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POSITRON-EMITTING RADIONUCLIDES - PRESENT AND FUTURE STATUS

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ABSTRACT

A tabulation of 157 positron-emitting radionuclides which have the physical characteristics deemed appropriate for radiopharmaceutical use in conjunction with positron emission tomography is provided. The most promising radionuclides are within the production capabilities of a variable energy cyclotron accelerating protons to about 40 MeV and deuterons, helium-3, and helium-4 to compatable energies. To date only 27 positron emitting radionuclides have been subjected to radiopharmaceutical consideration, whereas only ^{11}C, ^{13}N, ^{15}O, ^{18}F, ^{38}K, and ^{68}Ga have proved to be especially promising.

INTRODUCTION

A combination of recent events (1-3) has resulted in a significant movement towards cyclotron and generator produced short-lived positron emitting radionuclides and radiopharmaceutical chemistry to be used in combination with positron emission tomography for the quantitative determination of metabolic function and disease state. Earlier in these proceedings Silvester (4) and Wolf (5) have discussed the production of medical radionuclides and organic radiopharmaceuticals labeled with cyclotron produced radionuclides. Demonstratable success with short-lived positron emitting radionuclides has been limited to ^{11}C, ^{13}N, ^{15}O, ^{18}F, ^{38}K, and ^{68}Ga although ^{14}O, ^{21}Na, ^{30}P, ^{34m}Cl, ^{45}Ti, ^{49}Cr, ^{51}Mn, ^{52m}Mn, ^{55}Co, ^{61}Cu, ^{62}Cu, ^{66}Ge, ^{75}Br, ^{76}Br, ^{77}Kr, $^{81,82m}Rb$, ^{82}Rb, ^{104}Ag, ^{121}I, ^{122}I, and ^{123}Xe have been reported to be in various stages of development. A complete bibliography of papers and reports published on all aspects of the fundamental and applied studies with positrons, positron emitters, positron emission tomography are compiled elsewhere (6,7) as part of a continuing effort. The intent here is to present the first comprehensive survey of the positron-emitting radionuclides which are deemed to have appropriate physical characteristics and be within the production capability of a cyclotron.

DISCUSSION

Table 1 is a compilation of published nuclear data for 157 nuclides listed in order of increasing atomic number and identifying the half life and the abundance of positrons per 100 decay events. An indication of the complexity of the positron energy spectra is summarized by listing the number of distinguishable positrons, and the median positron energy of the spectra calculated by Hogan (8) using the Fermi theory of beta decay. The Table lists 8 nuclides with a t 1/2 of 60 s or less, 12 with t 1/2 of 1-2 m, 26 with t 1/2 = 2-10 m, 24 with t 1/2 of 10-30 m, 40 with t 1/2 of 0.5-2 h, 27 with t 1/2 of 2-10 h, and 20 with t 1/2 > 10 h.

The status of the nuclear data of several of the radionuclides is not without question. The status of nuclear data, not only decay but also production, of some of the medical radionuclides has been reviewed (9) or compiled without evaluation (10,11). The prerequisite positron abundance necessary to make a nuclide useful is not yet defined due to rapid developments in instrumentation and the absence of extensive studies. Data on the median energy of the positron spectra is presented as a criteria indicative of the degree of influence the positron spectra and range-energy distribution will have on the resolution of the emission tomograph. The data are intended as a more practical aid. The interpretation of the effect of positron range on the resolution with a volume element viewed with a positron emission tomograph is subject to argument (12-21). Fig. 1 depicts an interpretation of the variation in the relative FWHM of one spectrometer (12) as a function of the relative path length (20) of the relatively uncomplicated positron spectra of ^{18}F, ^{11}C, ^{13}N, ^{15}O, and ^{68}Ga. Derenzo and Budinger (19) have suggested that it will not be possible to achieve a resolution for a positron emission tomograph of greater than the square root of $(\text{angular spread})^2 + (\text{range spread})^2$ which for ^{68}Ga is 4 mm.

The minimum accepted positron abundance for the purposes of Table 1 was 2.5%. Of the 157 nuclides, 86 decay with $\geq$ 50% abundance of positrons, i.e. the annihilation radiation is $\geq$ 100 γ's per 100 decay events.

Lambrecht et al. (23) have reported that $^{81,82m}Rb$ seems satisfactory for emission tomography of the myocardium. The result was initially surprising due to a report (24) that the positron abundance of ^{81}Rb was ∿ 12%. Recent decay scheme studies have shown the positron abundance of ^{81}Rb and ^{82m}Rb to be 27% and 26%, respectively (24).

The choice of a radionuclide for radiopharmaceutical applications depends on a combination of physical, chemical, and physiological considerations. For example, ^{38}K (t 1/2 = 7.62 m, β^+ = 100%, γ = 2.17 MeV, 100%) has demonstrated (24) all the criteria required for an "ideal" radiopharmaceutical. The high energy gamma did not interfer with the tomographic quality obtainable with the PETT III. In fact, it has been suggested that the coincident annihilation radiation and the 2.17 MeV gamma should be considered (21,26) for dual tomographic and positron annihilation lifetime determinations *in vivo*.

The production and processing of simple inorganic species which have physiological properties to permit near-direct clinical use or which can be rapidly manipulated into synthetic intermediates for radiopharmaceutical preparation are key points in the widespread use of short-lived positron emitting radionuclides. These parameters will be discussed in detail elsewhere. The absorbed radiation dose becomes a more serious consideration with nuclides having a half life of > 3 h, except perhaps in therapeutic or high benefit-vs.-risk circumstances. Siegel (27) suggested ^{55}Co-labeled bleomycin may be one identifiable example, and research has progressed in this direction (28,29).

As the radiopharmaceutical sciences evolve in part into the direction of positron emission tomography, it is apparent that cyclotrons and accelerators (tandem, electron, and particle linacs) will be essential for production of radionuclides for near-direct clinical use, for on-line preparation of synthetic intermediates required in synthetic organic, metallo-organic and inorganic chemistry, and for production and distribution of longer-lived positron-emitters or their generators.

Justification of a medical cyclotron depends on the envisioned use. Initial economic factors argue for a single particle, low-energy machine devoted to the production of ^{11}C, ^{13}N, ^{15}O, and ^{18}F as the key positron-emitting radionuclides. Without doubt these radionuclides in combination with ^{68}Ge-^{68}Ga and possibly ^{82}Sr-^{82}Rb and ^{44}Ti-^{44}Sc generators will have a principle role in future radiopharmaceutical developments. However, scientific and philosophical arguments argue that too small a machine may be short-sighted.

Examples of other bioelements required for human metabolism are K, P, Se, Rb, and Mn. None of the appropriate radionuclides of these elements can be produced in acceptable yield and radionuclidic purity with a low energy single particle cyclotron. The $^{40}Ar(p,3n)^{38}K$ reaction with 32 MeV protons (26,30) and the $Cl(\alpha,n)^{38}K$ reaction with 15 MeV alphas (31) have been utilized for production of the ideal radionuclide. Sahakundu et al. (32) concluded the best route to ^{30}P was via the $^{27}Al(\alpha,n)^{30}P$ reactions with 28 MeV alphas. The production of ^{73}Se in acceptable radionuclidic purity and yield requires either ^{3}He or ^{4}He particles (33-35). Recently, $^{81,82m}Rb$ produced via the $^{82}Kr(p,xn)$ reactions on natural Kr was shown to be an appropriate radionuclidic pair for emission tomography of the myocardium (21) in instances where ^{38}K cannot be produced in-house. In addition, the production route with > 32 MeV protons is yet the most appropriate for routine production of ^{81}Rb-^{81m}Kr generators (36).

In many respects the ^{52}Fe-^{52m}Mn generator (37-39) is probably limited to research applications due to the fact that a high energy, high fluence accelerator is required to produce multi-mCi quantities of ^{52}Fe. The 8.27 h half life is a limitation since chemistry and shipment considerations will probably result in a generator of 1 day or less shelf life. By fortunate circumstance either of two Mn radionuclides can be produced in high yield and purity via the $^{50}Cr(p,2n)^{51}Mn$ or $^{50}Cr(p,n)^{52m}Mn$ nuclear reactions (30).

Positron emitting isotopes of presently used γ-emitting radionuclides which merit further consideration are listed in Table 2.

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TABLE 1. CHARACTERISTICS OF POSITRON EMITTING RADIONUCLIDES HAVING POTENTIAL APPLICATIONS IN THE RADIOPHARMACEUTICAL SCIENCES

Nuclide[a]	Half life[b]	No. of Positrons[b,c]	Abundance[b,c]	E_{median}[c] MeV
^{11}C	20.38 m	1	0.998	0.375
^{13}N	9.96 m	1	0.998	0.475
^{14}O	70.6 s	3	1.000	0.768
^{15}O	122 s	1	0.99+	0.723
^{17}F	64.5 s	1	1.000	0.729
^{18}F	109.8 m	1	0.969	0.245
^{18}Ne	1.67 s	2	1.000	1.385
^{19}Ne	17.3 s	1	0.99+	*
^{21}Na	22.47 s	2	1.000	1.085
^{23}Mg	11.3 s	2	0.950	1.308
^{30}P	2.50 m	2	1.000	1.455
^{34m}Cl	32.0 m	3	0.53	0.712
^{38}K	7.61 m	1	0.995	1.113
$^{42m+g}Sc$	62.0 s	2	1.000	2.200
^{43}Sc	3.89 h	3	*	*
^{44}Sc	3.93 h	1	0.95	0.616
^{45}Ti	3.09 h	2	0.853	0.417
^{47}V	32.6 m	1	0.970	0.812
^{48}V	15.967d	1	0.496	0.283
^{49}Cr	41.9 m	3	0.932	0.627
^{50m}Mn	1.74 m	2	0.990	1.476
^{50}Mn	0.28 s	1	1.000	3.552
^{51}Mn	46.2 m	3	0.972	0.935

Nuclide[a]	Half life[b]	No. of Positrons[b,c]	Abundance[b,c]	E_{median}[c] MeV
^{52m}Mn	21.1 m	1	*	*
^{52}Mn	5.59 d	2	0.276	0.215
^{52}Fe	8.27 h	1	0.565	0.336
^{53}Fe	8.51 m	3	0.975	1.064
^{54m}Co	1.46 m	1	1.000	2.078
^{54}Co	0.19 s	1	1.000	3.438
^{55}Co	17.5 h	3	0.77	0.510
^{57}Ni	36.0 h	3	0.40	0.338
^{59}Cu	82 s	6	0.984	1.531
^{60}Cu	23.4 m	5	0.936	0.957
^{61}Cu	3.41 h	4	0.622	0.480
^{62}Cu	9.73 m	4	0.978	1.290
^{64}Cu	12.70 h	1	0.190	0.273
^{61}Zn	89.1 s	4	0.991	1.882
^{62}Zn	9.2 h	2	0.069	0.279
^{63}Zn	38.1 m	5	0.93	0.911
^{64}Ga	2.62 m	2	0.990	1.975
^{65}Ga	15.2 m	4	0.86	*
^{66}Ga	9.4 h	6	0.567	1.712
^{68}Ga	68.1 m	2	0.875	0.811
^{66}Ge	2.3 h	6	0.27	0.574
^{67}Ge	19.0 m	4	0.96	1.200
^{69}Ge	39.0 h	4	0.361	0.492
^{69}As	15 m	1	0.98	1.290

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^{11}C	20.38 m	1	0.998	0.375
^{13}N	9.96 m	1	0.998	0.475
^{14}O	70.6 s	3	1.000	0.768
^{15}O	122 s	1	0.99+	0.723
^{17}F	64.5 s	1	1.000	0.729
^{18}F	109.8 m	1	0.969	0.245
^{18}Ne	1.67 s	2	1.000	1.385
^{19}Ne	17.3 s	1	0.99+	*
^{21}Na	22.47 s	2	1.000	1.085
^{23}Mg	11.3 s	2	0.950	1.308
^{30}P	2.50 m	2	1.000	1.455
^{34m}Cl	32.0 m	3	0.53	0.712
^{38}K	7.61 m	1	0.995	1.113
$^{42m+g}Sc$	62.0 s	2	1.000	2.200
^{43}Sc	3.89 h	3	*	*
^{44}Sc	3.93 h	1	0.95	0.616
^{45}Ti	3.09 h	2	0.853	0.417
^{47}V	32.6 m	1	0.970	0.812
^{48}V	15.967d	1	0.496	0.283
^{49}Cr	41.9 m	3	0.932	0.627
^{50m}Mn	1.74 m	2	0.990	1.476
^{50}Mn	0.28 s	1	1.000	3.552
^{51}Mn	46.2 m	3	0.972	0.935

Nuclide[a]	Half life[b]	No. of Positrons[b,c]	Abundance[b,c]	E_{median}[c] MeV
^{52m}Mn	21.1 m	1	*	*
^{52}Mn	5.59 d	2	0.276	0.215
^{52}Fe	8.27 h	1	0.565	0.336
^{53}Fe	8.51 m	3	0.975	1.064
^{54m}Co	1.46 m	1	1.000	2.078
^{54}Co	0.19 s	1	1.000	3.438
^{55}Co	17.5 h	3	0.77	0.510
^{57}Ni	36.0 h	3	0.40	0.338
^{59}Cu	82 s	6	0.984	1.531
^{60}Cu	23.4 m	5	0.936	0.957
^{61}Cu	3.41 h	4	0.622	0.480
^{62}Cu	9.73 m	4	0.978	1.290
^{64}Cu	12.70 h	1	0.190	0.273
^{61}Zn	89.1 s	4	0.991	1.882
^{62}Zn	9.2 h	2	0.069	0.279
^{63}Zn	38.1 m	5	0.93	0.911
^{64}Ga	2.62 m	2	0.990	1.975
^{65}Ga	15.2 m	4	0.86	*
^{66}Ga	9.4 h	6	0.567	1.712
^{68}Ga	68.1 m	2	0.875	0.811
^{66}Ge	2.3 h	6	0.27	0.574
^{67}Ge	19.0 m	4	0.96	1.200
^{69}Ge	39.0 h	4	0.361	0.492
^{69}As	15 m	1	0.98	1.290

Nuclide[a]	Half life[b]	No. of Positrons[b,c]	Abundance[b,c]	E_{median}[c] MeV
^{70}As	53 m	2 (3)	0.84	0.680
^{71}As	61 h	2	0.32	00.343
^{72}As	26.0 h	6	0.777	1.147
^{74}As	17.78 d	1 (2)	0.309	0.406
^{71}Se	4.9 m	2	1.000	1.477
^{73}Se	7.1 h	2 (4)	0.65	0.566
^{74m}Br	41 m	2	*	*
^{74}Br	25.3 m	1	1.000	2.004
^{75}Br	98 m	4 (3)	0.755	0.376
^{76}Br	16.1 h	7	0.57	1.200
^{78}Br	6.46 m	2	0.932	1.076
^{74}Kr	11.5 m	1	1.000	1.385
^{77}Kr	75 m	3 (4)	0.80	0.741
^{79}Kr	35.0 h	2	0.07	0.254
^{79}Rb	23.0 m	3	0.84	1.063
^{80}Rb	34 s	2	0.977	1.827
^{81m}Rb	32 m	1	0.500	0.603
^{81}Rb	4.58 h	3	0.27	0.372
^{82m}Rb	6.2 h	1	0.256	0.332
^{82}Rb	1.25 m	3	0.962	1.382
^{81}Sr	26 m	2	0.87	1.244
^{83}Sr	32.4 h	(3)	0.24	(0.494)
^{84}Y	39 m	5	0.865	1.201
$^{85(g)}Y$	2.7 h	(4)	0.70	*

Nuclide[a]	Half life[b]	No. of Positrons[b,c]	Abundance[b,c]	E_{median}[c] MeV
$^{85(m)}Y$	4.9 h	(3)	0.55	(0.875)
^{86}Y	14.74 h	6	0.34	*
^{87}Zr	1.6 h	1	0.830	0.920
^{89}Zr	78.4 h	1	0.223	0.390
^{89}Nb	66 m	1	0.74	*
^{89}Nb	2.0 h	1	0.910	1.270
^{90}Nb	14.6 h	3	0.53	0.602
^{90}Mo	5.67 h	1	0.25	0.498
^{91m}Mo	65 s	3	0.280	1.210
^{91}Mo	15.49 m	1	0.941	*
^{92}Tc	4.4 m	1	1.000	1.857
^{93}Tc	2.7 h	3	0.13	0.248
^{94m}Tc	52 m	1 (3)	0.72	0.434
^{94}Tc	293 m	3 (1)	0.11	1.036
^{95}Ru	1.65 h	3	0.15	0.459
^{97m}Rh	44 m	1	(1.000)	(0.722)
^{98}Rh	8.7 m	1 (2)	(0.170)	(1.181)
^{100}Rh	20.8 h	5	0.049	0.919
^{98}Pd	18 m	*	*	*
^{99}Pd	21.4 m	4	*	*
^{102}Ag	13.0 m	2	0.68	*
^{103}Ag	1.10 h	3	0.42	*
^{104m}Ag	33 m	1	0.590	1.202
^{104}Ag	69 m	1	0.150	0.434

Nuclide[a]	Half life[b]	No. of Positrons[b,c]	Abundance[b,c]	E_{median}[c] MeV
^{106}Ag	24.0 m	2	0.610	0.826
^{105}Cd	56.0 m	(2)	*	*
^{106}In	6.3 m	(1)	*	*
^{107}In	32.4 m	1	0.35	0.882
^{108}In	39.6 m	(1)	0.110	1.512
^{108}In	58.0 m	1	0.20	0.585
^{109}In	4.2 h	(1)	0.06	*
^{110}In	69 m	1	0.72	1.006
^{109}Sn	18.0 m	(5)	*	*
^{111}Sn	35 m	1	0.290	0.640
^{114}Sb	3.5 m	2	1.000	0.144
^{115}Sb	31.8 m	1	0.33	0.665
^{116m}Sb	60.4 m	2	0.192	0.634
^{116}Sb	16 m	2	0.28	0.931
^{117}Sb	2.80 h	1	0.025	0.288
^{118}Sb	3.5 m	2	0.828	1.364
^{117}Te	62 m	(1)	0.30	*
^{120m}I	53 m	1	*	*
^{120}I	1.35 h	1	0.46	1.802
^{121}I	2.12 h	1	0.064	0.506
^{122}I	3.6 m	1 (3)	0.77	1.386
^{124}I	4.2 d	3	0.25	0.788
^{121}Xe	39 m	1	0.08	1.233
^{123}Xe	2.08 h	1	0.13	0.666

Nuclide[a]	Half life[b]	No. of Positrons[b,c]	Abundance[b,c]	E_{median}[c] MeV
^{125}Cs	45 m	2	0.39	*
^{126}Cs	1.64 m	2	0.82	1.636
^{127}Cs	6.2 h	4	0.035	0.364
^{128}Cs	3.6 m	4	0.61	1.138
^{130}Cs	29.9 m	1	*	*
^{130}La	8.7 m	(1)	*	*
^{131}La	61 m	3	0.245	0.603
^{132}La	4.8 h	1	0.810	1.637
^{133}La	3.91 h	1	1.000	0.534
^{134}La	6.67 m	2	0.618	1.186
^{136}La	9.87 m	2	0.332	0.855
^{131}Ce	10 m	1	0.113	*
^{133}Ce	5.4 h	1	0.150	0.578
^{135}Pr	25 m	1	0.25	1.111
^{137}Pr	1.28 m	1	0.255	0.754
^{138m}Pr	2.1 h	1	0.236	0.622
^{139}Pr	4.4 h	1	0.062	0.449
^{140}Pr	3.30 m	1	0.487	1.075
^{139}Nd	30 m	1	0.256	1.382
^{141}Pm	20.9 m	1	0.57	1.156
^{142}Pm	40.5 s	1	0.69	1.693
^{142}Sm	2.49 m	1	0.10	0.451
^{143}Sm	8.83 m	1	0.465	1.111
^{145}Gd	22 m	1	1.000	1.066

Nuclide[a]	Half life[b]	No. of Positrons[b,c]	Abundance[b,c]	E_{median}[c] MeV
^{152}Tb	17.5 h	3	0.145	0.938
^{155}Ho	49 m	(1)	(1.000)	(0.933)
^{156}Ho	56 m	(3)	(1.000)	(0.933)
^{158m}Ho	27 m	1.320	1.000	0.593
^{158}Er	2.4 h	1	1.000	0.362
^{164}Tm	2.0 m	2	0.39	*
^{165}Yb	10 m	(1)	*	*
^{168m}Lu	6.7 m	2	0.012	*
^{168}Lu	5.5 m	1	1.000	0.544
^{178}Re	13.2 m	1	0.113	1.372
^{180}Re	2.42 m	1	0.08	0.847
^{186}Ir	1.7 h	5	1.000	0.856
$^{190m2}Ir$	3.2 h	1	0.050	0.891
^{194}Au	39.5 h	2	0.03	0.604

(a) Arranged in order of increasing atomic number

(b) Lederer, C. M., Shirley, V. S. (eds.), Browne, E., Dairiki, J. M., Doebler, R. E., Shihab-Eldin, A. A., Jardine, L. J., Tuli, J. K., Buyrn, A. B. Table of Isotopes, 7th Ed., J. Wiley and Sons, New York, 1978.

(c) Hogan, O. L., "Beta Spectra V. Spectra of Individual Positron Emitters" Report USNRDL-TR-1101, 14 November 1966, 126 pp.

* Insufficient nuclear decay data available for (c) calculation

( ) Parenthesis indicates the value in table may be in error

TABLE 2

POSITRON-EMITTING ISOTOPES OF γ-EMITTING RADIONUCLIDES

Nuclide	t 1/2	β^+ Abundance	Production*
^{110}In	69 m	72 %	$^{107}Ag(\alpha,n)$ $^{112}Cd(p,n)$
^{66}Ga	9.4 h	56.7%	$^{66}Zn(\alpha,4n)^{66}Ge \xrightarrow[2.27\ h]{} {}^{66}Ga$ $^{66}Zn(^{3}He,3n)$
^{95}Ru	1.65 h	15 %	$^{94,95}Ru(\alpha,xn)$ $^{94,95}Ru(^{3}He,xn)$

*isotopically enriched targets