PRODUCTION OF RADIOISOTOPES WITHIN A PLASMA FOCUS DEVICE

by

Ergisto ANGELI[1], Agostino TARTARI[1], Michele FRIGNANI[2], Vincenzo MOLINARI[2], Domiziano MOSTACCI[2], Federico ROCCHI[2], and Marco SUMINI[2]

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In recent years, research conducted in the US and in Italy has demonstrated production of radioisotopes in plasma focus devices, and particularly, on what could be termed "endogenous" production, to wit, production within the plasma itself, as opposed to irradiation of targets. This technique relies on the formation of localized small plasma zones characterized by very high densities and fairly high temperatures. The conditions prevailing in these zones lead to high nuclear reaction rates, as pointed out in previous work by several authors [1, 2, 3, 4]. Further investigation of the cross sections involved has proven necessary to model the phenomena involved. In this paper, the present status of research in this field is reviewed, both with regards to cross section models and to experimental production of radioisotopes. Possible outcomes and further development are discussed.

Key words: plasma focus, radioisotopes, cross-sections

INTRODUCTION

In recent years, the request for radioisotopes has been increasing steadily. In particular, short-lived radioisotopes (SLRs) for nuclear medicine have been in ever greater demand. In contrast with this growing need, production facilities have not increased quite as fast. This is due, largely, to the high cost of cyclotrons, the sole present-day commercial source of short-lived isotopes. Cyclotrons are the state-of-the-art in radioisotope production, and do an excellent job: however, they are very sophisticated and rather expensive. Expensive to buy, in the first place, and very demanding in shielding, requiring a large concrete bunker; a cyclotron facility can generally be afforded only by large hospitals. It is also quite expensive to run, as it requires a number of specialized personnel to operate it, and demanding maintenance. This said, if you can afford one, a cyclotron is an excellent way to produce radioisotopes.

Another side to the issue is peculiar to short-lived isotopes: due to their fast decay they cannot travel far lest the majority of the product is lost. Fluorine-18, for instance, with a half time of less than 2 hours, can hardly travel longer than an hour or two.

The third point to stress is that, albeit few hospitals can afford a cyclotron, many more can set up a nuclear medicine service, and the exams offered by such services are requested ever more often.

Summing up the problem, only hospitals within easy reach of a cyclotron facility can offer the type of services mentioned above, and still they saturate the capability of the existing facilities.

What can be asked of science and technology is to devise a novel approach to the production of radioisotopes, that could make them available to a larger class of medium hospitals thereby making it possible to increase significantly the number of patients examined.

In recent years, a research group in the USA [5, 6, 7] tried successfully to demonstrate the use of plasma focus (PF), to produce several SLRs with

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Authors' addresses:
[1] INFM-FE and Physics Department,
University of Ferrara, 12, Via Paradiso
44100 Ferrara, Italy
[2] INFM-BO and Laboratorio di Montecuccolino,
University of Bologna, 16, Via dei Colli
40136 Bologna, Italy

E-mail address of corresponding author:
domiziano.mostacci@mail.ing.unibo.it (D. Mostacci)

relatively high yields per discharge. An ongoing joint University of Ferrara and University of Bologna experimental campaign (*henceforth referred to as Bologna/Ferrara*) is being conducted, focusing on the production of a few positron-emitting radioisotopes through (d, n) reactions. The results obtained so far are within the same order of magnitude as those obtained by the Brzosko group. In the following sections the present scenario will be reviewed briefly and the results of the experimental campaign presented.

THE PLASMA FOCUS AS A RADIOISOTOPE BREEDER

The PF is a device that can generate, accelerate and pinch a plasma [8]. A "Mather-type" PF is composed of two coaxial cylindrical electrodes, closed and electrically insulated at one end and open at the other. The electrode assembly is contained in a vacuum chamber filled with an appropriate mixture of gases. Pressure ranges typically from 2 to 10 Torr (1 Torr = 133.322 Pa). The electrodes are connected by means of a fast switch to a capacitor bank, charged to tens of kV. A pulse is produced when the fast switch closes and the energy E_C stored in the capacitor bank is released to the electrodes in microseconds, producing currents of hundreds of kiloampere and more. The electrical discharge produced forms between the electrodes a thin sheath of plasma that evolves axially along the electrode assembly under the effect of the J B force. The plasma collapses at the open end where it is pinched by the electromagnetic fields: the pinch has duration of a few tens of nanoseconds. In the view of many authors, a number of localized small plasma zones, generally termed "hot spots", "plasma domains of enhanced nuclear reactivity" or "plasmoids", often form within the pinch volume; these zones are typically characterized by very high densities (strongly above the average levels in the pinch region and even above solid state values), fairly high temperatures (above 3 keV), small dimensions (20 to 300 microns), and magnetic fields sufficiently strong to trap ions with energies of the order of 5 MeV/nucleon. The time-stability of these domains is considered to be from 0.5 to 5 ns. If the reagents involved are captured within the plasmoids the density and temperature conditions inside these regions produce high nuclear reaction rates [1, 2, 3, 4]. The main advantage of producing SLRs within the plasma ("endogenous" production) are the significantly higher reaction rates that can be obtained within the plasmoids. Breeding is obtained filling the PF vacuum chamber with a mixture of a low atomic number gas (LZ) and one or more higher Z (HZ) gases.

EXPERIMENTAL RESULTS

The results obtained so far by the US group can be summarized in fig. 1, where they are confronted with the results of analytical calculations: the error bars refer to a 95% confidence level.

To obtain the analytical results of fig. 1, knowledge of the reaction cross sections is needed. Cross sections for some of the reactions tested by the US group are, to date, at best very approximate and at worst entirely unknown, either as experimental data or calculated values. Therefore the Bologna/Ferrara group has resorted to the EMPIRE II – Mondovì v. 2.18 code suite [9] as a tool for the re-evaluation of the cross sections of interest. Empire II is a modular system of nuclear reaction codes, comprising various nuclear models, and designed for calculations over a broad range of energies and particles. It accounts for the major nuclear reactions mechanisms (optical model, coupled channels, distorted-wave Born approximation, multi-step direct, multi-step compound, exciton model, Hauser-Feshbach *etc.*) and is very user-friendly, with a graphical interface based on Tcl/Tk libraries. In fig. 2 a comparison between the EMPIRE II calculation and the experimental data availabe for the ^{14}N(d, n)^{15}O reaction is shown as

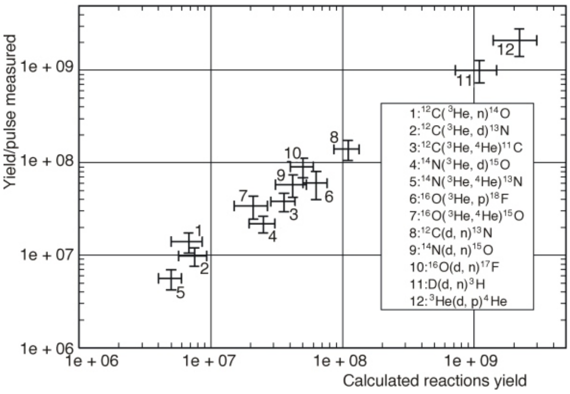

Figure 1. Experimental results and calculated yields (1e + 09 read as 10^9)

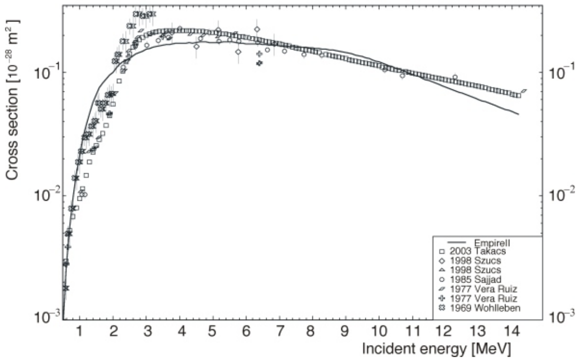

Figure 2. Computed cross section and experimental data points for ^{14}N(d, n)^{15}O [10, 11, 12, 13, 14]

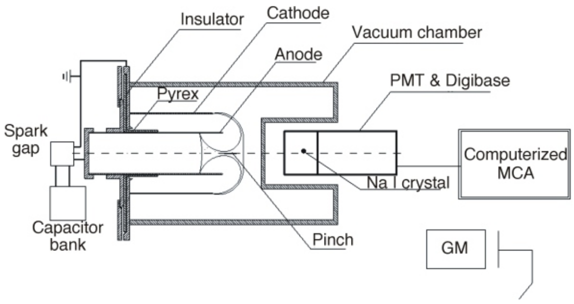

Figure 3. Schematic diagram of the experimental setup with a section of the PF vacuum

part of the preliminary results of this reevaluation work.

The Bologna/Ferrara experimental work was performed on the Ferrara University PF1 shown in fig. 3, whose technical features are summarized in tab. 1. The apparatus is similar in energy and geometry to that used by the US group; however, the measuring technique is different: the 511 keV annihilation photons were measured with gamma spectrometers located outside the vacuum chamber, whereas in the US experiments GM detectors were used to measure beta emission directly inside the chamber itself.

Table 1. Ferrara PF1 features

Total capacitance	48 F
Operating voltage	17 kV
Bank energy	7 kJ
Total inductance	30 nH
Electrode length L	13.3 cm
Inner electrode diameter	3.4 cm
Outer electrode diameter	10.7 cm
Pseudoperiod τ	9 s
Vacuum chamber volume	20 dm^3

The gas mixtures used in the Bologna/Ferrara experiments had D_2 as the LZ component and either N_2, O_2, and CO_2 or ambient air as the HZ component. The nuclear reactions involved were all of the type (d, n), with a positron emitter as product (see tab. 2).

Table 2. SLRs produced in the experiments described, with the reactions used for production, the half-lives and the decay daughters

Radioisotope	Reactions	$T_{1/2}$ [min]	Decay daughter
^{15}O	^{14}N(d, n)^{15}O	2.04	^{15}N
^{17}F	^{16}O(d, n)^{17}F	10.7	^{17}O
^{13}N	^{12}C(d, n)^{13}N	9.97	^{13}C

Gamma spectrometry was effected with a 3.28” thick, 3.22” diameter NaI(Tl) scintillator crystal, having a resolution of 7.5% at 511 keV; the scintillator phototube was coupled to an Ortec Digibase base & digital MCA, and the assembly was logged in a cavity so designed to place the detector as near as possible to the pinch region (fig. 3). Quantitative calibration of the gamma spectrometer was effected with a sealed 10 Ci (1 Ci = = 3,7 10^{10} Bq), ^{137}Cs reference source, located on the outer walls of the vacuum chamber, directly above the detector. The relative geometrical efficiency of the detector with the ^{137}Cs source and with the volume source was assessed with MCNP-5. An Ortec LO-AX coaxial HPGe detector with a 22.5 mm thick, 51 mm diameter crystal, having a resolution of 0.35% at 511 keV, was used independently to exclude the presence of unrelated nearby peaks. The neutron burst emitted by the PF has a time-spread (tens of nanoseconds) that is very short compared to the time-resolution of the available electronics: therefore, neutron counting was effected with a silver activation detector composed of four LND 72527 GM tubes wrapped in a thin silver foil, placed inside a paraffin block covered with a thin cadmium foil. The activation produced in the silver foil by the fast neutrons slowed down by the paraffin is recorded by a standard counting chain for 60 s. The cadmium around the paraffin block prevents ambient backscattering thermal neutrons from reaching the silver foil. To calibrate this neutron counting device a set of 8 high-sensitivity superheated emulsion detectors was used. A Rogowski coil was used to register the total current on a 1 GHz oscilloscope.

Figure 4 shows the gamma spectrum of a discharge in a mixture of about 7 Torr of D_2 and 0.5 Torr of N_2. The counting live time was 244 s, which corresponds to about 2 half-lives of ^{15}O. The peak is exactly at 511 keV.

The activity of ^{15}O measured in the discharge was 0.7 0.15 μCi, corresponding roughly to the breeding of approx. N = 4.7·10^6 nuclei of this radioisotope. In the same discharge, measurement of neutrons produced found a yield $Y_N = 6\ 10^8$, giving a ratio $Y_N / N = 127.7$. The ^{15}O breeding in this discharge, which can be considered typical, was one order of magnitude less than the average values published by Brzosko *et al.* (2001), as mentioned above. However, considering

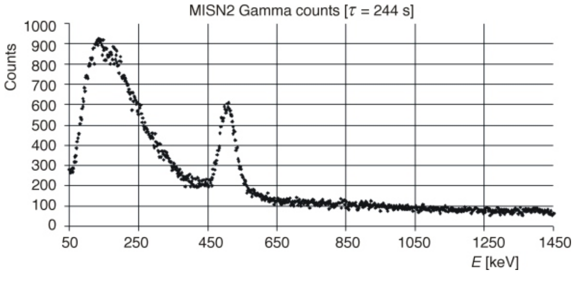

Figure 4. Photon spectrum (background not subtracted) for a typical discharge; the live time was 244 s

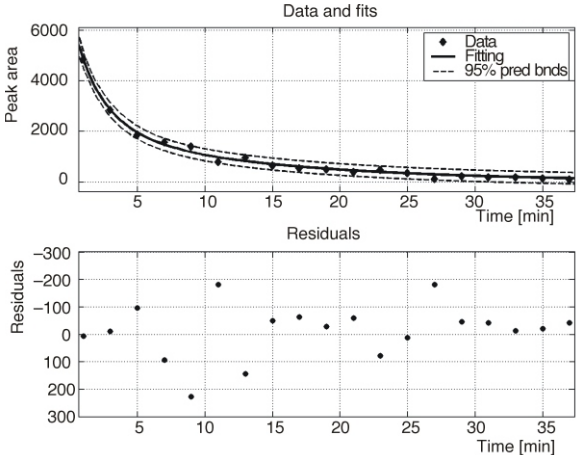

Figure 5. Upper plot – time decay of 511 keV peak (diamonds) and the Matlab fitting with 95% prediction confidence boundaries; Lower plot – scatter plot of the residuals of the fitting of the upper plot

that Ferrara PF1 is not a machine optimized for the production of SLR, the results obtained can be considered to agree with Brzosko's.

Figure 5 pertains to a discharge in a mixture of 8 Torr of D_2 + 0.5 Torr of air, with a total neutron yield of 1.7 10^8.

As expected, there are three isotopes simultaneously present: ^{15}O, ^{17}F, and ^{13}N, each decaying exponentially with its own half-life. The time decay of the 511 keV peak, due to the cumulative effect of the three isotopic species present, is shown in the top part of the figure, where the experimental data are shown together with the linear combination of the three decay exponentials as calculated with a best fitting standard Matlab package. The amplitudes a_i and the half-lives $T_{1/2}$ from the fitting are reported in tab. 3. The dashed lines in fig. 5 (upper part) are the 95% confidence prediction boundaries for the fitting; it can be seen how all the experimental points fall within these boundaries. The bottom part of the figure shows a scatter plot of the residuals of the fitting. The predicted half-lives agree very well with the known values pertaining to the three isotopes.

Table 3. Predicted half-lives and a_i coefficients, and confidence intervals for a multi-product discharge

Isotope	Predicted a_i	Predicted $T_{1/2}$ [min]	Known $T_{1/2}$ [min]	95% a_i confidence interval
^{15}O	2641	2.07	2.04	[1157, 4124]
^{17}F	2174	1.05	1.07	[286.1, 4062]
^{13}N	1959	9.9	9.96	[1722, 2197]

CONCLUSIONS

In concluding, activities of the order of 1 Ci of ^{15}O, ^{17}F, and ^{13}N were produced in a single discharge of a 7 kJ PF with the endogenous production method. These data seem to agree with those in literature; the fact that a different measurement technique was used confirms further the validity of the results. More research is needed and is in progress as far as the gas extraction from the vacuum chamber is concerned.

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Ергисто АЊЕЛИ, Агостино ТАРТАРИ, Микеле ФРИЊАНИ, Винћенцо МОЛИНАРИ, Домицијано МОСТАЋИ, Федерико РОКИ, Марко СУМИНИ

ПРОИЗВОДЊА РАДИОИЗОТОПА У УРЕЂАЈУ СА ФОКУСИРАНОМ ПЛАЗМОМ

Истраживања која се последњих година спроводе у САД и Италији, скренула су пажњу на производњу изотопа у уређајима са фокусираном плазмом, и нарочито, на оно што би се могло назвати ”ендогеном” производњом, то јест, производњом унутар саме плазме, насупрот озрачивања мета. Овај поступак заснива се на образовању локализованих малих зона плазме које карактеришу веома висока густина и прилично високе температуре. Као што је указано у ранијим радовима више аутора, услови који владају у овим зонама воде великим брзинама нуклеарних реакција. Даља истраживања укључених пресека за судар учврстила су потребу за моделовањем присутних феномена. У овом раду дат је преглед савремених истраживања у овој области, и са гледишта модела пресека за судар и са стране експерименталне производње радиоизотопа. Такође су размотрене могуће последице и даљи развој.

Кључне речи: плазма фокус, радиоизотопи, пресек за судар