Spallation Produced Radioisotopes for Nuclear Medical Application*)

G. J. Beyer

Academy of Sciences of the GDR, Central Institute of Nuclear Research Rossendorf, P.O. Box 19, DDR-8051 Dresden

A. F. Novgorodov, F. Roesch

Joint Institute for Nuclear Research Dubna, Head Post Office, P.O. Box 79, Moscow, USSR

H. L. Ravn

CERN EP/ISOLDE, CH-1211 Geneva 23, Switzerland

Spallation reaction opens new additional possibilities for production of a number of medical useful isotopes. In some cases all the present world demand is produced through spallation. Because of the high transmission of targetmaterials for medium or high energy protons lower cross-sections may be more than compensated by using massive targets. The unspecific nuclear reaction as well as the radiochemical processing of such massive targets lead to several difficulties. New ways for processing of large spallation targets are required. The possibilities of modern ISOL-techniques (ISOL = Isotope Separator On-Line) for the production of medical useful radio-isotopes will be discussed in some detail. Corresponding cross-section data are reviewed and discussed in relation to physico-chemical data requirements. In detail the following radio-isotopes will be discussed: 81-Rb, 82-Sr, 123-I, 167-Tm and 211-At.

Die Kernspaltungsreaktion eröffnet neue, zusätzliche Möglichkeiten der Herstellung einiger Isotope, die für die Medizin wertvoll sind. In einigen Fällen wird der gegenwärtige Weltbedarf durch die Kernspaltung gedeckt. Wegen der großen Durchlässigkeit des Targetmaterials für mittel- und höher-energetische Protonen werden die kleineren Wirkungsquerschnitte bei Verwendung von massiven Targets mehr als kompensiert. Die unspezifische Kernreaktion als auch die radiochemische Aufbereitung von solchen massiven Targets verursachen einige Schwierigkeiten. Es werden neue Wege zur Aufbereitung von größeren Kernspaltungstargets benötigt. Die Möglichkeiten der modernen ISOL-Technik (ISOL = Isotope Separator On-Line) zur Herstellung einiger medizinisch wertvoller Radioisotope werden im einzelnen behandelt. Die entsprechenden Daten der Wirkungsquerschnitte werden kritisch besprochen und im Zusammenhang mit den phsikalisch-chemischen Daten diskutiert. Im einzelnen werden die folgenden Radioisotope diskutiert: 81-Rb, 82-Sr, 123-I, 167-Tm und 211-At.

Keywords

1. Introduction

It is well known, that spallation reaction is the most convinient way to produce very neutron-deficient nuclei far from the line of beta-stability [1—3]. The use of intense beams of high energy protons has been also recognized and exploited for some time for the production of neutron deficient radioisotopes for practical application [4—11]. The main advantage of beams in the 0.5 to 1 GeV range lies at present in high product output through an ability to penetrate and induce nuclear reactions in target materials having thicknesses on the order of hundreds gramms per cm². The large number of target atoms can more than compensate for the generally low reaction cross sections (order of 10^{-30} m² or 10 mb) compared to the cross sections often associated with lower energy reactions used in more conventional ways for isotope production.

In interactions of high energy protons with heavy target elements three main typs of nuclear reactions accure: spallation, fission and fragmentation (see Fig. 1 taken from [16]). For crossection or yield calculations for the spallation still analytical methods basing on the work of *Rudstam* [12] are used. *Rudstam* observed, that the cross-section for isodiapheric nuclei grow with increasing mass-number or oposit decreases homogeneously with growing mass deficiet. Consequently he proposed an empirical exponential equation, which was than modified for special applications [13—15].

Cross-sections for spallation reaction products have been discussed at the IAEA Consultants Meeting in Vienne, April 13—15, 1981 [20]. In generall measurements of spallation cross-sections fall within a factor of approximately 2 of the value predicted by *Rudstam*'s empirical systematics [12] which seems to be sufficient for the moment. Optimization of target- and separation techniques are of similar importance. It is the aim of this paper to discuss the production of medical isotopes via spallation as a complex prob-
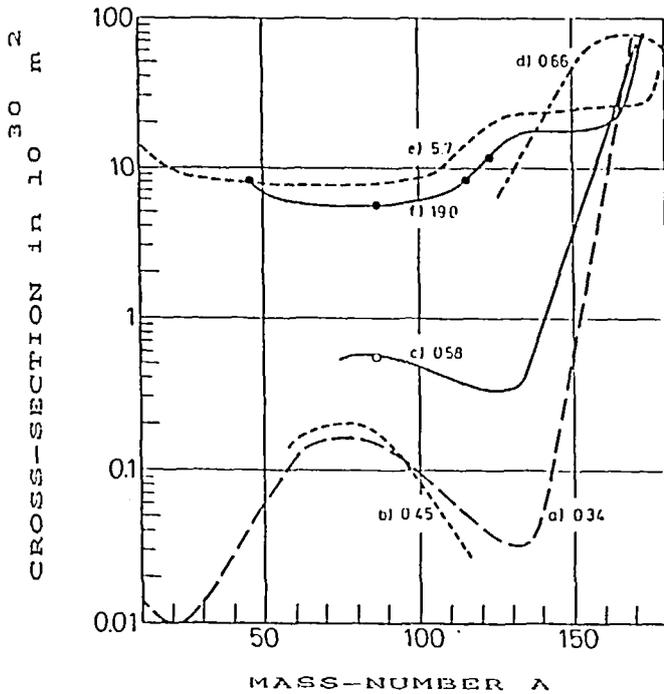
Fig. 1. Mass distribution for reaction products formed in interaction of Ta with protons of different energies (taken from [16])

lem of the nuclear reaction and chemical or physical principles of separation techniques.

The long range of the high energy protons permits the simultaneous irradiation of stacked thick targets as well as the use of a single massive target. The advantage is that several radioisotopes are available in both cases simultaneously. But this advatage is also an disadvantage because of the formation of isotopic impurities. This problem must be handled through optimized choices of irradiation and decay periods as demonstrated in the case of 123-I via 123-Xe [17, 18]. The plethora of waste and by-product radioelements to be dealt with are other complicating factors. Nevertheless practically all the present world supply of 82-Sr and 67-Cu and good fractions of 127-Xe, 109-Cd and 68-Ge are produced via spallation reaction [6, 7, 11, 19].

It is our suggestion, that new separation techniques could solve some of the problems and open possibilities to produce valuable spallation radioisotopes in a more sufficient way.

Today new rapid separation methods based on thermo-chemical and physical principles allow radioactive nuclei of allmost all elements to be contineously transferred from an accelerator irradiated target into an ion beam. These techniques often referred to as target ion-source system have in the last 20 years turned on-line mass separators into the most efficient tool for production and study not only of nuclei far from stability but radioactive nuclei in general [23]. The principle of such an ISOL-system is shown in Fig. 2. For more generall information see [1—3].

The production, transport and separation processes in such an integrated ISOL-system can be devided into the following main steps:

i.   Formation of the products in interaction of an intense proton beam of around 700 MeV
ii.   Diffusion of the product nuclei from the interior to the target surface
iii. Desorption from the surface
iv.   Transport of the product to an ion-source
v.   Ionization of the product
vi.   Mass-separation of the products.

For a complete description of the processes we need to know about the following data (others than nuclear data):

— Diffusion coefficients
— Adsorption enthalpies
— Ionization behaviour.

2.   High temperature diffusion

The solution of the second *Fick* law

$$\frac{dc}{dt} = D \cdot \frac{d^2c}{dx^2} \tag{1}$$

$$F(\%) = 100 - \frac{800}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \times$$

$$\times \exp\left[-\frac{(2n+1)^2\,\pi^2 Dt}{d^2}\right] \tag{2}$$
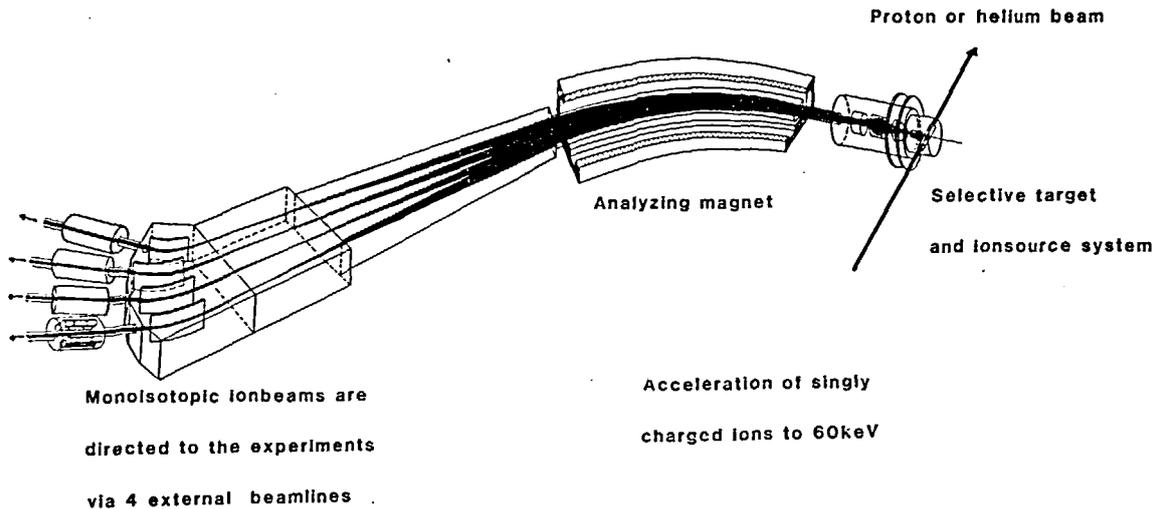
THE PRINCIPLE OF ON-LINE MASS SEPARATION
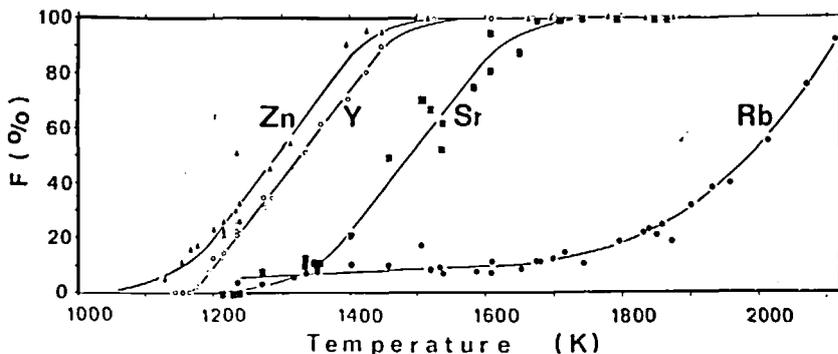
Proton or helium beam

Analyzing magnet

Selective target and Ionsource system

Acceleration of singly charged Ions to 60keV

Monoisotopic Ionbeams are directed to the experiments via 4 external beamlines

Fig. 2. The principle of on-line mass separation

describes the relation of fractional release (*F*) of a given nuclear reaction product, the diffusion coefficient (D) and the target-foil thickness (*d*) [24]. This equation (2) is applicable for the special sample conditions: foiles or plates where the foil-thickness d is small compared to the length and width (*d* ≫ *a* ≈ *b*) and with homogeneous distribution of the diffusing particles throughout the matrix. The experimental value *F* can be transformed into diffusion coefficients using the tables of *Zimen* [25]. The activation energy

E_A for the diffusion process can be obtained from an *Arrhenius* plot according equ. (3).

$$D = D_0 \exp \frac{E_A}{RT}, \qquad (3)$$

where R is the gas-constant.

Systematical studies have been performed in the following manner: target foils of refractory metals were labelled homogeneously with radio tracers by irradiating them with 660 MeV protons. Sampels of the irradiated foils were annealed in high vacuum at high temperatures. The samples were analyzed before and after heating by means of gamma-spectroscopy for determination of the fractional release of spallation reaction products. The results are published for the following target elements: Ti [26, 33], Zr [26, 27], Nb [28], Mo [29], Hf [30], Ta [31], W [32], Re [33], Ir [33], Th [33]. For illustration a few results are presented in Figs. 3—6.

The diffusion relase studies may be summarized in the following way:

i. In a given host metal of the IV B, V B or VI B group of the periodic table the diffusion coefficients for trace elements grow in the following sequence:

$D_{IA} < D_{IIA} < D_{IIIA}$.

For example Y diffuses faster than Sr, which is faster than Rb (see Fig. 3). The reason for this sequence is the decreasing radius of the diffusing particles.

ii. The diffusion coefficient for a given trace element decreases for different host metals in the following sequence:
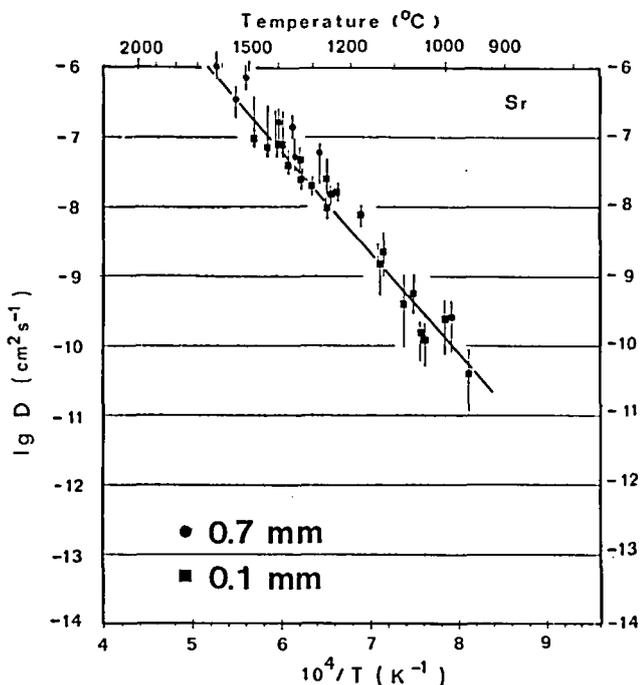
$D_{IVB} < D_{VB} < D_{VIB}$.

Fig. 4. *Arrhenius*-plot for the diffusion of Sr in polycristalline Zr [26]
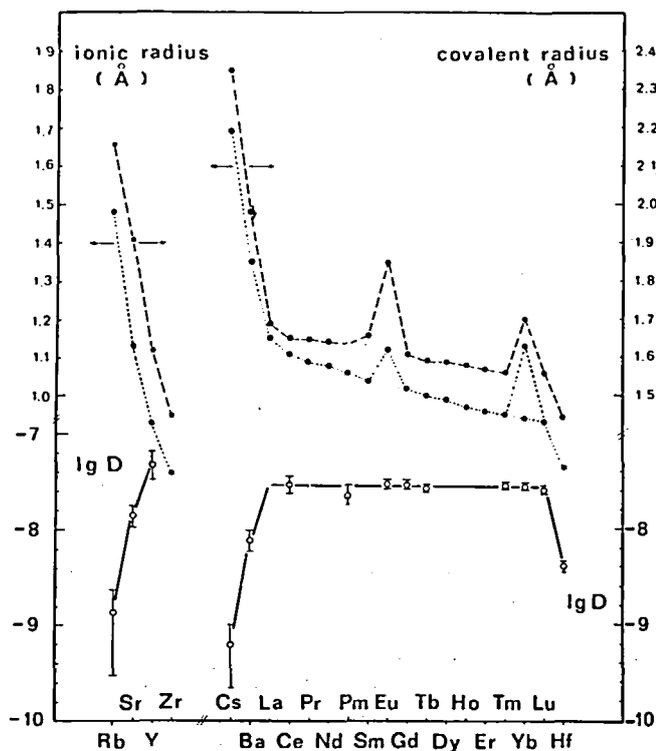
4
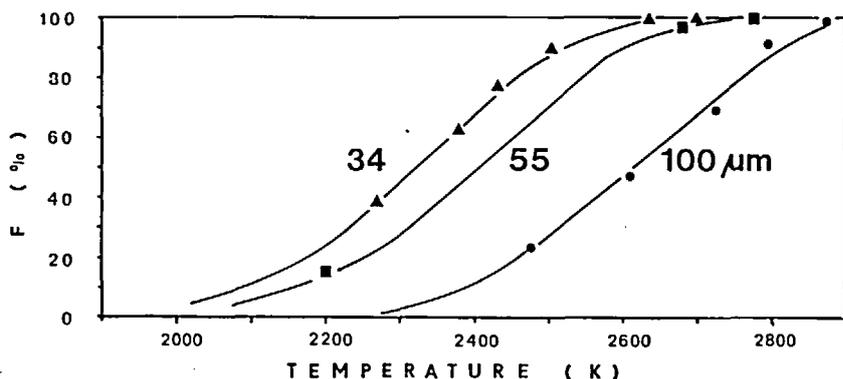
Fig. 6. Diffusion coefficients of spallation reaction products in poly-crystalline Ta (lower part) in comparison with the atomic and ionic radii of the diffusing particles (upper part). Ta-foils of 83 μm thickness were irradiated with 660 MeV protons. The diffusion coefficients were determined for the indicated elements simultaneously from release studies, the annealing temperature was 2750 K, annealing time 300 s. No significant difference in the diffusion behaviour of the rare earth elements has been observed, they all diffuse faster then Sr and Ba

For example Y diffuses fastest in Zr and in Nb faster than in Mo. Compare Figs. 3 and 5. The reason for this effect is the growing lattice density of the host metals.

iii. In a given host metal the elements of one group of the periodic table diffuse according to growing atomic masses: Sr diffuses faster than Ba in Ta (see Fig. 6).

iv. From the systematical study one can evaluate, that all rare earth elements diffuse in metal matrixes as M^{3+}-particles, the II A-elements as M^{2+}, the alkaline elments as M^+ and the nobel gases as neutrals (see Fig. 6 and Fig. 3).

In generall one can conclude that solid phase diffusion processes very often are fast in order to use refractory metals as high temperature targets. A large number of other refractory materials such as metaloxides, borides, carbides as well as molten metals and alloys has been investigated to develope suitable targetsystems for ISOLDE (review papers see [61, 60, 44]).

3. Surface effects

In our release studies we found that in Ta the diffusion of the rare earth elements is more rapid than of Ba followed by Cs. For the desorption from the surface the sequence is reverse with Cs being fastest followed by Ba, lanthanoides
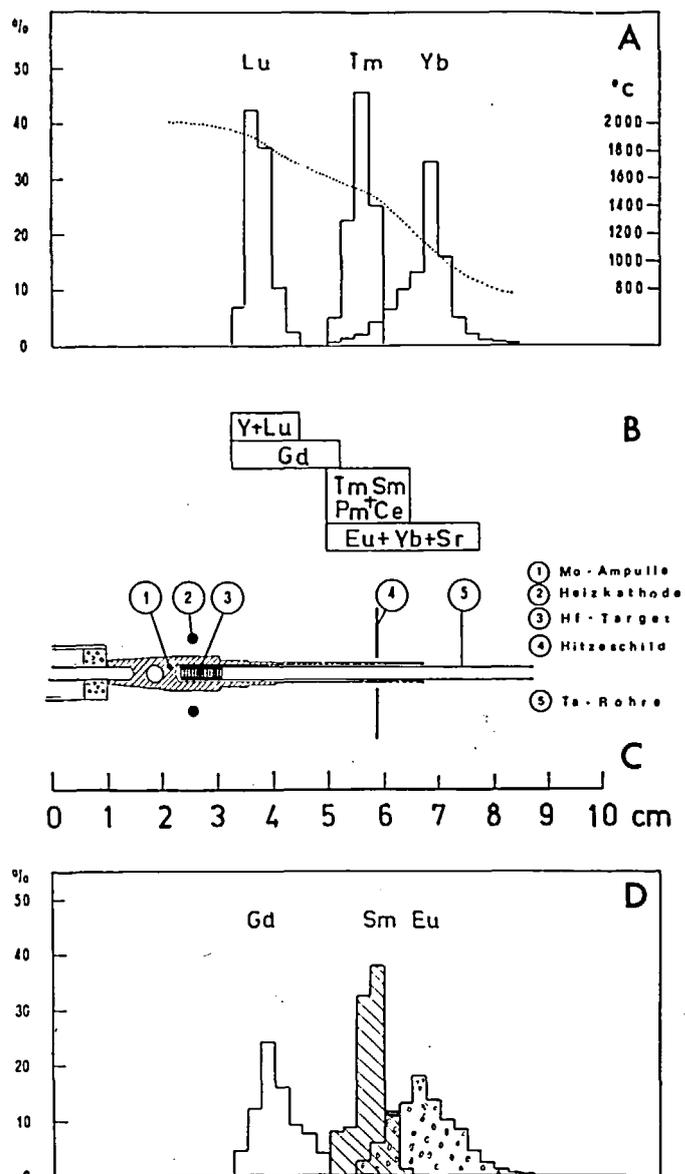

Fig. 7. Thermochromatographical separation of carrier free rare earth radio isotopes in vacuum. The rare earth radio nuclides are formed in spallation reaction irradiating a Hf-target (3) with 660 MeV protons. The Hf-target was inserted into Mo-crucible (1) carriing a thermochromatographic Ta-tube (5). The crucible containing the target was heated to 2170 K by electron bombardment (2). All rare earth radio isotopes are released and adsorped according theire adsorption energies along the Ta-tube. Everage annealing time was 10 min [30, 31]

and Hf. Thus in a Ta-powder target the rare earth elements will be delayed compared to Cs and Ba due to the large total surface. Consequently from a Ta-foil target with optimized foil thickness the rare earth elements will be released more rapidly because the surface is deminished. The surface adsorption behaviour or in other therms the adsorption enthalpies play an important role in the release of nuclear reaction products from a target as well as for the transport of the products in the gas phase.

Desorption studies for rare earth elements have been performed in a similar way as the release studies [34]. Fig. 7 illustrates the possibilities to use differences in the adsorption enthalpies for thermochromatographic separations.

A systematical study of the adsorption of trace metals at metallic surfaces hase recently been published by *Roesch*

and *Eichler* [35]. The wide variation of the surface material, the composition of the used carrier gases, variation of the gas preasure and temperature opens the wide range of separation techniques starting from gaschromatography over gasthermochromatography and thermochromatography to vacuum thermochromatography.

4. Ionization

A selective ionization is a modern radiochemical separation technique. For the production of radioisotopes via spallation the following ion-source principles are suitable to be operated in an integrated target ion-source system:

i. Positive surface ionization ion-source with Ta-ionizer kept at 1300 K for selective ionization of the alkaline elements K, Rb, Cs and Fr [36]. The ionization efficiency is of the order of 90%.

ii. Positive surface ionization ion-source with W-ionizer (or Re) at high temperatures (2700 ... 3000 K) for ionizing the lanthanoides, In, Ga, Sc, Al, Ca, Sr, Ba, Li, Na and Tl. The ionization efficiency varies from a few % up to 80% [36, 37, 38].

iii. Negative surface ionization ion-source with LaB_6-ionizer for selective ionization of Cl, Br, I and At. Ionization efficiencies of up to 40% have been obtained [36, 39].

iv. The plasma discharge ion-source with cooled line at 300 K and ionization temperatures around 1300 K is used for selective ionization of nobel gases. Efficiences archieved for He, Ar, Kr, Xe and Rn are 0.5%, 2%, 5%, 15%, 30% and 40% respectively [36]. A similar source with a cooled line kept at 700 K is used to ionize the volatile elements of the group IIB Zn, Cd and Hg with efficiences of 10 ... 60% [36].

v. A high temperature version of the plasma discharge ion-source (line at 220 K) is intended to ionize the less volatile elements from group IB, III, IV and V. This ion-source can be used in conjunction with CF_4 to form volatile fluorides of the mentioned elements [36].

5. Chemical selectivity

As mentioned the product mixture of a massive target irradiated with protons with E_p 0.5 GeV is rather complex. The difficulties to handle such targets and product mixtures can be compared with the fission product mixture in isotope production plants only. Nevertheless chemical selectivity can be obtained using the above mentioned techniques by one or several of the following means:

i. Selective release from the target (Sr from Zr [40]).

ii. Selective adsorption of unwanted species at suitable surfaces (selective release of Yb from Ta-powder [33]) or selective transport of the required product (^{52}Fe from Ni [41]).

iii. Selective ionization.

iv. Mass-separation of molecular side bands (iodine as AlI+-particles [36]). Isobaric separations using differences in the adsorption enthalpies [42]).

6. ISOL-technique for production of medical isotopes via spallation

In this chapter we like to illustrate the possibilities of the discussed techniques for the production of some of the most important medical radioisotopes via spallation route.

6.1. 81-Rb

Because of the short halflife there is no much sense to produce 81-Rb via spallation, but the isotope separator opens the way to produce a new type of 81-Rb-81m-Kr-generator. 81-Rb is best produced at ISOLDE using a Nb-powder target with yields of $3.8 \cdot 10^{10}$ atoms per second for 1 μA proton beam current (Figs. 8, 9 [44]). This production rate corresponds to 1.6 MBq/μAs (or 20 mCi in 7.7 min collection time for 1 μA proton beam current). By implanting the 81-Rb+-ions into mylar foils (or other plastic material like polyethylene etc.) an implantation type of 81-Rb-81m-Kr-generator is obtained. The 81m-Kr can be eluted nearly quantitatively by blowing air over the implanted Rb-source as well as by elution with isotonic saline solution suitable for direct infusion [43]. No 81m-Kr-elution will be achieved by using metallic foils as implantation backing. The 81-Rb break through is as low as in conventional 81-Rb-81m-Kr-generator systems. The implantation type generator could be realized of extremly small size since for an implantation density of 10^{14} atoms/cm² less then 0.5cm² foil area is required to make a 800 MBq (20 mCi) generator.

Tab. 1. Review of spallation rection cross-section data for the radio-isotopes discussed in this paper. The proton energy was limited to the 500 MeV region

Radionuclide	Target	E_p [MeV]	Cross-Section [10^{10} m² = mb]	Ref.
82-Sr	Zr	590	19 ± 4	53
	Nb	590	22 ± 4	53
	Mo	590	15 ± 3	53
	Mo	800	21.5 ± 0.8	56
	Mo	800	23 ± 1	46
	RbBr	800	2.1 ± 0.2	54
85-Sr	Mo	800	48 ± 0.7	46
	Mo	800	50 ± 2	56
123-Xe	La/Cu	590	36 ± 5	53
	La/Cu	590	45 ± 6	54
	I	660	3.8 ± 0.4	18
	Cs	660	29.3 ± 1.7	18
	Ba	660	26 ± 4	18
	La	660	31.9 ± 1.6	18
125-Xe	La/Cu	590	57 + 9	53
	La/Cu	590	43 ± 9	54
	I	660	6.6 ± 0.5	18
	Cs	660	45 ± 0.5	18
	Ba	660	38 ± 6	18
	La	660	47.7 ± 3.9	18
127-Xe	La/Cu	590	53 ± 11	53
	La/Cu	800	51 ± 7	56
123-I	La/Cu	590	57 ± 6	54
	La/Cu	590	57 ± 9	53
	La	800	51 ± 3	56
167-Tm	Lu	590	56 ± 6	58
	Hf	590	51 ± 10	58
	Ta	590	49 ± 7	58
	W	590	49 ± 7	58
168-Tm	Lu	590	5 ± 1	58
	Hf	590	6 ± 2	58
	Ta	590	0.8 ± 0.4	58
	W	590	0.4 ± 0.2	58
211-Rn	Th	660	13 ± 6	63
211-At	Th	660	18.2 ± 2.2	64
	U	660	5.2 ± 1.4	64

6

VALVE

TRANSFER
LINE

TARGET
CONTAINER
WITH
HEAT
SCREENS

TARGET

SURFACE
IONIZER

EXTRACTION
ELECTRODE

LINE AND
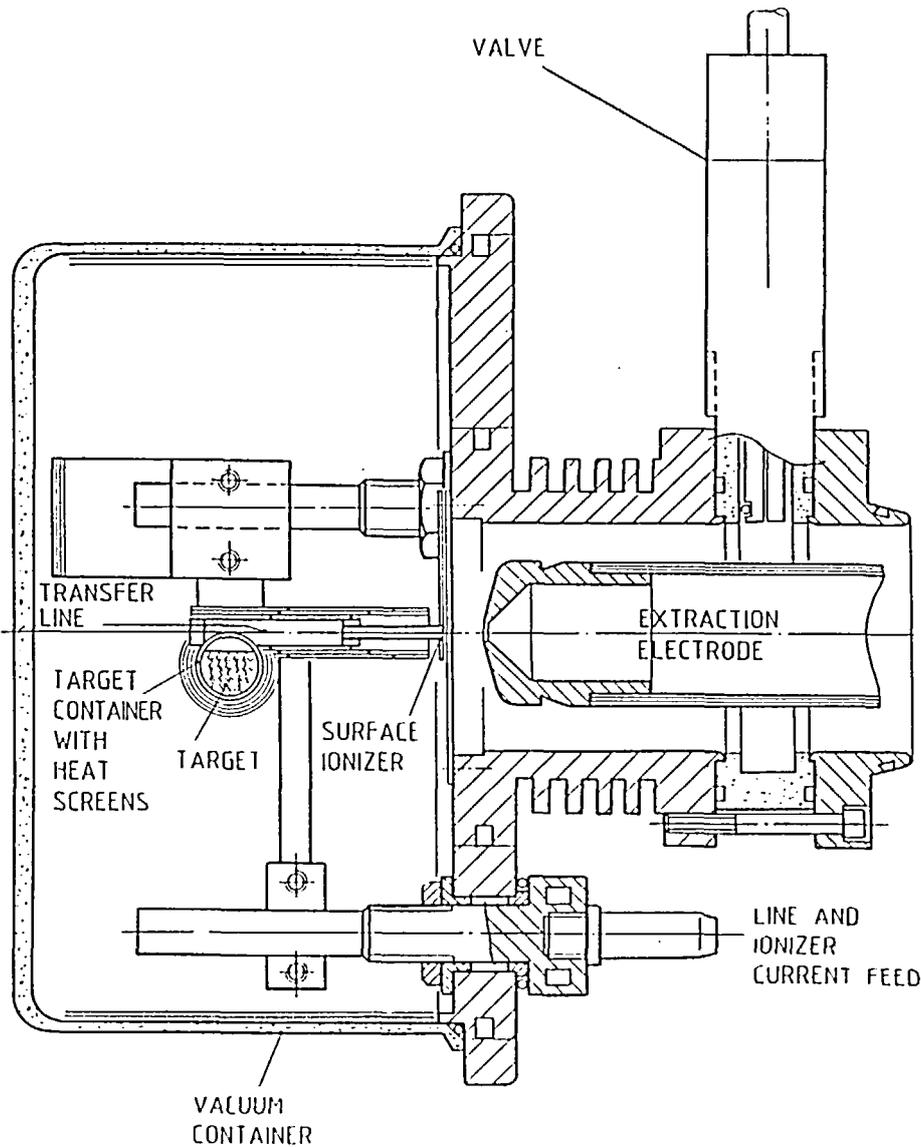IONIZER
CURRENT FEED

VACUUM
CONTAINER

Fig. 3. Scheme of target ion source system for the case of a surface ionization ion source. In addition the extraction electrode is shown fixed to the acceleration chamber. (taken from [44])

6.2. 82-Sr

Recently the complete problems about 82-Sr-82-Rb-generators have been published [45]. The cross-section for the formation of 82-Sr in Mo irradiated with 800 MeV protons is $(2.3 \pm 0.1) \cdot 10^{-30} \, m^2$ (23 mb) and for 85-Sr $(4.8 \pm 0.7) \cdot 10^{-30} \, m^2$ (43 mb) [46] (Tab. 1). The radiochemical procedure has been modified several times [47, 48, 48]. High dose rates of the irradiated targets (10^7 R/h in 1 cm distance [48]), large amounts of process solutions and unsatisfied purity of the final 82-Sr-preparation were the main problems in the wet radichemical process [48, 49]. The isotopic byproduct 85-Sr remains still a serious problem. At EOB an everage isotopic ratio 82/85-Sr of only 1.0 to 1.2 is obtained depending on irradiation time. During the clinical use of the generator the 85-Sr grows up to an excess of a few 100% compared to the 82-Sr activity. It was allready proposed to solve this problem by using an isotope separator [48].

We recommend to use additional ISOL-target technique. From a 50 g/cm² Zr-foil target (foil-thickness up to 0.5 mm) the Sr is released rather selective at 1750 K (Fig. 3). A high temperature surface ionization ion-source could give an ionization yield of better then 50%. At the collector of an isotope separator pure 82-Sr and 85-Sr could be collect-

ed. The cross-contamination for routine practical use is allmost less then 0.1% in this mass region. Recent studies at CERN demonstrated, that a Zr-foil target could withstand a well focused proton beam of 100 μA [50]. It is expected to obtain 10^{10} atoms 82-Sr per 1 μAs or 10^{10} Bq/1000 μAh (10 h, 100 μA). The advantage of this technology is first the high isotopic purity; secondly practically no liquid waste is produced. The disadvantage is that no other product as 77-Br or 88-Y are available simultaneously from the same target.

6.3. 123-I

Special interest was directed to the production of radioxenon via spallation [18, 20, 51]. It has been demonstrated, that by optimized choice of irradiation and decay periods ruther pure 123-I could be obtained. Also important was the possibility to produce 125-I and 127-Xe simultaneously. From a 175 g CsCl-target after 4 h irradiation and 4 h grow up period for 123-I via 123-Xe decay 850 mCi 123-I with 0.4% 125-I contamination is obtained [52]. The 127-Xe produced via spallation at TRIUMF from 0.8 cm thick CsCl-pellet contains 15% of 129m-Xe and 12% 131m-Xe [11]. Cross-section data are summerized in Tab. 1.
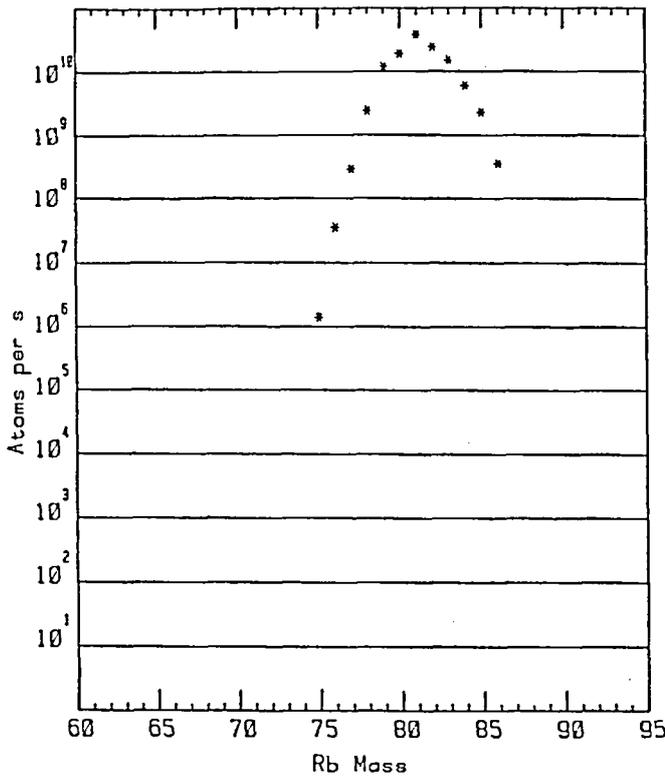
Rubidium (37-1)

Fig. 9. Yield curve for the production of Rb-isotopes at ISOLDE from a 50 g/cm² Nb-powder target taken from [44]
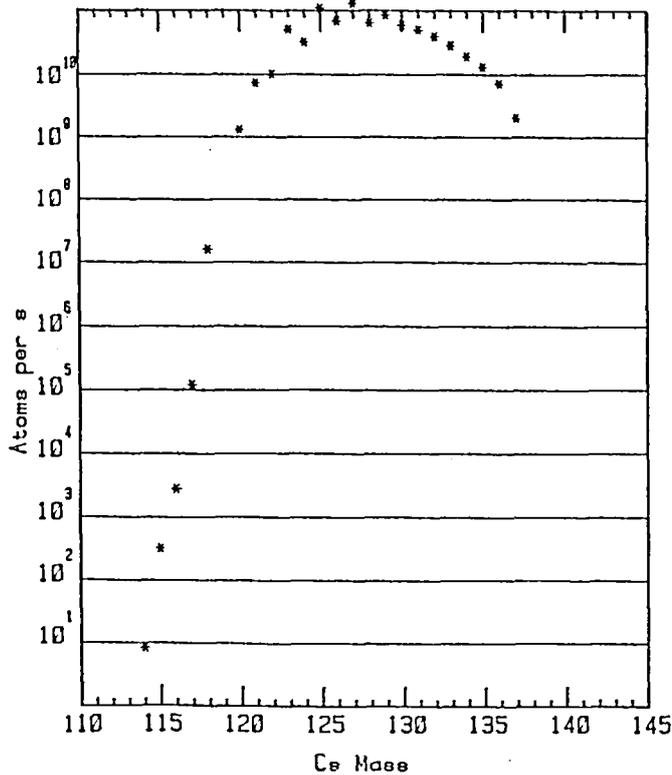
Caesium (55-1)

Fig. 10. Yield curve for the production of Cs-isotopes from a 140 g/cm² molten La target and a 1 μA 660 MeV proton beam in 1 s (taken from [44])

The 125-I contamination in spallation produced 123-I preparations presently is higher than those for the (p, 5n)-production route. One possibility to solve this problem is the application of ISOL-technique. From a molten La-target kept at 1770 K Cs is released fast and transported to a surface ionization ion source [57, 36]. This combination is highly selective for Cs. The total efficiency is of the order of 80 ... 90%. The Cs-yield courve is shown in Fig. 10 [44]. The yield as well as the activity of the corresponding products are as follows:

Cs-isotope	yield [atoms/μA]	product	yield [Bq/s]	[mCi/h]
123-Cs	$5.0 \cdot 10^{10}$	123-I	$7.3 \cdot 10^{8}$	71
125-Cs	$1.1 \cdot 10^{11}$	125-I	$1.4 \cdot 10^{4}$	1.5
127-Cs	$1.3 \cdot 10^{11}$	127-Xe	$2.9 \cdot 10^{4}$	2.8

Note that these activities are obtained for 1 μA proton beam intensity only. The molten La-target of the given design could withstand a beam current of 50 μA [50]. From this figures one can easy estimate: the system is able to produce multi-Ci quantities of 123-I and allmost Ci-quantities of 125-I and 127-Xe simultaneously within a few hours. The isotopic impurities are determined from the mass-resolution of the isotope separator. The 125-I contamination in the final 123-I preparation will be of the order of 10^{-3}%.

6.4. 167-Tm

The isotope 167-Tm is suitable for scintigraphic in vivo studies [59]. The best way to produce 167-Tm is via spallation reaction [58, 59]. The most elegant way would be the application of ISOL-technique. As allready mentioned a Ta-powder target give highly selective Yb-isotopes [60], the other rare earth elements are discriminated by surface effects. A Ta-foil target connected to the same ion-source produces isobars of the rare earth elements [33] (Fig. 11).
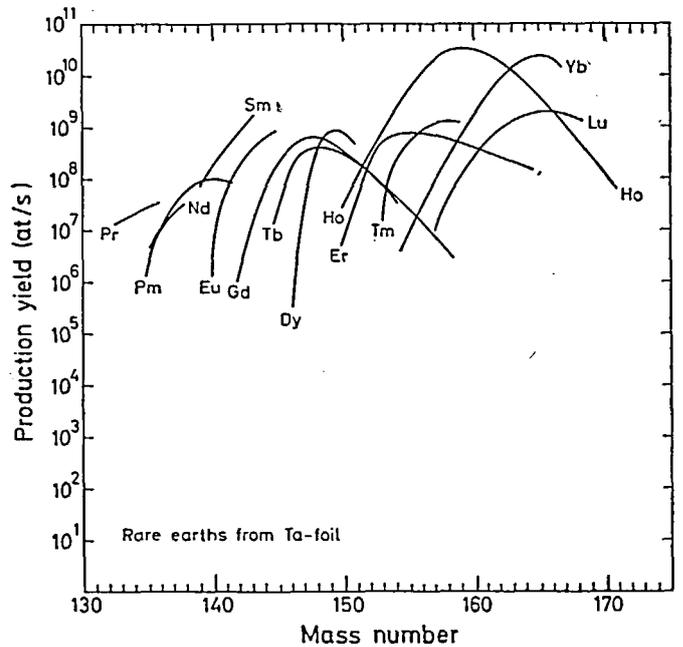

Fig. 11. Production yields of lanthanoid isotopes from a 122 g/cm² Ta-foil target connected to a positive surface ionizer. Individual measurement points are not indicated. (taken from [33])

The production rate for 167-Yb is $1.1 \cdot 10^{10}$ atoms/s and μA proton beam intensity. This gives a practical production yield for 167-Tm of approximately 1 mCi/μAh. The Ta-foil target of the given design could be irradiated with 100 μA proton beam current [50].

6.5. 211-At

In similarity to 123-I the medical important 211-At (radionuclide therapy) can be produced with satisfied isotopic purity via the 211-Rn [62]. The cross-sections for the formation of 211-Rn as well as for 211-At from uranium are significant smaller than for thorium as target [65] (Tab. 1). ISOL-technique can be used to produce 211-At via the mother-nuclide in a very elegant way. ThO_2 or ThC_2 targets [61] are connected to a plasma discharge ion source with cold line [36]. Practical yield of nearly 10^9 211-Rn atoms per second and μA proton beam intensity have been obtained [61, 36].

7. Conclusions

i. Spallation reaction is a very suitable route for production of some medical radioisotopes. Because of the limited number of corresponding accelerators as well as because of the priority of experiments at such facilities a regular supply of shortlived isotopes like 81-Rb or 123-I produced in this way seems to be unrealistical. For longlived isotopes however a few production runs per year will be sufficient to organize a regular supply.

ii. Stable impurities in the targetmaterial, isotopic impurities due to the lack of reaction selectivity, waste problems and others are the main complicating factors in radiochemical processing of large spallation targets. ISOL-techniques open new ways in spallation radioisotope production and application. Isotope separation technique itself is also useful to be applied in "classical" RI-production to improve the isotopical purity.

iii. Modern physico-chemical data are of same relevance as nuclear data in radioisotope production. By knowing data for release, surface adsorption and ionization ISOL-technique is a suitable tool to determine simultaneously cross-sections.

References

[1] *P. G. Hansen, O. B. Nielsen* (eds.), Proc. 4th Int. Conf. on Nuclei far from Stability, 1981 (CERN 81-09, Geneva, 1981)

[2] *H. L. Ravn, E. Kugler, S. Sundell* (eds.), Proc. 10th Intern. Conf. on Electromagnetic Isotope Separators and Techniques Related to their Application EMIS-10, Zinal, 1980, Nucl. Instrum. Methods 186 (1981) (special issue)

[3] *J. Crawford, J. M. D'Auria* (eds.), Proc. TRIUMF-ISOL Workshop, Mont Gabriel (Quebec) 1984

[4] *H. A. O'Brien*, "Status of the Los Alamos Physics Facility For Radionuclide Production" Sci. Sem. of Nucl. Med., DHEW Publication 74-8012 (FDA) 14—19, U.S. Gov. Printing Office 1973

[5] *J. N. Bradbury*, IEEE Trans. Nucl. Sci. NS-26 (1979) 139

[6] *H. A. O'Brien, A. E. Ogard, P. M. Grant*, "Radioisotopes from a medium energy accelerator: The Clincton P. Anderson Meson Physics Radioisotope Programe" LA-Report 74-1263

[7] *P. Richards*, "Status of the BLIP Facility for Radionuclide Production" Sci. Sem. of Nucl. Med., DHEW Publication 74-8012 (FDA) 22—26, U.S. Gov. Printing Office 1973

[8] *B. D. Pate*, Am. Nucl. Soc. (1979) 283

[9] *L. G. Strang*, Progr. Nucl. Med. 4 (1978) 34

[10] *J. J. Burgerjon, B. D. Pate, R. E. Blaby, E. G. Page, J. Lenz, P. T. Treretti, A. D. Wilson*, Am. Nucl. Soc. (1979) 285

[11] *R. Robertson, D. Graham, I. C. Travena*, "Radioisotope Production via 500 MeV Proton-induced Reactions" Third Int. Smp. on Radiopharmaceutical Chemistry, Juelich (FRG) 1982

[12] *G. Rudstam*, Z. Naturforschung 21a (1966) 1027

[13] *A. K. Lavrukhina, R. I. Kuznetzova*, JINR 6-4756 Dubna 1969, pp. 125 (in Russian)

[14] *E. Rupp, T. Fenes*, "Cross-sections for the formation of spallation products using 660 MeV protons", JINR 6-4998, Dubna 1970 (in Russian)

[15] *M. E. Schilacci*, LASL Report MPDOT/MES-1, 1970

[16] *U. Trabitzsch, K. Bacchmann*, Radiochimica Acta 16 (1971) 15

[17] *I. Huzar, E. Loepfe*, "Production of Cyclotron Isotopes for Medical Purpose at SIN/EIR", Physics Report No. 3, p. 139 (1981), Swiss Inst. for Nucl. Res. Villigen, Switzerland

[18] *M. Adilbish, C. G. Chumin, V. A. Khalkin, O. Knotek, M. Ja. Kuznetsova, Ju. V. Norceev, V. I. Fominykh, N. G. Zaitseva*, Int. J. Appl. Radiat. Isot. 31 (1980) 163

[19] *C. D. Hoffman*, "Isotope and Nuclear Chemical Division, Annual Report FY 1983 Oct. 1982-Sept. 1983" ed. by *J. H. Heiken* and *H. A. Lindberg* LA-10130 PR Progress Report UC-4 May 1984

[20] *H. A. O'Brien Jr.*, "Tables of spallation cross-sections" Paper presented to the IAEA Consultants Meeting on Nuclear Data for Medical Radioisotope Production held in Vienna, April 13—15, 1981

[21] *G. G. Jonsson, K. Lindgren*, Phys. Scripta 7 (1973) 49

[22] *G. G. Jonsson, K. Lindgren*, Phys. Scripta 15 (1977) 308

[23] *H. L. Ravn*, "Radioactive Ion-beams available at on-line Mass Separators", CERN-EP/86-128, 12. Sept. 1986, Proc. 11th Int. Conference on Electromagnetic Isotope Separators and Techniques related to Their Application Los Alamos, N. M., August 18—22, 1986

[24] *R. M. Barrer*, "Diffusion in and through solids" (*E. K. Rideal*, ed.). New York: Macmillan Co. 1941

[25] *K. E. Zimen*, „Tabellen zur Auswertung der Messungen der Diffussion radioaktiver Gase" HMI-B 16 (1962)

[26] *G. J. Beyer, A. F. Norgorodov*, "Tracer Diffusion einiger Spallations-Produkte in polykristallinem β-Zr und β-Ti", ZfK-328 Rossendorf 1977

[27] *A. F. Norgorodov, G. J. Beyer, A. S. Kovalev*, "Tracer-diffusion of spallation products in polycristalline β-Zirconium" (in Russian) JINR 6-12113 Dubna 1979

[28] *G. J. Beyer, A. F. Norgorodov*, Radiochem. Radioanal. Letters 27 (1976) 341

[29] *G. J. Beyer, A. F. Norgorodov*, "Tracer Diffusion einiger Spallationsnuklide in polykristallinem Molybdaenen", ZfK-317 Rossendorf 1976

[30] *G. J. Beyer, A. F. Norgorodov*, "Tracer Diffusion einiger Kernreaktionsprodukte in Hafnium-Metal", ZfK-303 Rossendorf 1976, JINR P6-9916 Dubna 1976 (in Russian)

[31] *G. J. Beyer, W. D. Fromm, A. F. Norgorodov*, Nucl. Instrum. Methods 146 (1977) 419

[32] *G. J. Beyer, A. F. Norgorodov*, "Tracer Diffusion einiger Kernreaktionsprodukte in Wolfram", ZfK-310 Rossendorf 1976

[33] *T. Bjornstad, E. Hagebo, P. Hoff, O. C. Jonsson, E. Kugler, H. L. Ravn, S. Sundell, B. Vosicki*, "Recent developments of high temperature metal targets for ISOLDE", CERN-EP/86-130 Sept. 1986, Proc. 11th Int. Conference on Electromagnetic Isotope Separators and Techniques related to Their Application, Los Alamos, N. M., August 18—22, 1986

[34] *G. J. Beyer, A. F. Norgorodov*, „Thermische Desorption trägerfreier Radionuklide der Seltenerden von polykristallinen Ta-Oberflächen", ZfK-305 Rossendorf 1976, JINR P6-9917 Dubna 1976 (in Russian)

[35] *F. Rösch, B. Eichler*, „Adsorption von Metallen auf metallischen Oberflächen und Möglichkeiten ihrer Nutzung in der Kernchemie", ZfK-606 Rossendorf 1986 (ISSN 0138-2950)

[36] *T. Bjornstad, E. Hagbo, P. Hoff, O. C. Jonsson, E. Kugler, H. L. Ravn, S. Sundell, B. Vosicki*, "Methods for production of intense beams of unstable nuclei: New developments at ISOLDE", CERN-EP 86-130 Sept. 1986, Physica Scripta, in print

[37] *G. J. Beyer, E. Herrmann, A. Piotrovski, V. I. Raiko, H. Tyrroff*, Nucl. Instrum. Methods 96 (1971) 41

[38] *R. Kirchner, O. Klepper, D. Schardt, T. Sekine*, "Ion source development for the GSI on-line isotope separator", GSI-86-30 Preprint Aug. 1986; *R. Kirchner*, "An ion-source with bunched beam release", GSI-86-28 PREPRINT Aug. 1986

[39] *B. Vosicki, T. Bjornstad, L. C. Carras, J. Heinemeier, H. L. Ravn*, Nucl. Instrum. Methods 186 (1981) 307

[40] *G. J. Beyer, O. Knotek, J. Juschkevich, M. Jachim, A. F. Norgorodov*, Nucl. Instrum. Methods 148 (1978) 543

[41] *A. F. Norgorodov, A. Kolochkovski, Nguen Kong Tchang, A. Zelinski, M. Kaskevich*, "High temperature release of some spallation products from Ni", JINR P6-85-11 Dubna 1985

[42] *G. J. Beyer, A. F. Norgorodov, A. Kovalev, F. Prazak, V. A. Khalkin, Yu. V. Yushkevich*, Nucl. Instrum. Methods 186 (1981) 401

[43] *G. J. Beyer, H. L. Ravn, Y. Huang*, Int. J. Appl. Radiat. Isot. 35 (1984) 1075

[44] *H. J. Kluge* (ed.), "CERN USERS GUIDE", CERN 86-05 Juli 1986, to be published as CERN-report

[45] *S. L. Waters, B. M. Coursey* (eds.), "The Strontium-82-Rubidium-82 Generator" Special Issue Int. J. Appl. Radiat. Isot. 38 (1987)

[46] *P. M. Grant, B. R. Erdal, H. A. O'Brien Jr.*, Int. J. Appl. Radiat. Isot. 34 (1983) 1631

[47] *P. M. Grant, H. A. O'Brien, M. Kahn*, J. Inorg. Nucl. Chem. 37 (1975) 413

[48] *P. L. Harlock, J. C. Clark, I. W. Goodier, J. W. Barnes, G. E. Bentley, P. M. Grant, H. A. O'Brien*, J. Radioanal. Chem. 64 (1981) 257

[49] *K. E. Thomas*, Int. J. Appl. Radiat. Isot. 38 (1987) 175

[50] *T. W. Eaton, H. L. Ravn*, "Beam heating of thick targets for on-line mass-separators" CERN-EP/86—135, Sept. 1986, Proc. 11th Int. Conference on Electromagnetic Isotope Separators and Techniques related to Their Application Los Alamos, N.M., August 18—22, 1986

[51] *N. P. Peck, F. Hagedues*, Int. J. Appl. Radiat. Isot. 30 (1979) 631

[52] *J. W. Barnes, M. A. Ott, K. E. Thomas, P. M. Wanek*, "123-Iodine" Isotope Production and Application, Annual Report FY 1983, D. C. Heiken, H. A. Lindberg (eds.), LA-10 130 PR, UC-4, May 1984, pp. 85

[53] *H. A. O'Brien Jr.*, "Radioisotope Programme at LAMPF", Proc. of the Symp. on Practical Application of Accelerators, Session I: Radioisotopes, Los Alamos, Aug. 1973 LA-5535-C (1974) 93—107

[54] *G. Rudstam, G. Sorensen*, J. Inorg. Nucl. Chem. 28 (1966) 771

[55] *P. M. Grant, D. A. Miller, G. S. Gilmore, H. A. O'Brien Jr.*, Int. Appl. Radiat. Isot. 33 (1982) 415

[56] *B. R. Erdal, P. M. Grant, V. R. Casella, A. E. Ogard, H. A. O'Brien*, "Spallation cross-sections and the LAMPF medical radioisotope programme" LA-UR-75-359, in: *R. A. Schrack, C. B. Bowman* (eds.), Proc. of Conf. "Nuclear Cross Sections and Technology, Vol. II", Washington, March 3—7, 1975, 492—495

[57] *T. Bjornstad, L. C. Carraz, H. A. Gustavsson, J. Heinemeyer, B. Jonson, O. C. Jonsson, V. Lindfors, S. Mattsson, H. L. Ravn*, Nucl. Instrum. Methods 186 (1981) 391

[58] *K. L. Scholz, V. Sodd, J. W. Blue*, Int. J. Appl. Radiat. Isot. 27 (1976) 263

[59] *G. J. Beyer, R. Muenze, W. D. Fromm, W. G. Franke, H. Henke, V. A. Khalkin, N. A. Lebedev*, "Spallation produced 167-Tm for medical application", in: "Radionuclide Imaging 1980". Vienna: IAEA 1981 Vol. 1, 587—598 (IAEA-SM-247/60)

[60] *H. L. Ravn*, Phys. Rev. 54 (1979) 201

[61] *H. L. Ravn, T. Bjornstad, P. Hoff, O. C. Jonsson, E. Kugler, S. Sundell, B. Vosicki*, "Use of refractory oxides, carbides and borides for on-line mass separation", CERN-EP/86-147, Oct. 1986, presented at the 11th Int. Conf. on Electromagnetic Isotope Separators and Techniques related to Their Applications, Los Alamos, Aug. 18—22, 1986

[62] *R. M. Lambrecht*, Radiochimica Acta 34 (1983) 9

[63] *Yu. V. Norseev, S. Suez, V. A. Khalkin, F. Rösch*, to be published

[64] *V. A. Khalkin, E. Herrmann, Yu. V. Norseev, R. Dreyer*, Chemiker-zeitung 101 (1977) 470

[65] *M. Lindner, R. N. Osborne*, Phys. Rev. 103 (1956) 378

Structure of 9Be Nucleus Using the Generator Coordinate Method and the Three-Body Model

A. Osman, M. Zaky

Cairo University, Faculty of Science, Physics Department, Cairo, Egypt

The structure of the 9Be nucleus is considered as being composed of three clusters (two alpha particles and a neutron). The bound state of the 9Be nucleus is studied using the approaches of the generator coordinate method as well as the three-body model formalism. The numerical calculations are carried out using different types of two-body interactions. Yamaguchi, Gaussian, Tabakin and Volkov type potentials are used. The three-body model is also used in treating the three clusters forming the 9Be nucleus by following the Faddeev formalism. In both approaches, the low-lying energy levels of the 9Be nucleus as well as its binding energy are numerically calculated. Good agreements are obtained between the present theoretically calculated values and the experimental data.

Die Struktur des 9Be-Kerns wird als ein Cluster aus drei Teilchen betrachtet (zwei α-Teilchen und ein Neutron). Der gebundene Zustand des 9Be-Kerns wird unter Anwendung der Methode der erzeugenden Koordinaten und des Dreikörper-Kernmodell-Formalismus untersucht. Die numerischen Berechnungen wurden mit verschiedenen Zweikörper-Wechselwirkungen ausgeführt, wobei Yamaguchi-, Gauß-, Tabakin- und Volkov-Potentialen verwendet werden. Das Dreikörper-Modell wurde auch bei der Behandlung des Dreiteilchen-Clusters angewendet, das den 9Be-Kern nach dem Faddeev-Formalismus bildet. Für beide Näherungen wurden die tiefliegenden Energieniveaus des 9Be-Kerns und ihre Bindungs-energien numerisch berechnet und es konnten befriedigende Übereinstimmungen zwischen den berechneten und den experimentellen Daten erzielt werden.

Keywords

1. Introduction

The nuclear structure of light nuclei have been shown as one of the important tools for extracting some static properties of these nuclei. The nuclear shell model has been extensively used in an attempt to explain certain features of light nuclei. The shell model calculations lead to the result that both of the L-S and J-J coupling schemes do not give an adequate explaination of the relevant experimental data. The intermediate coupling in the p-shell for all nuclei has been extensively calculated and have been shown that the energies of almost all known low-lying levels are reproduced satisfactorily with good agreement with the experimental magnetic moments, M1 transition probabili-