THE POSSIBILITIES OF FAST POWER REACTORS TO CREATE HIGH INTENSITY RADIOACTIVE SOURCES

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ABSTRACT

Fundamental possibilities of fast power reactors to produce useful radioactive isotopes by creating special irradiating assemblies and side blanket realignment have been considered. The experience of the possibility substantiation and the pilot production of cobalt-60, chromium-51 and argon-37 isotopes have been described and briefly analyzed. It has been shown that the isotope production can be arranged keeping the other main reactor neutron and physical parameters. The evaluation of the isotope production capabilities in typical fast reactor of mean power has been presented for BN-600 as an example. On the whole, the conclusion has been made that this production possesses good potential and supplements organically the main NPP reactor function of power production.

1. INTRODUCTION

One of the certain advantages of fast reactors is the neutron plenty not participating in chain reaction. The most well-known and explored way of the neutrons utilization is production of the secondary nuclear fuel – plutonium. Nowadays, when not the problem of plutonium production but plutonium stock removal problem is really actual, the research on this neutron surplus application for long-lived radiotoxic nuclide transmutation is really extensive. Why not to use the fast reactor potential in solving the other worthy problem – artificial radioactive isotope production applied in radiation engineering, medicine and science?

The isotope current demand has reached so large scale that the research reactors supplying these isotopes so far can satisfy this demand with more and more difficulties. It becomes clear that the isotope production in the research reactors will never be profitable under conditions of diminishing state support and total accounting of all expenses. That's why we need to look for new profitable and efficient ways of radionuclide production if we make our aim to maintain the possibility of their useful medical or engineering application.

The use of fast power reactors, able to solve their economic problems without the state support in general, for radionuclide production seems to be very advanced way for solving the problem under discussion. However, the use of such Russian thermal neutron reactors as WWER (PWR analog) or RBMK is seriously restricted by relatively low neutron flux density, which does not allow to obtain the preferred radionuclide specific activities. Our experience has shown that the problems of different radionuclide production with very attractive parameters of specific activity and total yield can be solved quite flexibly in BN type reactors. In this paper we would like to summarize the Russian experience in this field with the examples of several specific projects. International experience is also available in this field. The work performed at the US FFTF reactor [1] can be mentioned, in particular. Unfortunately, this activity has not got further serious development.

2. MAIN PROBLEMS AND APPROACH

One of the reasons according to which fast reactors were not recently considered as promising isotope producers is too low values of neutron reaction cross sections in the "fast" energy area. This fact limits the level of Co-60 specific activity, for example, by a value of 10-20Ci/kg, which is absolutely unacceptable for the most applications. Obtaining high levels of radionuclide specific activity is the first key problem. By now three main approaches have been determined.

The first one is the use of threshold reactions with the outlet of charged particles and the target further chemical processing and specific nuclide recovery. In this case the nuclide is carrier free and its specific activity is very high as a rule (close to maximum). Large amount of high-energy neutrons in the fast reactor spectrum naturally presents the most favorable possibilities for the use of (n,p) and (n,α) reactions. In particular, $^{32}S(n,p)^{32}P$, $^{33}S(n,p)^{33}P$, $^{35}Cl(n,\alpha)^{35}S$, $^{89}Y(n,p)^{85}Sr$ reactions have already found their practical application – for ^{32}P, ^{33}P, ^{35}S and ^{89}Sr isotope production, correspondingly. The project of the intense neutrino source production on the base of ^{37}Ar isotope produced in $^{40}Ca(n,\alpha)^{37}Ar$ reaction will be considered below in more detail.

As far as large amount of radionuclides is produced in neutron reactions running on thermal neutrons mainly an idea of irradiation assemblies of "trap" type has been proposed. It consists of creation of effective moderator – zirconium hydride – containing assembly, optimized in such a way that it provides the best conditions (neutron spectrum, in particular) for one or another radionuclide local production within one assembly. In this case it should be kept in mind that this assembly, or irradiating assembly (IA), as it is often called, also affects the reactor neutron field where it is installed. If the assembly with zirconium hydride is put into BN reactor core, the high-energy release spikes take place in the fuel elements nearest to IA. That's why special measures shall be made preventing the leak of moderated neutrons out of IA. The simplest method is the use of absorbents with large cross sections in thermal energy range at the assembly periphery. Thus, IA works according to the principle – "let in and not let out", i.e. "trap". Namely the use of "traps" allows considering the fast reactor as a real producer of different isotope products because it allows the most popular isotopes - ^{60}Co, ^{14}C,

^{192}Ir production and many others as well. We consider the **second** basic approach to the isotope production in the use of the "trap" type assemblies.

Some applications of this approach will be considered below. The experience obtained in this case has completely revealed the following problem. In any case the isotope production in the power-producing BN reactor will be the secondary one supplementing the energy production process. That's why it should be carried out with the minimal impact on the reactor core. Only in this case good economic characteristics can be reached as well as the isotope acceptable price. However, the use of moderator and strong absorbent leads to the reactor neutron field disturbances. In particular, the reactor reactivity reserve reduction can become noticeable. The acceptable level of disturbances can be reached if the number of assemblies is relatively small, for example, at the expense of such assembly arrangement in the blanket. In this reactor area the fuel assembly heat release is far from the limiting value and that's why some variations of the energy release field are permissible. Reactor reactivity variation can be compensated, as a rule, by insignificant additional fuel loading.

However, if the number of IA is large the more effective approach is the creation of special spectral sub-zones intended for the industrial large-scale isotope production. At present namely this **third** approach must be the most defensible. Modern BN reactor designs such as BN-600 and BN-800 reactor hybrid core designs propose to refuse from reproducing shielding with depleted uranium and replace it by the steel reflector. Neutrons leaking from the core and formerly providing secondary nuclear fuel production are lost irreversibly in this case without any profit, moreover, activating the structure materials. Besides, as the steel reflector is more transparent for neutrons the additional efforts should be made on reducing the neutron charge on the reactor internal irremovable structures. Taking this into account the construction of a sub-zone made of zirconium hydride in the area of the side steel reflector will facilitate the neutron surplus most effective utilization.

While creating such sub-zones we can refuse from strong absorbers applied on the "core-moderator" border. As the calculation shows the moderating assemblies with zirconium hydride layout in the second-third rows of the steel reflector does not result in noticeable energy release spike in the core. Rejection of absorbers results in their less influence on the reactor reactivity and less fuel additional charge, correspondingly. In this concept the reactivity loss can be compensated by the fuel enrichment variation. In BN-600 reactor hybrid core it can be done by plutonium share increasing in MOX fuel. Moreover, this increasing entirely corresponds to the basic idea of hybrid core because it will facilitate the improvement of ex-weapons plutonium utilization characteristics.

Summarizing the brief conceptual consideration let us state the second main problem, which we have to face during investigation of the isotope production in NPP power reactors. This is **the minimization of reactor neutron field disturbances and complete maintenance of electric power production**. The search of compromise between the achievement of the needed parameters of specific activity and general radionuclide production and reactor nuclear and physical characteristics connected with these variations defines the scope of problem for scientific investigation. It should be also mentioned that the assembly complex heterogeneous structure with moderator presents specific difficulties in the design forecasting of their characteristics. In this connection a set of calculation and experimental work is carried out allowing the nuclear and radiation safety reliable substantiation of fast reactor cores intended for radionuclide production and verification research.

3. EXPERIENCE OF SOME ISOTOPE PRODUCTION AND ESTIMATION OF PROSPECTS.

3.1. STUDY OF THE POSSIBILITY OF COBALT-60 PRODUCTION.

This isotope is one of the most popular, is it in extreme and stable demand at the world market. That's why no wonder that we have paid much attention to this isotope production.

In order to prove the possibility of Co-60 production of about 100Ci/g specific activity in BN-350 reactor two IA have been tested. Their diagram is presented on the Fig.1. Container with initial target and zirconium hydride moderator is in the center of IA. The rest space of IA is furnished with the fuel elements made of depleted uranium oxide. Before the irradiation at the power level the experiment has been carried out at the minimal power level in order to study and test IA neutron and physical characteristics and substantiate the safety of industrial irradiation. In particular, the rate of cobalt-59 capture reaction has been measured, which allowed the cobalt-60 activity estimation.

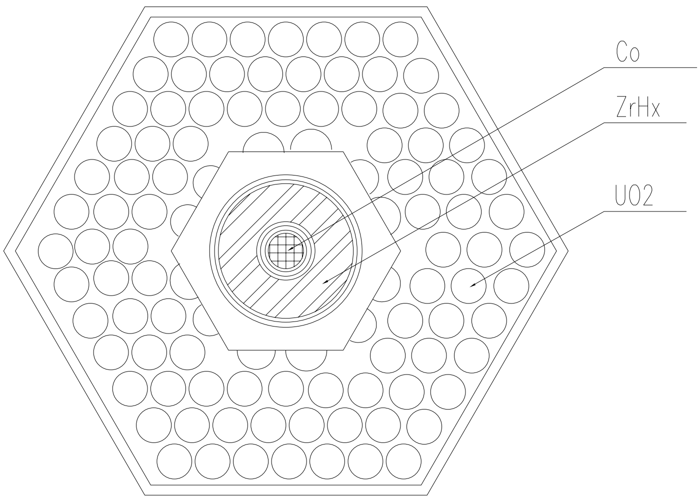

Fig.1. Diagram of experimental IA, tested in BN-350 reactor.

As it was the first experience of such assembly testing in BN-350, the decision had been made to irradiate the first IA during a half of the designed period. After irradiation and fabrication of 6 sources they were calibrated as well. The results are presented in the Table 1. It can be seen from the Table 1 that both calculated and extrapolated values at small power level are in good agreement with the actual activities obtained. Although the actual activity was less than 100Ci/g it will be certainly reached after the full-scale designed irradiation period. Unfortunately, the test of the second IA has not been completed due to organization reasons.

Table1. Measurement results of Co-60 source activity, produced in the 1st IA of BN-350, irradiated about a half of the designed period.

Nsource	*Extrapolated specific activity, Ci/g	Calibrated specific activity, Ci/g	Calculated specific activity, Ci/g	Calc./Calibr.
019	51.5	50.6	49.1	0.97
013	45.9	45.3	43.8	0.97
021	16.4	18.6	15.6	0.84
018	31.1	30.4	29.7	0.98
024	31.1	32.0	29.7	0.93
017	47.5	46.7	45.3	0.97

*Extrapolation of low power experiment.

IA influence on the energy release in fuel assembly has been also studied in the experiment at low power level. Uranium-235 fission rate distribution directly near IA is presented on the Fig.2. The energy release spike is observed on IA border. Nevertheless, this spike was not dangerous due to the energy release gradient on the core border and low power level of BN-350 reactor (at that time reactor was operated at 75% of standard power level).

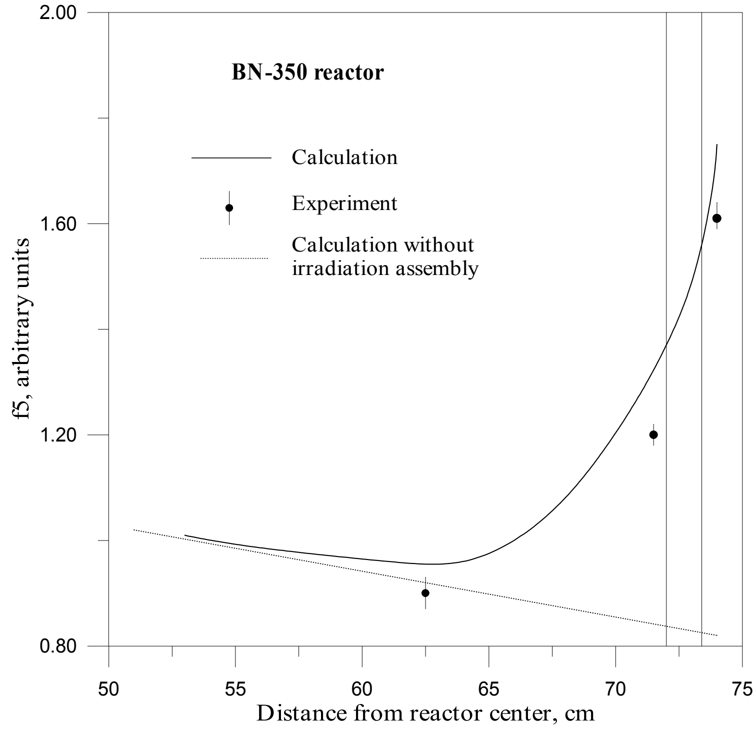

Fig.2. Distribution of U-235 fission rate near irradiating assembly.

In spite of the result achieved in Co-60 specific activity, the total production was not large in the assembly described above – not more than 30KCi. Our further effort was directed on the research of the production capability of greater amounts of product. The trap versions containing several cobalt rods and much more moderator in one assembly have been considered and tested at the KBR critical facility of SSC RF IPPE. This, in turn, had led to the necessity of the use of more strong absorbers at the assembly periphery such as europium and boron carbide. In optimization process the amount, dimensions and geometry of cobalt and moderating elements varied as well as the density, material and geometry of absorbing elements. The study has resulted in the following IA diagram (see Fig.3). It has been successfully applied by the efforts of several Russian enterprises (OKBM, BNPP, Polymetal plant, etc.) and tested in the second row of BN-600 reactor side blanket. Comparison of the cobalt-60 expected activities and the measurement results carried out by the personnel of Beloyarskaya NPP is presented in Table 2. One can see from the table that production parameters are extremely high both in specific activity and total production.

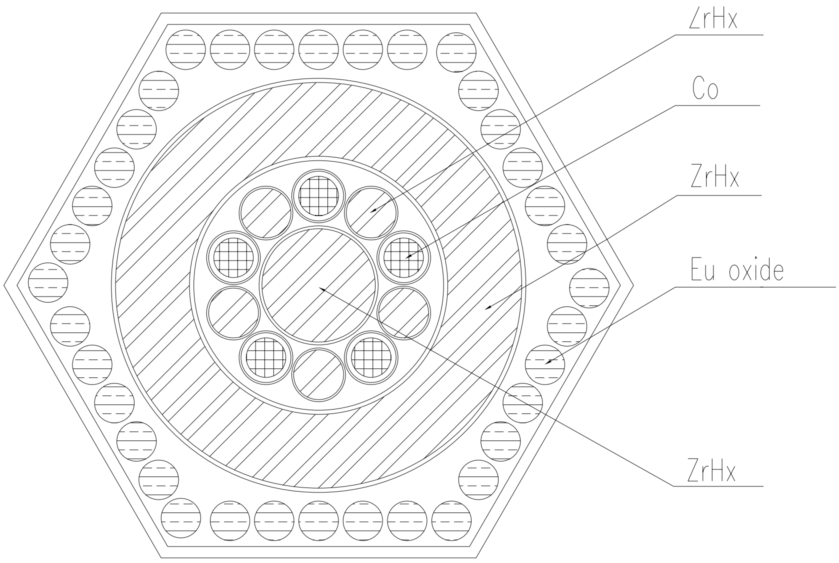

Fig.3. Experimental irradiating assembly for Co-60 production in BN-600 reactor.

Table 2. Measurement results of Co-60 source activity, produced in 1st experimental IA in BN-600.

	Max. specific activity Ci/g	Aver. specific activity Ci/g	Co-60 production per IA, KCi
Calculation	112	89	149
Calibrated	109±8	87±6	130±9

The presence of very efficient europium oxide absorber not only has prevented the energy release spikes typical to BN-350 IA but also resulted in some “fall” of the field in IA vicinity. It has been measured that each IA brings negative reactivity, about -0.04% Δk/k. Parameters of Co-60 production in BN-600 have been estimated taking this into account. They are presented in Table 3. Under the core version available the reasonable amount of IA not greatly distorting the reactor neutron field is estimated as 12 – 15. Corresponding Co-60 production can become a very distinct value – up to 0.8 – 1.0 MCi. Much more possibilities are being opened in connection with the proposed transfer to steel side reflector (SSR). In this case the use of IA mentioned is also possible. However, it is expedient to replace it into the third row of reflector in order to reduce its impact on the core. Production parameters are still the same in this case. From the other side production parameters improve greatly if the absorbing elements are removed from IA composition. When the number of IA in steel reflector is big an actually moderating sub-zone is being developed where up to 10MCi per year of Co-60 can be produced. It is certainly a great amount and this shows the real potential of the fast reactor of mean power for the isotope production.

Table3. Estimation of Co-60 production parameters in the BN-600 reactor.

Parameter	Production in separate IA		Production in moderating sub-zone		
Number of IA	12	12	12	75	79
IA location	2^{nd} row side blanket	3d row SSR	3d row SSR	3d row SSR	2^{nd} row SSR
Reactivity variation, %Δk/k	-0.60	-0.60	-0.65	-1.72	-3.8
Compensation method	10 FA HEZ	10 FA HEZ	11 FA HEZ	FA with MOX	FA with MOX
Irradiation period, eff. days	675	725	580	1160	725
Co-60 average specific activity, Ci/g	90	97	101	82	93
Co-60 total production in one IA, KCi	154	173	346	281	319
Co-60 annual total production in BN-600, MCi	0.79	0.84	2.09	5.30	10.2

3.2. DEVELOPMENT OF ARTIFICIAL NEUTRINO SOURCES OF HIGH ACTIVITY

Direction connected with the use of artificial neutrino sources is recently developed in neutrino fundamental physics. The so-called problem of solar neutrino is the most urgent. In the first and the single for a long time experiment with chlorine-argon radiochemical detector the solar neutrino flux appeared to be three times less than the theoretic one. At present the experiments with gallium-germanium radiochemical detectors are carried out in Russia (SAGE collaboration [3]) and in Italy (GALLEX collaboration [4]). It is very important that both experiments give the information of the main (p-p) thermonuclear reaction on the Sun and confirm the solar neutrino deficiency.

The essential part of this work is the direct confirmation of the facility correct operation by the measurement of neutrino flux from the source of known intensity – the so-called calibration experiment. Artificial ^{51}Cr source is very suitable for gallium-germanium telescope.

The projects of the other neutrino detectors are also under consideration, iodine-xenon one, for example, which is developed at Pennsylvania University, USA. Chromium source cannot be used for calibration due to its different sensitivity energy range. That's why we consider the possibility of neutrino source development based on ^{37}Ar for this detector [5].

There are also other problems demanding the development of powerful neutrino sources. Extremely high requirements presented to them both in total activity (0.5-1 MCi and even more) and specific activity (1000Ci/g and more) are essential for us. We consider their development as the research of the maximal ability of fast reactors from the point of view of high quality radionuclide production.

3.2.1. ^{51}Cr neutrino source for SAGE (Russian and American gallium-germanium solar neutrino experiment).

Our experience on ^{51}Cr source development in BN-350 reactor for SAGE experiment is described in this paragraph. In view of high requirements to specific activity special experiments studying the possibility of such neutron spectrum forming, which allows ^{51}Cr production of needed condition, have been carried out providing reactor safety. For that purpose the models simulating IA with different moderator share have been developed. Radiation capture average cross section has been measured on Cr-50 isotope (see Fig.4). As it is seen from the figure the cross section of this reaction is very small in the fast energy range (millibarns). However, zirconium hydrate moderating ability is quite high to allow forming a spectrum with large share of thermal neutrons and cross section increasing up to several barns within one assembly of 9.6cm size. We decided to use chromium-50 of about 90% enrichment to achieve high specific activity. In this case the spatial shielding effects begin to play essential part. Nevertheless, these effects do not present an important obstacle under the target chosen dimension 7mm.

As the result of optimization IA has been developed, which diagram is presented on the Fig.5. It differs by the maximal use of the assembly volume for moderator layout and thinner europium oxide absorbing elements. Their thickness was minimal and there were the energy release spikes on the border between IA and the core. However, at the moment of irradiation the reactor power was only a half of standard one. That's why the mentioned disturbance was permissible.

514 g of 90% Cr-50 irradiated in two IA on the border of BN-350 core has been used for the source production. Up to 1330Ci/g specific activity of Cr-51 has been achieved and the average one was about 1007Ci/g. The source total activity was 0.66±0.01MCi by the end of irradiation. The source has been successfully used in calibration experiment at Baksan neutrino observatory and confirmed the experimental facility registration efficiency and the solar neutrino problem presence.

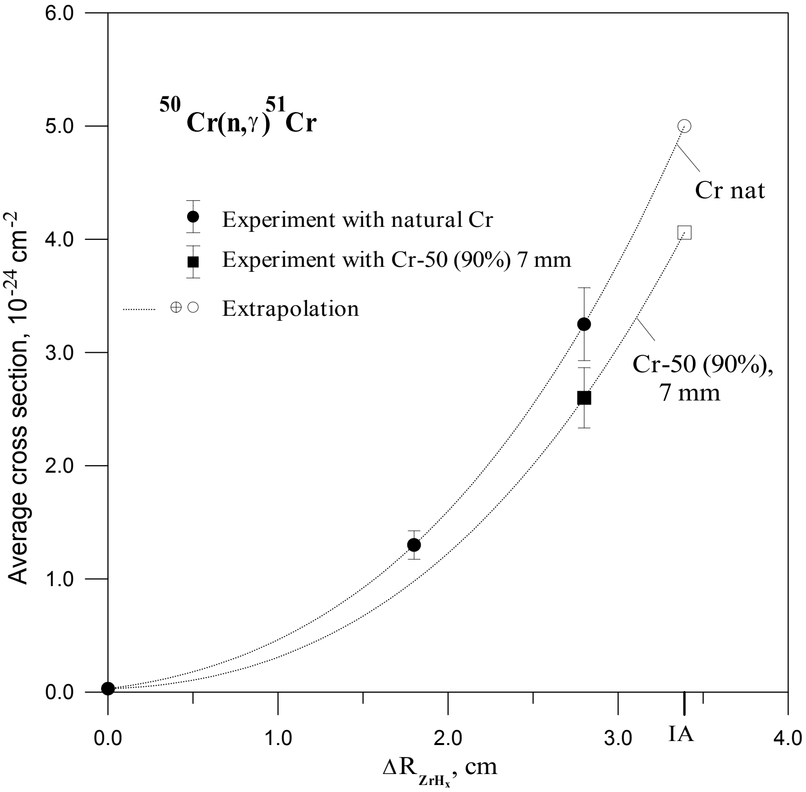

Fig.4. Cr-50 average capture cross section dependence on ZrH_x thickness.

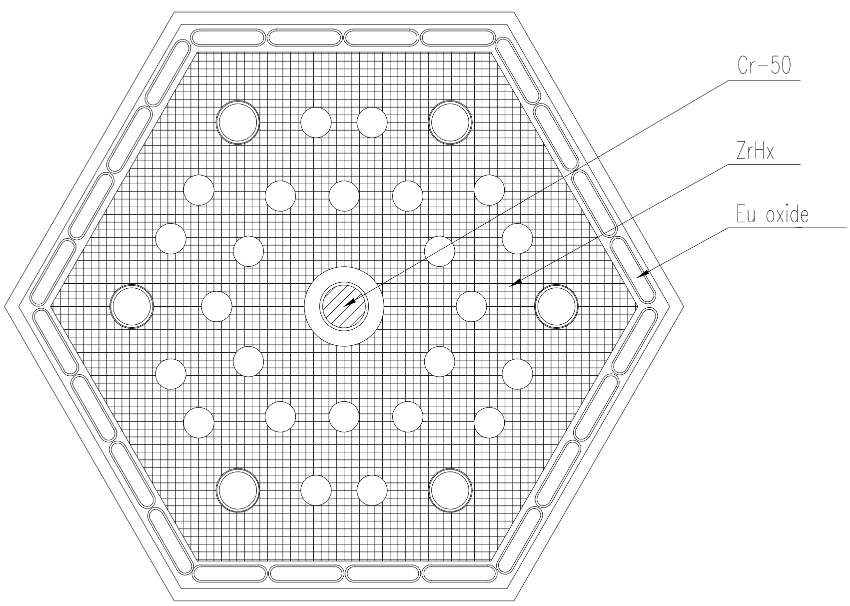

Fig.5. Irradiation assembly for Cr-51 production.

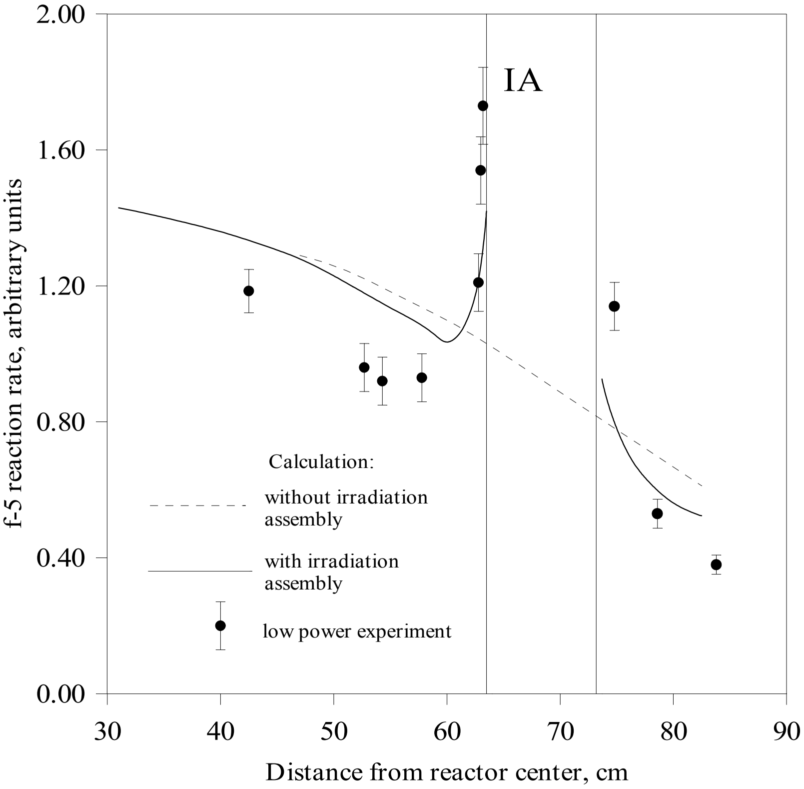

Fig.6. F-5 rate distribution in Cr-51 irradiation assembly area.

3.2.2. Research of the possibility of neutrino source creation on the base of ^{37}Ar isotope.

At present the research is carried out on substantiation of a possibility of 1-2MCi neutrino source production in the BN-600 reactor. For this purpose it is proposed to use (n,α) reaction on calcium with further argon recovery from the target irradiated. This reactor cross section has a threshold character and is relatively small (millibarns). This disadvantage can be compensated at the expense of massive target application. In this case large blanket can be used and it is possible to irradiate up to several hundreds kg of calcium-containing target.

The choice of the target is important in principle. As calculation shows the use of calcium oxide as a target does not result in any noticeable variation of reactor reactivity reserve and energy release distribution even when the blanket is loaded much. Thus, the target can be irradiated under the standard operation mode of BN-600 reactor. Metal calcium target results in some reduction of reactivity reserve. An influence on the energy release field is also present. However, it is not considerable.

Forecasting estimation of expected activity depending on the number of CaO irradiating assemblies located in BN-600 radial blanket and duration of the target chemical processing after irradiation are presented on the Fig.8. The last factor is of no small importance due to relatively small argon-37 half-life period 35 days. It can be seen from the figure that 1-2MCi activity production is completely workable. Currently a team of specialists from several Russian enterprises and Beloyarskaya NPP develops the first experimental pilot model of the source of about 0.4MCi activity. It is expected to produce the source and use it in neutrino research within nearest two years.

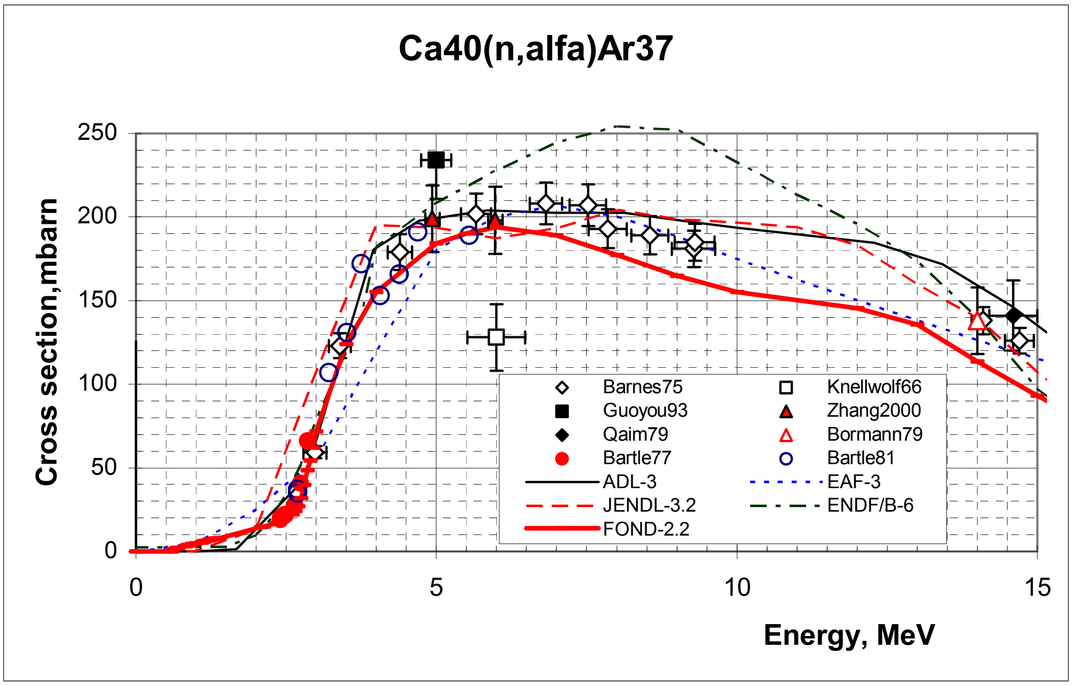

Fig. 7. Experimental data and estimation of ^{40}Ca(n,α) reaction energy progress.

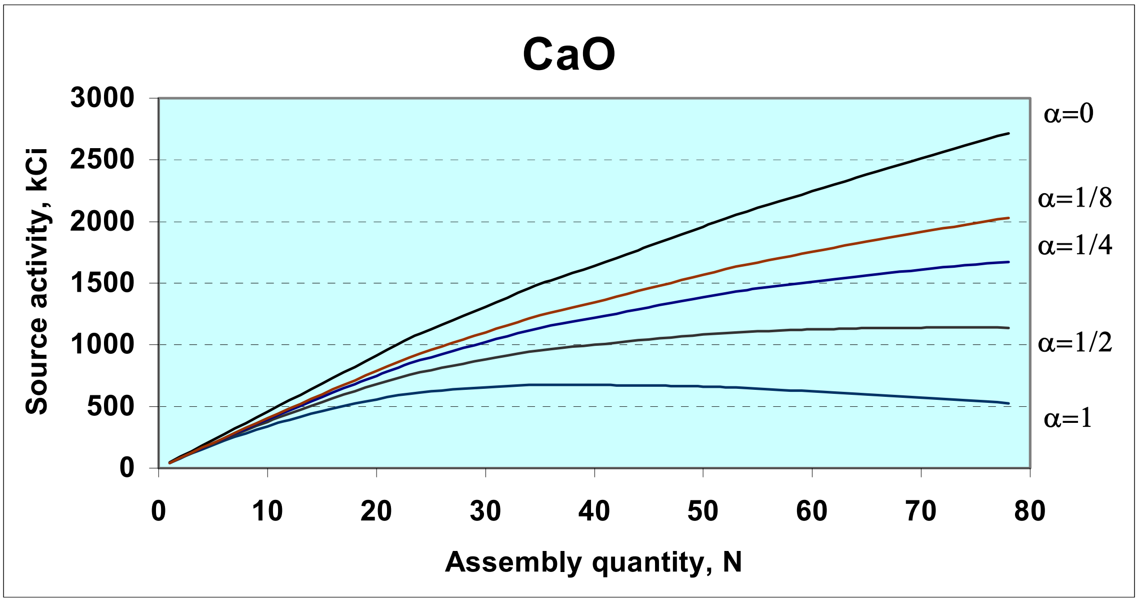

Fig.8. The expected activity of Ar-37 depending on IA amount and handling rate α (day/assembly).

3.3. PROSPECTS FOR SOME OTHER ISOTOPE PRODUCTION.

By present we have considered and estimated the mean power fast reactor capability to produce, besides the considered nuclides, the other isotopes such as C-14, Sr-89, Ir-192, Pu-238 with the use of above mentioned approach in BN-600 reactor as an example. Final data summarizing the potential for challenging isotope production in BN-600 reactor are presented in the Table 4.

Table 4. Production parameters of great potential in the BN-600 reactor.

Aim isotope	Target	Production parameters		
		Specific production, Ci/g	Production per one assembly, KCi	Annual production in reactor
^{14}C	AlN	0.041	0.056	0.9 KCi
^{51}Cr	^{50}Cr	484	390	4.7 MCi
^{60}Co	Co	93	319	10.2 MCi
^{89}Sr	Y_2O_3	0.016	0.270	4.8 KCi
^{192}Ir	Ir-nat	712	82	960 KCi
^{238}Pu	^{237}Np	1.0	32	3.8 MCi
^{238}Pu	^{241}Am	0.42	14	1.8 MCi

CONCLUSION

Performance capability of such reactor as BN-600 in its total activity and expected profitability greatly overlaps the capability of research high-flux reactors such as, for example, CM-2 or HFIR, approaching to them in specific activity characteristic. However, the authors do not consider BN-600 to be an alternative, most likely, it is the supplement to these specialized isotope reactors. The real role of power-producing BN reactors is the isotope industrial batch commercial production.

Under conditions of demand reduction to the nuclear fuel extended reproduction the useful radionuclide production can supplement organically the main functions of fast reactor on energy production together with limited production of secondary nuclear fuel. That's why, evidently, it is expedient to research this possibility as applied to the fast reactor innovation projects.

Being a development of initial ideas of BN reactors the artificial radioactive isotope production requires more technical flexibility during the project development for one or another isotope recovery, which is connected with specific individual requirements to every product. The examples presented in the report show its presence. Thus:

- Considering Co-60 production one can see the capabilities of industrial isotope production - BN-600 reactor can double Co-60 Russian production capability in practice;
- Considering Cr-51 production one can get an idea of solution of the most complicated problem in radionuclide production – achievement of extremely high specific activities;
- Considering argon-37 production one can see the possibility of threshold reaction application and production of great amounts of carrier free radionuclides.

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