ACCELERATOR-BASED GAMMA NEUTRON TRANSMUTATION OF RADIONUCLIDES AS A NEW TECHNOLOGY FOR THE NUCLEAR FUEL CYCLE

I.P.Eremeev, International Business Nucleonic, Moscow, Russia

ABSTRACT

A new transmuting technology for incinerating long-lived nuclear waste and breeding fissile fuel is proposed as an application of the physical approach [1, 2]. The physical parameters of the technology, such as a transmuting rate, an intensity of photoneutrons generated in the gamma transmutation process, an energetic "cost" of a transmutation event have been determined by multigroup calculations. It is shown that the approach proposed allows any long-lived radionuclide (both transuranium actinide and fission product) to be transmuted at a much greater rate than that of its build-up in any operating NPP reactor at a much less energy consumption than an energy accompanying their production in a reactor core. To realize the technology proposed requirements to a cyclic electron accelerator have been formulated.

I. THE GNT METHOD

The problem of radioactive waste (RW) would be solved cardinally, if it were a success in finding such feasible and economically expedient method, which would allow, on the one hand, long-lived fission radionuclides (FR) to be transmuted to stable isotopes and actinide radionuclides (AR) to be incinerated or transmuted to isotopes fissionable by thermal neutrons and, on the other hand, additional fissile fuel to be produced to compensate at least in part the energy consumption for RW transmutation.

The approach proposed provides an unique opportunity to do it.

The method of gamma neutron transmutation (GNT) is based on photoneutron reactions (γ, *xn*) induced by magnetic bremsstrahlung (synchrotron) radiation of an ultrarelativistic electron beam turned periodically in a uniform or spatially periodic magnetic field [1, 2]. The possibility of industrial use of the GNT method, its distinctions and advantages as compared with the known ones stem from the properties of magnetic bremsstrahlung (MB) such as a great integral intensity raising as the third power of electron energy, a high spectral and spatial densities, a specific shape of the spectrum falling exponentially in the high-energy range with the relatively low average energy of γ-quanta.

These properties provide the great spatial and spectral densities of RW transmutation and photoneutron production.

The opportunity to use of MB for transmuting FR follows from the fact that long-lived FR, such as Sr-90 and Cs-137, are neutron-rich to be on the verge of nuclear stability and to have the cross-section of neutron capture near to zero. In the MB gamma field they loss the excess neutrons through the (γ, xn) reactions and transform into a stable state either directly during the above reactions or via the β-decay of short-lived daughter nuclei.

Photoneutrons produced in this process could be used to transmute by the reaction (*n*, γ) : (a) FR with a noticeable cross-section of neutron capture, such as Tc-99, J-129 and Cs-135; (b) even-even and odd- odd AR, such as Np-237, Pu-238 and Pu-240, to isotopes fissionable by thermal neutrons.

It is desirable that Pu-242 should be also transmuted in the MB gamma field so that the chain of production and accumulation of minor actinides (Am, Cm) were broken.

To reach the maximum intensity and hence the maximum rate of transmutation it is essential that a "critical" energy of the MB spectrum be comparable with the (γ, *n*) threshold energies $E_{\gamma n}$ of nuclei transmuted. For medium-mass nuclei with A = 90-150 and for transuranium actinides they are equal to $E_{\gamma n}$ = 6 - 12 MeV. Therefore, the electron energies required are E = 50-100 GeV at magnetic fields on the orbit H = 2-6 T. Under such conditions, the spectral density of generated gamma radiation in the energy range of a giant dipole resonance in the cross-section of the (γ, *xn*) reactions becomes sufficient to ensure a value of the transmuting rate suitable for industrial use.

II. CHARACTERISTICS OF THE METHOD

The spectral density of gamma transmutation in the MB field with allowance in a first approximation for the contributions from the Compton scattering of γ-quanta and the bremsstruhlung of Compton electrons and electron-positron pairs is described by the expressions:

$$N_{tr}\left(E_\gamma\right) = N_\Sigma \frac{\Sigma_x \sigma_{\gamma,xn}\left(E_\gamma\right)}{\sigma_\gamma\left(E_\gamma\right)}\left(1 - e^{-\Sigma_\gamma\left(E_\gamma\right)l}\right)\Phi_\gamma\left(E_\gamma\right)$$

where

$$\Phi_\gamma(E_\gamma) = \Phi_{\gamma 0}(E_\gamma) + \sum_{i=1}^{3}\Phi_{\gamma i}(E_\gamma)$$

$$\Phi_{\gamma 0}(E_\gamma) = k_0 \eta_\gamma(E_\gamma / E_c)\frac{1}{E_\gamma}$$

N_Σ is the MB integral intensity; $\sigma_\gamma(E_\gamma)$ and $\sigma_{\gamma,xn}(E_\gamma)$ are the total cross-section and the cross-section of the (γ, *xn*) reactions for γ-quanta of an energy E_γ, respectively; $\Phi_{\gamma 0}(E_\gamma)$ is the spectrum of MB γ-quanta normalized to unity; $\Phi_{\gamma i}(E_\gamma)$ are the spectra of γ-quanta for the above processes; $\eta_\gamma(E_\gamma / E_c)$ is the MB spectral function; $E_c \propto E^2 H$ is the so-called "critical" energy determining the position of the maximum in the MB spectrum; k_0 is the normalization factor; l is the target dimension along the MB beam, $l >> 1/\Sigma_\gamma(E_{\gamma n})$. The spectral density of photoneutrons emitted in the gamma transmuting processes can be determined from the expressions in [2].

The spectral density of gamma transmutation and photoneutrons produced and, hence, their integral intensity are

determined by the (γ, *n*) reaction threshold, the shape of the giant resonance of the reaction (γ, *xn*) and the value of the function $\eta_\gamma(E_\gamma / E_c)$ (see [2]).

The rate of gamma transmutation, the spectra of photoneutrons emitted hereby, the integral intensity of neutron production and other parameters of the GNT method were calculated by numerical integration for 40 energy groups in the range $E_{\gamma n} \le E_\gamma < 32$ MeV for a typical FR Sr-90 and for AR such as Pu-240 or Pu-242. In view of the absence of any data on $\sigma_{\gamma,xn}$ for FR and AR the cross-sections for their nearest even-even analogs Sr-88 [3] and U-238 [4, 5] were used in the calculations. Allowance was made for both the contribution from the above secondary processes and the production of photoneutrons in a Be moderator surrounding from above and below a plane target of radionuclides transmuted. In the case of Be the data [6, 7] were used.

The parameters were estimated for MB formed by an electron beam with E = 100 GeV and a current of 0.1 A in a uniform magnetic field H = 2 T (E_C = 13.3 MeV). It is proposed that electron bunchs should turn in a certain cyclic accelerator (storage ring) assembled as a magnetic and accelerating sectioned structure. In each of the sections the MB would be formed by a bending magnet within an angle of 3.6°. The radiation losses of electrons would be about $5 \cdot 10^{-3}$ E to be compensated by afteracceleration within the following accelerating section. Under these conditions the MB integral intensity would be $N_\Sigma = 8.2 \cdot 10^{21}$ q/s within 2π-radian, the MB flux being $F_\Sigma = 3 \cdot 10^{17}$ q/cm^{2}s with a 50 m length of the radiation exit channel.

The spectra of the photoneutrons are shown in Fig. 1 and 2 for the cases of the plane model FR (Sr-90) and AR targets (U-238) and for the same targets surrounded by the Be moderator of neutrons. It is seen that in the last case the spectral yield of photoneutrons grows considerably owing to production in the Be moderator by gamma radiation scattered from the plane targets.

The calculated parameters are presented in Tables 1 and 2. The symbols in the tables denote as follows: N_{tr}^γ and N_n^γ are the intensities of gamma transmutation and photoneutron production, respectively, in the target in the field of incident MB gamma radiation; $N_{tr}^{\gamma'}$ and $N_n^{\gamma'}$ are the same in the field of scattered radiation; N_{tr} and N_n are the total intensities of the above processes with allowance for the contributions from $\Phi_{\gamma i}(E_\gamma)$ and the neutron multiplication in the Be moderator through both (γ, *xn*) and (*n*, 2*n*) reactions; Q_γ is the specific energy consumption per transmutation event. The productivity of the GNT method is defined by the sum of $N_{tr} + N_n$. It was made allowance for the contribution from the (*n*, 2*n*) reaction approximately and only for Be.

The results were compared with those determined from the data on amounts of various FR and AR in spent fuel from NPP reactors (see, for example, [8]) and summarized in Table 3. The symbols in Table 3 denote as follows: $T_{1/2}$ is the half-life of the i-th radionuclide; q^i is its yield in *g* per GW × day of thermal energy output; N_r^i is the rate of build-up of this radionuclide per GW of thermal power; Q_f^i is the specific

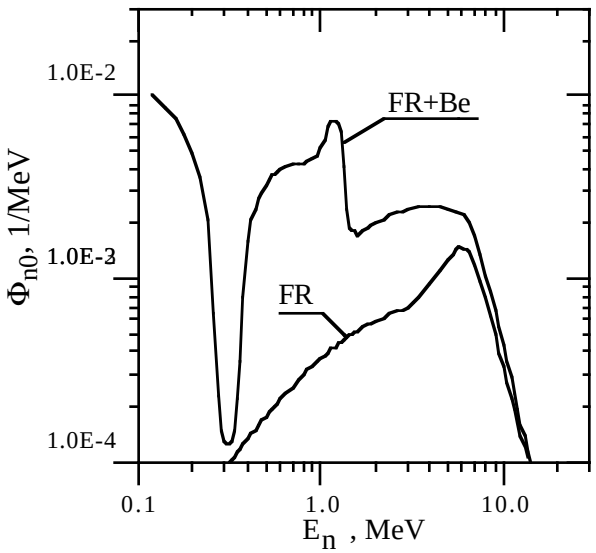

Fig. 1. The photoneutron spectra Φ_{n0} (E_n) from the model FR target (Sr) without a moderator and in Be moderator enveloping it.

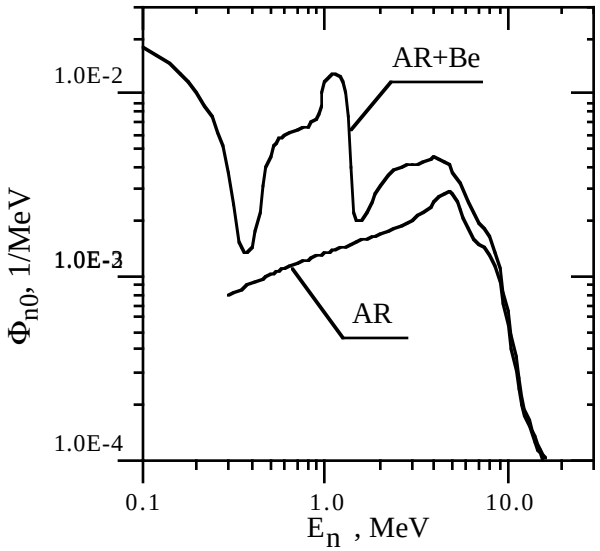

Fig. 2. The photoneutron spectra Φ_{n0} (E_n) from the model AR target (U-238) without a moderator and in Be moderator enveloping it

energy output per nucleus of the i-th radionuclide; $< Q_f^i >_i$ is the same averaged over FR or AR. The total rate of RW build-up amounts to about $1{,}8 \cdot 10^{19}$ nucl./GW ·s.

The comparison of the data presented shows that the GNT method allows the transmuting rate to be made essentially higher than the accumulation rate of any radionuclide in any operating NPP reactor: for RW with a maximum yield, such as Sr - 90 and Cs-137 by a factor of 4-5 and such as Pu - 240 and Pu - 242 by a factor of 10-15, for other RW - more than 30 times.

Table 1. The GNT parameters expected for FR-Be systems.

Parameter	FR	Be	FR + Be
N_{tr}^{γ} , 1/s	$1.7\cdot 10^{19}$	–	$1.7\cdot 10^{19}$
N_{n}^{γ} , n/s	$2.0\cdot 10^{19}$	–	$2.0\cdot 10^{19}$
$N_{tr}^{\gamma'}$, 1/s	$1.8\cdot 10^{18}$	–	$1.8\cdot 10^{18}$
$N_{n}^{\gamma'}$, n/s	$2.0\cdot 10^{18}$	$3.6\cdot 10^{19}$	$3.8\cdot 10^{19}$
N_{tr} , 1/s	$1.9\cdot 10^{19}$	–	$1.9\cdot 10^{19}$
N_{n}^{2n} , n/s	to be determ.	$4.3\cdot 10^{19}$	$4.3\cdot 10^{19}$
N_{n} , n/s	$2.2\cdot 10^{19}$	$3.6\cdot 10^{19}$	$5.8\cdot 10^{19}$
$N_{tr}+N_{n}=12.0\cdot 10^{19}$ 1/s; $Q_{\gamma}=270$ MeV/nucl.			

Table 2. The GNT parameters expected for AR-Be systems.

Parameter	AR	Be	AR + Be
N_{tr}^{γ} , 1/s	$1.8\cdot 10^{19}$	–	$1.8\cdot 10^{19}$
N_{n}^{γ} , n/s	$3.3\cdot 10^{19}$	–	$3.3\cdot 10^{19}$
$N_{tr}^{\gamma'}$, 1/s	$4.2\cdot 10^{18}$	–	$4.2\cdot 10^{18}$
$N_{n}^{\gamma'}$, n/s	$7.0\cdot 10^{18}$	$4.1\cdot 10^{19}$	$4.8\cdot 10^{19}$
N_{tr} , 1/s	$2.2\cdot 10^{19}$	–	$2.2\cdot 10^{19}$
N_{n}^{2n} , n/s	to be determ.	$5.2\cdot 10^{19}$	$5.2\cdot 10^{19}$
N_{n}^{f} , n/s	to be determ.	–	–
N_{n} , n/s	$4.0\cdot 10^{19}$	$4.1\cdot 10^{19}$	$8.1\cdot 10^{19}$
$N_{tr}+N_{n}=15.5\cdot 10^{19}$ 1/s; $Q_{\gamma}=210$ MeV/nucl.			

The energetic value of a transmuting event can be estimated from the expression:

$$Q_{\gamma}=\frac{N_{\Sigma}<E_{\gamma}>}{N_{tr}+N_{n}/\left(1-k_{eff}\right)}$$

where $<E_{\gamma}>=0.3\ E_C$ is the average energy of the MB spectrum and k_{eff} is the multiplication factor of neutrons in a blanket (for that of FR + Be $k_{eff}=0$).

Under the above conditions and for a nonmultiplicating matter of the blanket Q_{γ}amounts to about 210 MeV per AR nucleus and 270 MeV per FR nucleus. It can be somewhat minimized at the expense of a more optimal choice of the parameter E^2H for each of radionuclides to be transmuted as well as the more exact allowance for the processes of multiple scattering of γ-quanta and the $(n, 2n)$ reaction in the Be moderator. The contributions from processes $(n, 2n)$ in FR targets and $(n, 2n)$, (n, f) in AR targets should be included also.

As follows from the data of Table 3, the specific energy output averaged over RW with allowance for their cumulative yield amounts to about 5700 MeV per AR nucleus and about 4600 MeV per FR nucleus.

If we assume that the efficiency of energy transformation in designed NPP reactors and storage rings were raised in the future up to k_{NPP} = 0.38 and k_{ACC} = 0.65, respectively, we would obtain a ratio of the specific energy consumption for transmutation to the specific energy output $Q_{\gamma}/k_{NPP}k_{ACC}<Q_f>$ = 0.15 and 0.24 in the case of AR and FR, respectively. This means that 15 - 25 % of electricity generated by an NPP unit should spent to transmute any RW produced by this unit.

For a multiplicating blanket whose composition is similar to the AR - FR percentage in spent fuel it is not difficult to ensure the multiplication factor k_{eff} = 0.8 - 0.95 for thermal neutrons (with the system remaning conservatively subcritical). In consequence the values of Q_{γ} would be reduced considerably (by a factor of 5-20). This means that the above estimate for the NPP unit power spent to transmute RW produced by the unit itself should be considered as an upper limit of its possible values. In this case the productivity of the method $N_{tr}+N_{n}$ would grow much more than the total rate of build-up of FR and AR (see Table 3). Therefore the opportunity would arise to realize the breeding of fissile fuel and thereby to compensate at least in part for the energy consumption for transmutation.

III. CONCLUSION

The data presented demonstrate that the method proposed for gamma neutron transmutation of RW allows: (a) any long-lived radionuclide both AR and FR to be transmuted, with FR transmuted to a stable form; (b) the rate of transmutation to be made much greater than that of RW build-up in any operating NPP reactor; (c) the energetic value of a transmuting event to be made economically attractive; (d) the partial compensation of the energy consumption for transmutation to be obtained at the expense of the breeding of fissionable isotopes in the flux of neutrons produced.

One can expect that this method would be cleanest as compared with the known ones, because the fission is not here a basic nuclear reaction.

In realizing the method, the challenges are: (a) development and creation of a 50-100 GeV cyclic electron accelerator (a storage ring) with an efficient system of multiple afteracceleration to compensate for the beam radiation losses; (b) development and creation of RF systems capable of generating and transfering dozens MW of RF power to the beam with an efficiency of at least 60-65%; (c) development of methods and creation of facilities for the ejection of powerful MB beams from the accelerator vacuum chamber onto RW targets of great activity.

The problems concerning chemical extraction and separation of RW, radiation protection and safety of these processes which are also very important are complicated to solve. Nevertheless, all they must be overcome, if we wish to create a high technology for the closed nuclear fuel cycle ("wasteless nuclear technology") and thereby to provide the safe and ecologically clean future.

Table 3. The characteristics of the RW build-up process determined for the NPP reactors of WWER-440, WWER-1000 and RBMK

Fission radionuclides (FR)

Parameter	Sr-90	Zr-93	Tc-99	Pd-107	I-129	Cs-135	Cs-137
$T_{1/2}$, years	29.0	$9.5\cdot 10^5$	$2.1\cdot 10^5$	$6.5\cdot 10^6$	$1.6\cdot 10^7$	$2.3\cdot 10^7$	30.1
Transmut. type	γ	γ	*n*	γ	*n*	*n*	γ
q^i , *g*/GW· day	17.3	20.8*	26.6*	8.35*	5.29	40.1	37.4
N_r^i , 10^{18} nucl./GW· s	1.33	1.56	1.86	0.544	0.286	2.07	1.90
Q_f^i , Mev/nucl.	4700	4000	3350	11500	21900	3000	3300

Total rate of FR build-up per GW thermal power: $\sum_{i=1}^{7} N_r^i = 9.55\cdot 10^{18}$ nucl./GW· s

Energy output per FR nucleus averaged over FR: $< Q_f^i >_i^{FR} = 4580$ MeV/nucl.

Actinide radionuclides (AR)

Parameter	Np-237	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241
$T_{1/2}$, years	$2.6\cdot 10^6$	87.8	24380	6537	14.54	$3.9\cdot 10^5$	433
Transmut. type	*n*	*n*	*n*	*n*(γ)	*n*	γ(*n*)	γ(*n*)
q^i , *g*/GW· day	9.31	2.54	160.6	73.3	35.8	14.8	3.20
N_r^i , 10^{18} nucl./GW· s	0.274	0.074	4.69	2.13	1.04	0.426	0.092
Q_f^i , Mev/nucl.	22800	84400	1350	2950	6050	14700	67800

Total rate of AR build-up per GW thermal power: $\sum_{i=1}^{8} N_r^i = 8.75\cdot 10^{18}$ nucl./GW· s

Energy output per AR nucleus averaged over AR: $< Q_f^i >_i^{AR} = 5720$ MeV/nucl.

* The interpolation on Sr-90 and Cs-137.

REFERENCES

[1] I.P. Eremeev. JETP Lett., Vol. 27, No 1, p. 10, 1978.

[2] I.P. Eremeev. Paper to be presented to this Conference.

[3] A. Lepretre , H. Beil, R. Bergere et.al. Nucl. Phys., Vol. A 175, p. 609, 1971.

[4] J.T. Caldwell, E.J. Dowdy, B.L. Berman, R. A.Alvarez, P. Meyer. Phys. Rev., Vol. C 21, p. 1215, 1980.

[5] A. Veyssiere, H. Beil, R. Bergere, P. Carlos, A.Lepretre, K.Kernbach. Nucl. Phys., Vol. A 199, p. 45, 1973.

[6] B.L. Berman, R.L. Van Hemert, C.D. Bowman. Phys. Rev., Vol. 163, p. 958, 1967.

[7] R.J. Hughes et al. Proc. Int. Conf. Photonuclear Reactions and Applications, p. 151. Asilomar, 1973.

[8] S.N. Begichev, A.A. Borovoi, E.V. Burlakov et. al. Preprint IAE- 5268/3. Moscow, 1990.