01
Cr50	9.06E+22	7.51E+00
Cr52	1.75E+24	1.51E+02
Cr53	1.98E+23	1.74E+01
Cr54	4.93E+22	4.42E+00
Fe54	4.47E+23	4.00E+01
Fe56	7.02E+24	6.52E+02
Fe57	1.62E+23	1.53E+01
Fe58	2.16E+22	2.07E+00
Ni58	6.98E+23	6.72E+01
Ni60	2.69E+23	2.68E+01
Ni61	1.17E+22	1.18E+00
Ni62	3.73E+22	3.83E+00
Ni64	9.50E+21	1.01E+00

Simulation of activation processes was done according written above scheme for 1, 5, 10, 20 and 30 years of irradiation time. Results of this simulation for produced isotopes amount more then 10^8 atoms in H18N10T stainless steel after 30 years irradiation time is shown in Table 2.

Table 2. *Isotopes composition in H18N10T stainless steel after 30 years irradiation time*

Isotope	Number of atoms	Mass, gm	Activity, Bq	Half-life, s	Isotope	Number of atoms	Mass,gm	Activity, Bq	Half-life, s
H1	1.26E+22	2.10E-02	-	Stable	Cr50	6.83E+22	5.67E+00	-	Stable
H2	3.60E+19	1.20E-04	-	Stable	Cr51	6.92E+19	5.86E-03	2.01E+13	2.39E+06
H3	6.59E+15	3.30E-08	1.17E+07	3.89E+08	Cr52	1.72E+24	1.49E+02	-	Stable
He3	1.21E+14	6.05E-10	-	Stable	Cr53	1.66E+23	1.46E+01	-	Stable
He4	4.92E+21	3.27E-02	-	Stable	Cr54	1.09E+23	9.77E+00	-	Stable
Li6	8.16E+14	8.15E-09	-	Stable	Cr55	2.35E+14	2.14E-08	7.65E+11	2.12E+02
Li7	7.76E+11	9.04E-12	-	Stable	Mn53	2.26E+16	1.99E-06	1.35E+02	1.16E+14
Be9	4.80E+18	7.19E-05	-	Stable	Mn54	1.12E+20	9.99E-03	2.86E+12	2.70E+07
Be10	7.93E+16	1.32E-06	1.09E+03	5.05E+13	Mn55	1.42E+22	1.30E+00	-	Stable
B10	1.54E+10	2.56E-13	-	Stable	Mn56	6.35E+16	5.90E-06	4.74E+12	9.28E+03
B11	8.25E+11	1.51E-11	-	Stable	Mn57	4.79E+12	4.53E-10	3.44E+10	9.66E+01
C12	4.96E+22	9.88E-01	-	Stable	Mn58	1.34E+09	1.29E-13	1.43E+07	6.52E+01
C13	5.60E+20	1.21E-02	-	Stable	Fe54	4.26E+23	3.82E+01	-	Stable
C14	2.53E+16	5.87E-07	9.68E+04	1.81E+11	Fe55	2.28E+21	2.08E-01	1.83E+13	8.63E+07
N14	4.51E+13	1.05E-09	-	Stable	Fe56	6.70E+24	6.23E+02	-	Stable
N15	5.95E+10	1.48E-12	-	Stable	Fe57	4.66E+23	4.41E+01	-	Stable
Ar38	9.37E+09	5.91E-13	-	Stable	Fe58	4.04E+22	3.88E+00	-	Stable
Ar39	3.72E+10	2.41E-12	3.04E+00	8.49E+09	Fe59	6.63E+18	6.49E-04	1.20E+12	3.85E+06
Ar40	1.89E+14	1.26E-08	-	Stable	Fe60	1.11E+18	1.10E-04	1.62E+04	4.73E+13
Ar42	1.81E+10	1.26E-12	1.20E+01	1.04E+09	Fe61	1.02E+10	1.03E-12	1.97E+07	3.59E+02
K39	9.84E+08	6.37E-14	-	Stable	Co57	5.76E+15	5.45E-07	1.70E+08	2.35E+07
K41	1.48E+12	1.01E-10	-	Stable	Co58	2.99E+19	2.88E-03	3.38E+12	6.12E+06
K42	9.15E+08	6.38E-14	1.43E+04	4.45E+04	Co58m	1.24E+17	1.19E-05	2.66E+12	3.22E+04
K43	1.66E+10	1.19E-12	1.44E+05	7.99E+04	Co59	1.45E+21	1.42E-01	-	Stable
Ca42	1.46E+15	1.02E-07	-	Stable	Co60	3.07E+20	3.05E-02	1.28E+12	1.66E+08
Ca43	4.54E+17	3.24E-05	-	Stable	Co60m	8.71E+14	8.67E-08	9.61E+11	6.28E+02
Ca44	9.46E+17	6.91E-05	-	Stable	Co61	2.38E+14	2.41E-08	2.77E+10	5.94E+03
Ca45	1.34E+16	1.00E-06	6.62E+08	1.41E+07	Co62	4.29E+09	4.41E-13	3.30E+07	9.00E+01
Ca46	6.35E+16	4.85E-06	-	Stable	Co62m	2.00E+10	2.06E-12	1.66E+07	8.35E+02
Ca47	2.31E+12	1.80E-10	4.08E+06	3.92E+05	Ni58	6.38E+23	6.14E+01	-	Stable
Ca48	6.03E+10	4.80E-12	-	Stable	Ni59	1.97E+22	1.93E+00	5.71E+09	2.40E+12
Sc45	5.17E+17	3.86E-05	-	Stable	Ni60	2.85E+23	2.83E+01	-	Stable
Sc46	1.00E+17	7.66E-06	9.60E+09	7.24E+06	Ni61	2.50E+22	2.53E+00	-	Stable
Sc46m	5.52E+10	4.21E-12	2.05E+09	1.87E+01	Ni62	2.99E+22	3.07E+00	-	Stable
Sc47	6.56E+15	5.11E-07	1.57E+10	2.89E+05	Ni63	5.93E+21	6.20E-01	1.32E+12	3.12E+09
Sc48	3.92E+14	3.12E-08	1.73E+09	1.57E+05	Ni64	1.09E+22	1.16E+00	-	Stable
Sc49	2.11E+12	1.72E-10	4.27E+08	3.43E+03	Ni65	4.10E+15	4.43E-07	3.14E+11	9.07E+03
Ti46	1.03E+22	7.83E-01	-	Stable	Cu63	6.99E+20	7.30E-02	-	Stable
Ti47	9.15E+21	7.13E-01	-	Stable	Cu64	5.06E+15	5.37E-07	7.67E+10	4.57E+04
Ti48	8.12E+22	6.46E+00	-	Stable	Cu65	2.65E+20	2.86E-02	-	Stable
Ti49	1.82E+22	1.48E+00	-	Stable	Cu67	1.38E+09	1.53E-13	4.28E+03	2.23E+05
Ti50	7.02E+21	5.82E-01	-	Stable	Zn64	1.02E+19	1.08E-03	-	Stable
Ti51	1.24E+13	1.05E-09	2.47E+10	3.48E+02	Zn65	6.79E+15	7.32E-07	2.23E+08	2.11E+07
V50	2.27E+20	1.88E-02	3.34E-05	4.70E+24	Zn66	6.27E+18	6.86E-04	-	Stable
V51	2.10E+22	1.78E+00	-	Stable	Zn67	4.10E+16	4.55E-06	-	Stable
V52	6.78E+14	5.85E-08	2.09E+12	2.25E+02	Zn68	3.06E+15	3.45E-07	-	Stable
V53	7.97E+11	7.01E-11	5.68E+09	9.72E+01	Zn70	2.41E+09	2.80E-13	-	Stable
V54	1.09E+09	9.77E-14	1.52E+07	4.98E+01	Ga69	2.34E+13	2.68E-09	-	Stable

Analysis of obtained results indicated that main isotopes which were produced by neutron irradiation are hydrogen, deuterium, helium, vanadium, calcium, manganese, cobalt. After a long time of irradiation additionally cooper and zinc were produced. The hydrogen production depending on irradiation time was shown in Fig. 3. Production of other major isotopes was shown in Fig. 4.

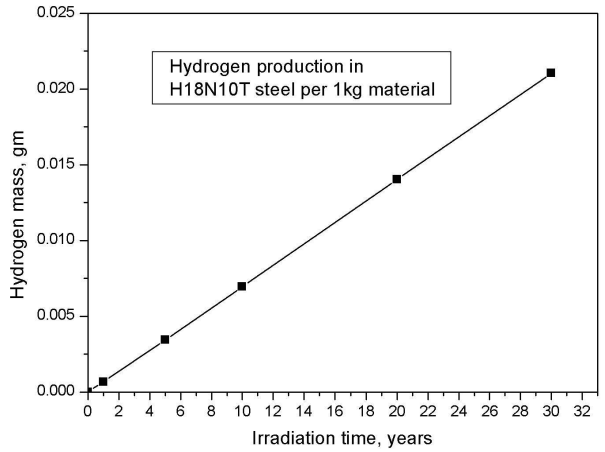

Fig.3. *Hydrogen production depending on irradiation time*

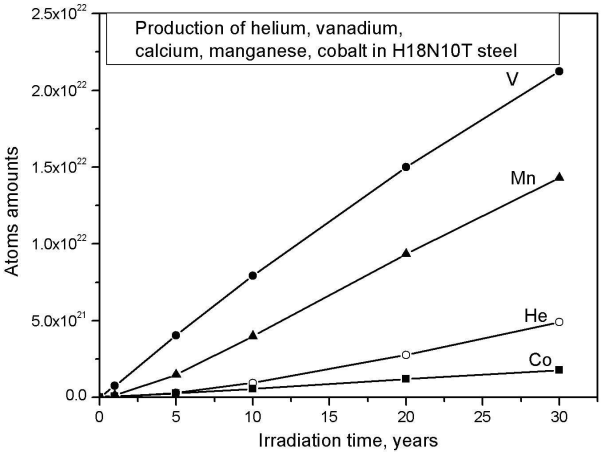

Fig.4. *Some major isotopes production depending on irradiation time*

Obtained results indicated that production of hydrogen is a result of $(n,p) \rightarrow p + e^- \rightarrow^1 H$ reaction. Reaction of such type lead to other isotopes production for example production of vanadium, manganese, cobalt as a result of ^{56}Fe(n,p)^{56}Mn, ^{52}Cr(n,p)^{52}V, ^{58}Ni(n,p)^{58}Co reactions. It is necessary to note that production of other isotopes causes to other reactions for example ^{58}Ni(n,γ)^{59}Ni(β^+)^{59}Co. Helium production is exist because of $(n,\alpha) \rightarrow \alpha + 2e^- \rightarrow^4 He$ reaction. Such kinds of reaction for iron lead to chromium production for chromium lead to titanium production and for titanium lead to calcium production also because of such reaction for nickel production of iron is occur.

In a long irradiation time also production of other isotopes is possible, for example a cooper production from ^{62}Ni(n,γ)^{63}Ni(β^-)^{63}Cu(n,γ)^{64}Cu reactions. Velocity analysis of isotopes accumulation will be realized in future investigation. But it is necessary to note that velocity of isotopes accumulation mainly depending on reaction cross-section and numbers of active pathways during isotope production. Moreover the main factor of accumulation velocity is a diffusion and adhesive processes which were not taking into account in this investigation.

Developed simulation methodic create possibility to make analysis of modification of isotopic composition of stainless steel H18N10T under long time irradiation such steel by neutrons with energy distribution corresponding to spectrum of VVER-1000 reactor. In our simulation we taking into account all active pathways of isotopic production. Also the modern reaction and decay data bases during simulation processes were used. Using our simulation methodic modification of composition steel by nuclear reaction of transmutation was obtained. Obtained results proved that main process under steel activation is productions of such isotopes as hydrogen, helium, vanadium, manganese and cobalt.

In future investigation will be necessary to make analysis of chemical composition changes of a given material after irradiation in more detail. Hence, it is possible to modify methodic for specific purposes and choose from obtained isotopes most important ones with relation to a solid state physics. Also we will take into account diffusion and other processes which will have an influence on gases migration in material.

Applying of the given simulation methodic is more reasonable as a part of radiation physics investigation. It is necessary for adequate model development with taking into account major physical processes which will be active in material under irradiation.

4. CONCLUSIONS

Developed simulation methodic create possibility to provide simulation of activation processes in condition of high intensity neutron fluxes of stainless steel H18N10T during long irradiation period.

This simulation methodic by applying of the most modern data bases such as EAF Cross Section and EAF Decay allow to simulate modification of isotopic composition of a given material with taking into account all possibly reactions of isotopic production which will be active under neutron irradiation.

Analysis of obtained results indicates that major elements which will be accumulate during irradiation time in material are helium and hydrogen.

In future plans we are going to make co-operative investigation in a nuclear physics field and in a radiation damages physic field for the purpose of development of complex simulation model of construction materials behavior under neutron irradiation. According this activity it is necessary to have collaboration with specialists in a field of radiation damages and diffusion processes.

References

1. G.A. Filippov, P.A. Antikajn. Primenenie suschestvuyuschih konstrukcionnyh materialov dlya izgotovleniya vnutrikorpusnyh ustrojstv i teplovydelyayuschih sborok legkovodnyh reaktorov // *Teplo'energetika.* 2005, v.8, p.2-8 (in Russian).

2. *GEANT4 Physics Reference Manual. GEANT4 Working Group.* CERN, June 21, 2004.

3. J. Breismeister. Ed. *MCNP - A General Monte Carlo N-Particle Transport Code. LA-13709-M.* Los Alamos National Laboratory: Los Alamos, NM, 2000.

4. R.A. Forrest. *FISPACT-2003: User manual.* UKAEA FUS 485. 2002.

5. J. Sidell. *EXTRA - A digital computer program for the solution of stiff sets of ordinary value, first order differential equations.* Atomic Energy Establishment of Winfrith (AEEW-R).-799, 1972.

КОМПЬЮТЕРНОЕ МОДЕЛИРОВАНИЕ ИЗМЕНЕНИЯ СОСТАВА НЕРЖАВЕЮЩЕЙ СТАЛИ, ИНДУЦИРОВАННОЕ НЕЙТРОННЫМИ ЯДЕРНЫМИ РЕАКЦИЯМИ

Е.В. Рудычев, С.И. Прохорец, М.А. Хажмурадов, Д.В. Федорченко

Рассмотрена проблема изменения состава конструкционных материалов реактора ВВЭР-1000 в результате ядерных реакций и соответствующих активационных процессов, которые проходят в материале под воздействием нейтронного облучения в течение длительного периода эксплуатации материала, в условиях нейтронного потока реактора ВВЭР-1000. Для моделирования подобных задач разработана специальная методика, основанная на современных методах компьютерного моделирования. Проведено моделирование изменения изотопного состава аустенитной стали Х18Н10Т под действием нейтронного облучения в течение длительного времени.

КОМП'ЮТЕРНЕ МОДЕЛЮВАННЯ ЗМІН СКЛАДУ НЕРЖАВІЮЧОЇ СТАЛІ, ІНДУКОВАНИХ НЕЙТРОННИМИ ЯДЕРНИМИ РЕАКЦІЯМИ

Є.В. Рудичев, С.І. Прохорецъ, М.А. Хажмурадов, Д.В. Федорченко

Розглянуто проблема зміни складу конструкційних матеріалів реактора ВВЭР-1000 у результаті ядерних реакцій і відповідних активаційних процесів, які проходять у матеріалі під впливом нейтронного опромінення протягом тривалого періоду експлуатації матеріалу в умовах нейтронного потоку реактора ВВЭР-1000. Для моделювання подібних завдань розроблена спеціальна методика, заснована на сучасних методах комп'ютерного моделювання. Проведено моделювання зміни ізотопного складу аустенітної сталі Х18Н10Т під дією нейтронного опромінення протягом тривалого часу.