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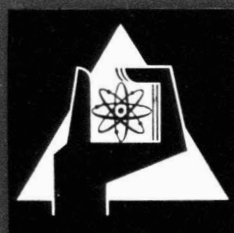
Institut für Neutronenphysik und Reaktortechnik

Present Status and Future Prospects of Production and Utilization  
of Radioisotopes in Germany

K. Wirtz

Gesellschaft für Kernforschung m. b. H.  
Zentralbücherei

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by

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1. Introduction

Up till 1949 there was no utilization of radioisotopes whatsoever in Germany, apart from a few radium sources which have always been kept for medical and scientific purposes. In 1949 the British Military Government of that time gave permission to the "Max-Planck-Gesellschaft for the Advancement of Science" to import artificially produced radionuclides and for this purpose recommended the creation of a so-called Isotope Laboratory of Max-Planck-Gesellschaft. As is known, Max-Planck-Gesellschaft succeeded the former "Kaiser-Wilhelm-Gesellschaft for the Advancement of Science"; it sponsors a large number of government-financed research institutes in all scientific fields. The Isotope Laboratory was erected in Göttingen and started work on the basis of radioisotopes imported mainly from Harwell. At that time Germany did not yet have a nuclear reactor, not even permission to build one. Moreover, as there were no regulations to govern handling, trading and processing of radioactive isotopes, the military authorities wished to have the distribution of isotopes centralized. For all these reasons the tasks of the Isotope Laboratory concentrated upon distributing radionuclides and supervising their use and in advising all persons interested in the use of artificial radioactive isotopes, particularly with regard to safety questions.

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The consumption of radioactive substances increased rapidly after 1949, and already in the first six months of 1952 these isotopes were imported:

Phosphorus-32	2612 millicuries
Iodine-131	7112 mc
Cobalt-60	5045 mc
Iridium-192	10500 mc
Sodium-24	} 7970 mc
Copper-64	
Gold-198	
Bromine-82	
Carbon-14	42 mc
Others	1548 mc
Total*	34829 mc

These quantities were shipped in 435 individual shipments to more than 100 research institutes, hospitals and industries. The Isotope Laboratory of Max-Planck-Gesellschaft was incorporated into the Karlsruhe Nuclear Research Center in 1957. We will refer to its work in Karlsruhe later on.

The second important date for our topic is that up till the end of 1962 radioisotopes could not be produced on a larger scale in the Federal Republic of Germany. Up to the end of 1962 no powerful research reactor was available for isotope production. Thus, the major part of the radioisotopes presently used in the Federal Republic is imported. This will change in the course of 1963 as the research reactor FR 2 of the Karlsruhe Nuclear Research Center, a natural-uranium, heavy-water reactor of 12 MW power which has been finished in the meantime will be available for routine work. Also at the Nuclear Research Center Jülich a powerful reactor starts work in 1963.

\*In 1952 a total amount of 100 Curies was reached.

## 2. Regulatory Provisions

As late as in 1960 the so-called "First Regulation Covering Protection Against Damage by Radiation of Radioactive Substances" was issued. Up to that year there had been no legislative rules in this field apart from older regulations covering work with X-rays. The "Strahlenschutzverordnung" (Radiation Protection Regulations) covers these fields:

1. Handling of radioactive substances (extraction, production, storage, processing, other uses, and removal)
2. Shipment and transport of radioactive substances
3. Import and export of radioactive substances
4. Trade in radioactive substances (purchase and sale to third parties).

The regulations distinguish between sealed and unsealed samples. Above all, the new set of rules decides under what conditions somebody may get permission to handle, import or export, or trade radioactive matter. Moreover, maximum permissible exposure levels are set in the regulations for persons occupied in radiation work (5 rem per year; 100 mrem per 40 hours; 2.5 mrem per hour). The maximum permissible concentrations of radioactive substances inside so-called restricted areas and outside are also determined. Basically, the German Radiation Protection Regulations should not differ very much from those of other countries.

## 3. Present State of Isotope Distribution in the Federal Republic

In the years following 1955 (cancellation of restrictions in the nuclear field), especially since the Radiation Protection Regulation became law (1960), more licenses were granted for import and distribution of radioactive substances. Today these institutions hold licenses:

1. Buchler und Co., Braunschweig
2. Farbwerke Hoechst AG., Frankfurt/Main
3. Farbenfabrik Bayer AG., Leverkusen
4. Isotopenlaboratorium Dr. Sauerwein, Düsseldorf
5. Karlsruhe Nuclear Research Center, Isotope Laboratory
6. Chemical Works E. Merck, Darmstadt
7. Dr. E. Uhlhorn und Co. GmbH., Wiesbaden-Biebrich
8. Abbott Gesellschaft für pharmazeutische Präparate, Frankfurt/Main
9. Laboratory Dr. Berthold, Wildbad
10. Schering und Kahlbaum, Berlin

Some of these firms are equipped with excellent laboratories for higher activities. Since five years Farbwerke Hoechst AG. for instance are producing chemical compounds marked with radionuclides as well as radiation sources. Later on, we will refer to the installations of the Karlsruhe Nuclear Research Center as an example. Today these distributors essentially import radioisotopes from the United States, Canada, United Kingdom and France. To a much lesser extent radioisotopes are occasionally imported also from some other countries. It has to be made clear, that the Karlsruhe Research Center for an initial period only acted as a dealer of imported radioisotopes, and will in the long run concentrate only on research, leaving all trading to industry.

Because of the multitude of distributors active in the Federal Republic it is impossible to give a quantitative survey of the percentage of foreign shipments to Germany. Especially for the Karlsruhe Isotope Laboratory the Radiochemical Center of Amersham, England can be regarded as one of the main suppliers. Today the RCC is able to satisfy the demand for the primary isotopes to the largest extent. Mainly C-14 compounds as well as tritium, krypton-85, thallium-204, and more recently iridium-192 (more than 1.000 Curie per year) are purchased in Britain. The contacts to Saclay have grown into relations of similar scope. Lately Saclay has been employed also increasingly in executing irradiation orders. The irradiated substances are then processed in the Radiochemistry Institute or the Isotope Laboratory, respectively. This is mainly true of Au-198, S-35, and P-32.

Afterwards, these isotopes are shipped directly to the customer. The main customers are medical institutions and hospitals. The growing capacity of Amersham and Saclay as well as the completion of our own institutes are contributing to the fact that purchases from overseas countries, above all from the U.S. and Canada, show a slightly decreasing tendency except for multicurie sources.

4. Statistics of imported radioisotopes \*

Statistical data have been compiled on imported radionuclides since 1949. Tab. 1 shows the increase in imports.

Tab. 1: Import of Radionuclides to the Federal Republic of Germany.

Year	1949	1950	1951	1952	1953	1954	1955
Curie	50	50	80	100	170	300	450

Year	1956	1957	1958	1959	1960	1961	1962
Curie	2349	3388	11091	11077	19676	66935	44364
Curie (without multicurie sources)	814	1388	1380	2777	2876	3035	5868

Although remarkable, the increase does not quite correspond to the increase experienced in the U.S. over the same years.

A better survey of isotope demand can be gained from Tab. 2 showing the individual imports.

Tab. 3 gives a survey of the use of some familiar multicurie installations (cobalt-60 and cesium-137).

Multicurie sources are used by large industrial concerns, among other chemical industries. Thus Farbenfabriken Bayer, Farbwerke Hoechst (1000 c of Co-60), and BASF (5000 c of Co-60) each have a multicurie source of cobalt they probably use above all for radiation chemistry research.

\* Most data compiled in ten tables of this section have been collected by the Federal Ministry of Scientific Research.

Tab. 2: Quota of Individual Nuclides in the Total Import of Radioactive Substances, 1956-1962.  
(without fissile and source materials as under Sec. 2, No.1 and 2, German Atomic Energy Law)

(in Curie)

Radionuclide	1956	1957	1958	1959	1960	1961	1962
<u>1. Below 50 c</u>							
Gold-(Au-)198	71	113	136	157	177	146	259
Cobalt-(Co-)60	209	289	34	354	169	244	600
Chromium-(Cr-)51	-	-	-	-	-	5	22
Cesium-(Cs-)137	41	4	34	18	16	18	29
Iridium-(Ir-)192	386	494	884	1 067	1 339	1 277	1 362
Hydrogen-(H-)3	-	328	146	945	782	970	1 845
Iodine-(I-)131	39	50	50	53	68	60	91
Krypton-(Kr-)85	1	-	52	82	36	4	28
Phosphorus-(P-)32	14	10	10	9	11	11	21
Promethium-(Pm-)147	-	-	5	43	197	213	447
Polonium-(Po-)210	-	25	2	6	2	17	46
Radium-(Ra-)226	-	-	-	-	-	-	4
Sulfur-(S-)35	2	3	3	4	5	6	8
Antimony-(Sb-)124	-	-	-	-	16	-	2
Strontium-(Sr-)90	3	8	5	14	30	28	57
Thulium-(Tm-)170	11	-	-	2	-	1	2
Xenon-(Xe-)133	18	45	5	15	12	-	-
Yttrium-(Y-)90	-	-	-	-	-	24	1
Others	19	19	14	8	16	11	19
<b>Total:</b>	<b>814</b>	<b>1 388</b>	<b>1 380</b>	<b>2 777</b>	<b>2 876</b>	<b>3 035</b>	<b>4 303</b>
<u>2. 50 - less than 1,000 c</u>							
Cobalt-(Co-)60	-	-	-	-	-	-	900
Cesium-(Cs-)137	-	-	-	-	-	-	50
Iridium-(Ir-)192	-	-	-	-	-	-	375
Antimony-(Sb-)124 <sup>3)</sup>	-	-	-	-	-	-	240
<b>Total:</b>	<b>-</b>	<b>-</b>	<b>-</b>	<b>-</b>	<b>-</b>	<b>-</b>	<b>1 565</b>
<u>3. High-curie Equipment<sup>2)</sup> above 1,000 c</u>							
Cobalt-(Co-)60	1 535	2 000	9 711	8 300	12 300	56 400	36 496
Cesium-(Cs-)137	-	-	-	-	1 500	4 000	2 000
Antimony-(Sb-)124 <sup>3)</sup>	-	-	-	-	3 000	3 500	-
<b>Total:</b>	<b>1 535</b>	<b>2 000</b>	<b>9 711</b>	<b>8 300</b>	<b>16 800</b>	<b>63 900</b>	<b>38 496</b>
<b>Total (1,2,3):</b>	<b>2 349</b>	<b>3 388</b>	<b>11 091</b>	<b>11 077</b>	<b>19 676</b>	<b>66 935</b>	<b>44 364</b>

1) Figures for 1956-1961 are included in group 1 (below 50 c);

2) including installed closed radioactive material above 1,000 c;

3) neutron sources of antimony



Tab. 3 High-Curie-Instruments and Their Use

Year		Radiotherapy	Radiation Chemistry	Other Use	Total
1956	Number Curie	- -	1 1535	- -	1 1535
1957	Number Curie	1 2000	- -	- -	1 2000
1958	Number Curie	3 5498	3 4213	- -	6 9711
1959	Number Curie	5 8300	- -	- -	5 8300
1960	Number Curie	10 16800	- -	- -	10 16800
1961	Number Curie	10 23950	7 34450	1 2000	18 60400
1962	Number Curie	10 20500	3 16696	1 1300	14 38496
Total	Number Curie	39 74048	14 56894	2 3300	55 134246

Other multicurie sources have been installed in many research institutions, such as Max-Planck-Institut für Kohleforschung (M.P.I. for Coal Research), Institute of Physical Chemistry of Cologne University, Erdölforschungsinstitut Hannover, Karlsruhe Nuclear Research Center (10000 c of Co-60).

The consumption of low-curie amounts of radioactive substances in engineering, medicine, and research gives a better picture. Tab. 4 shows the number of users of these radionuclides as per early in 1960. Later detailed data have not yet been accumulated. On January 1, 1960 the total number of users was 1233. In the United States these figures are always higher by a factor of 10. Later figures are not known.

Tab. 4 Number of Users of Radioisotopes in Research, Medicine and Industry in 1960

Iron and steel	27
Noniron-metals	16
Chemistry and mineral oil	80
Stones, earth, glass, ceramics	7
Rubber	16
Wood, cellulose, paper	126
Textile, leather, leatheroid	22
Machines, vehicles, shipbuilding	100
Electrical engineering, precision mechanics, optics	36
Mining	8
Gas, water, electricity	22
Other industry	41
Testing stations	17
Schools	109
Research and medicine	606
<b>Total</b>	<b>1233</b>

Tab. 5a Total Number of Users of Radionuclides in Research, Medicine and Industry in the Federal Republic

Year	1957	1958	1959	1.1.1960	1.9.1960	1962
Nb. of users	143	215	507	1233	1416	4700

Tab. 5b Number of Different Radionuclides in Research, Medicine and Industry

Year	1956	1957	1958	1959	1960	1961	1962
Number	61	62	69	70	74	79	97

By the end of 1962 there were about 5000 users in research, medicine and engineering. Some 900 schools using radioactive substances must be added to this figure.

Tab. 6a and 6b show import and export values of radioisotopes. Table 7 gives import values of the Karlsruhe Isotope Laboratory. For Karlsruhe there are also data on the ad valorem shares of the supplier countries (Tab. 8).

Exact statistics about the isotopes Karlsruhe has supplied to medical and scientific institutions as well as to industry are included in Tab. 9. These data have not yet been compiled for 1962, but there has been an increase for Ir-192, C-14, H-3 and Au-198. Tab. 10 shows the total value of the turnover of the Karlsruhe Isotope Laboratory over the past few years. Tab. 11 is a statistics of the corresponding users.

Tab. 6a: Development of Import Values of Radioactive Substances (in DM 1000)

Year	artificial radioactive isotopes and their compounds	other radioactive elements, isotopes and their compounds	total
1956	549	842	1 391
1957	682	832	1 514
1958	974	1 002	1 976
1959	1 152	783	1 935
1960	1 581	13 148	14 729
1961	2 066	1 099	3 165
1962	1 331	3 228	4 559
total:	8 335	20 934	29 269

Tab. 6b: Development of Export Values of Radioactive Substances (in DM 1000)

Year	artificial radioactive isotopes and their compounds	other radioactive elements, isotopes and their compounds	total
1956	-	-	-
1957	10	65	75
1958	52	63	115
1959	50	166	216
1960	91	141	232
1961	240	149	389
1962	268	473	741
total:	711	1 057	1 768

Tab. 7: Value of Import of Radioisotopes of the Isotope Laboratory Karlsruhe

Year	1958	1959	1960	1961	1962
Value in Mio DM	406.000	517.000	555.000	572.000	632.000

Tab. 8: Percentage of Imports from Different Countries by Karlsruhe

Year	1960	1961	1962
England	54%	55%	66%
France	20%	22%	10%
USA and Canada	26%	23%	24%

#### 4. Laboratories for High-level Isotopes

In the Federal Republic as in other countries of western Europe industry and research centers have some laboratories for handling multicurie sources. To mention just a few names: Farbwerke Hoechst AG and Farbenfabriken Bayer stand for industry, the Karlsruhe and Jülich centers for research. These facilities are characterized, above all, by their number of hot cells and the curie level they can handle. The modern installations of the Karlsruhe Nuclear Research Center will be briefly described as an example.

Similar to the development in the United States the design principle of stationary, roomy, heavily shielded cells with highly developed remote-viewing and operating facilities and extensive installations dominates hot cell design in the Federal Republic. This type of facility is flexible and can be adapted to various purposes. As a rule, the cells are lined up side by side. Certain types of hot cell designs have emerged over the years.

Tab. 9 Radioisotopes in mC (without Co-60) delivered from Karlsruhe

Isotope	Medical Institutes			Scientific Institutes			Companies		
	1959	1960	1961	1959	1960	1961	1959	1960	1961
H-3	476	560	76874	259220	304960	129975	10	4	34450
C-14	21	24	22	335	393	706	345	404	595
P-32	1632	1920	2720	1802	2212	2888	391	460	4280
S-35	-	-	1955	-	-	707	-	-	2408
J-131	22796	26818	33932	403	474	3165	68	80	210
Cr-51	-	-	328	-	-	4018	-	-	5312
Cs-137	-	-	1	4446	5225	240	90	102	6526
Ir-192	90	106	4	69020	81	77880	243100	286000	651842
Au-198	21913	25780	23540	1105	1300	715	-	-	37589

Tab. 10 Total Value of Sale of the Isotope Laboratory Karlsruhe including Co-60 and some Isotopes not shown in Tab. 9

Year	DM
1958	575.000
1959	665.000
1960	803.000
1961	907.000
1962	ca.965.000

Tab. 11 Total Number of Users of Radioisotopes from Karlsruhe

Year	1959	1960	1961	1962
Medical Institutes	125	130	93	94
Research Institutes	275	300	337	346
Industrial companies	130	145	136	134
	530	575	566	574
Percentage in Germany	75 %	-	34 %	decreasing

There is a smaller type with one working place and dimensions of some 2 to 2.5 m length and depth. It is used for experiments with smaller equipment or for analytical chemical work. Mostly these cells are equipped with a Master Slave (MS) manipulator. A larger type of cell includes two working places and has dimensions of 4 to 5 m length and 2.5 to 3 m depth. Apart from one Master Slave manipulator these cells mostly include a one-arm electric power manipulator (EM) installed in a carriage running on a bridge which can be moved inside the cell. Cell height may be up to 5 m. Cells for handling gamma or beta emitters are closed on all sides, but they are not airtight. An underpressure is created by a strong ventilation system to prevent contamination from spreading. Hot cells for alpha-gamma emitters, above all for plutonium, are made gastight as a rule and can be operated on a protective gas atmosphere. Tab. 12 shows a compilation of hot cells available in the Karlsruhe and Jülich Research Centers.

The restricted area of the Karlsruhe Isotope Laboratory B 7a/b is some 1800 sq.m. The special storage cell permits storage of 45000 curies of gamma radiation of 1 MeV. The laboratory contains all facilities for calibrating samples, standard sources in particular.

##### 5. Radioisotope Production Facilities in the Federal Republic of Germany

For producing radioisotopes the Karlsruhe research reactor FR2 (12 MW,  $\phi_{\max} \approx 4 \times 10^{13}$ ) and the Jülich research reactor DIDO (10 MW,  $\phi_{\max} \approx 10^{14}$ ) will soon be available. The Karlsruhe reactor features 41 irradiation channels for isotope production; the channels extend vertically through the top shield between the fuel elements. On principle, the FR2 could supply about 1 mol of neutrons per year for isotope production. If they were used for producing Co-60 this would yield a source of some 100000c. The capacity of DIDO in Jülich will be of the same order.

Tab. 12 Hot Cells of the National Laboratories Karlsruhe and Jülich

Location	start-up	number of cells	working space, m <sup>2</sup>	max. γ-activity of 1 MeV; Curie	wall thickness; cm	wall density	remote control <sup>+</sup>	notes
Karlsruhe B 32	1964	3	7	10 <sup>6</sup>	92	4.5	MS, EM )	gastight; for investigation of fuel elements
		2	4	10 <sup>4</sup>	92	3.5	MS, EM )	
		2	4	10 <sup>5</sup>	92	4.0	MS, EM )	
		3	3	10 <sup>2</sup>	92	2.4	M	
Karlsruhe B 32a	1963	2	4	10 <sup>5</sup>	92	4.0	MS, EM )	for hot chemistry
		3	3	10 <sup>2</sup>	92	2.4	M	
Karlsruhe B 7a/b	1962	3	4	10 <sup>4</sup>	92	3.5	MS )	Isotope laboratory
		2	2	10 <sup>2</sup>	92	2.4	MS	
Karlsruhe B 36	1965	2	4	10 <sup>6</sup>	92	4.5	MS, EM	EURATOM-Transuranium Institute Investigation of Pu fuel elements -- gastight
		6	6	10 <sup>6</sup>	92	4.5	MS	
		6	6	10 <sup>4</sup>	92	3.5	MS --	
Jülich	1965	4	7	5 · 10 <sup>5</sup>	110	3.5	MS, EM	gastight } investigation of fuel elements
		2	3	10 <sup>4</sup>	85	3.5	MS, EM	
		3	8	10 <sup>4</sup>	85	3.5	EM	

+ MS = Master Slave  
EM = Electrical heavy load Manipulator



The isotope irradiation channels incorporated in the Karlsruhe reactor FR2 are aluminum tubes of 48 mm inner diameter, closed at the bottom and suspended, as I said, in the top shield in such a way that they are directly submerged in the core. These immersion tubes are closed on top by shielding plugs permeated by a multiple-coiled duct for admitting cooling air. At every shielding plug the so-called capsule carrier tube, an aluminum tube equipped with longitudinal fins, is suspended in the immersion tube. It holds the irradiation capsules with the substances to be activated. The heat generated in the capsules during irradiation is carried off by a coolant air flow. Coolant air enters the capsule carrier tube on top, cools the capsules in its downward flow and is sucked off again through the annulus between carrier tube and immersion tube. At some 30 m<sup>3</sup> per hour rate of air flow some 500 watts of heat can be carried off without the capsules heating to dangerous levels.

The irradiation capsules housing the substances to be activated are capsules of the so-called Harwell-type, i.e. screwed capsules of pure aluminum 68 mm high and 25.5 mm in internal diameter, or half capsules of 28 mm internal height and 25.5 mm inside diameter, or so-called small capsules of 10.5 mm height and 19 mm inside diameter. Three small capsules fit into a large one. One isotope channel may take up as many as 29 large capsules.

The capsule carrier tube can be withdrawn from its immersion tube without any disturbance of reactor operation, under the proper safety measures it can be transported to an adjacent auxiliary building.

There is a rabbit system for activating shortlived nuclides. The sample to be activated is pneumatically shot into and out of a horizontal channel of the reactor. A few seconds after the irradiation has been finished the sample can be investigated in a special rabbit system laboratory.

We estimated also the cost of producing some radioisotopes in the FR2 reactor. We found that for the time being we cannot produce them at the cost we are offered them from abroad. The reasons for this are not yet quite clear. It has already been mentioned that it is not our aim, to produce radioisotopes at Karlsruhe for commercial purposes. Isotope trading in future will be entirely a matter of industry in Germany.

Finally it must be mentioned that the Federal Republic like some other European countries is able to produce radionuclides in cyclotrons as well as in reactors. The following short table lists the cyclotrons used for that purpose in Europe.

Country	Location	Owner
Germany	Karlsruhe	Nuclear Research Center
Belgium	Leuven	Univ. Leuven
France	Saclay	CEA
Netherlands	Amsterdam	Philips-Dupar
Britain	Amersham	UKAEA Univ. Birmingham
Yugoslavia	Zagreb	FNEC

The Karlsruhe cyclotron is one of the most modern medium-sized particle accelerators in Europe. The Germany AEG company designed and built a so-called isochronous cyclotron with fixed frequency differing from earlier types in the layout of the magnetic field. It permits accelerating atomic nuclei up to high energies and yet achieving high beam intensity. A deuteron beam can be generated with an energy of 50 MeV at current of up to 100 microamperes. Part of it may then be directed out of the cyclotron as a focused beam. Protons can be accelerated up to 25 MeV, alpha particles up to 100 MeV. In producing radionuclides the cyclotron is used in particular for generating positron emitters. The high current intensity, roughly fifty times higher than in conventional cyclotron designs, permits to make the target also an intense neutron emitter with a maximum source power of  $10^{14}$  neutrons per second whose energy may attain 50 MeV.

Practically all radionuclides can be produced in the cyclotron. Usually the occurring reactions are not  $n-\gamma$  reactions, as in a reactor, but reactions where deuterons or  $\alpha$ -particles are shot onto a target; for this reason the produced radionuclide differs from the target substance and can be separated by chemical processes. This makes for the high specific activities of the radionuclides produced in a cyclotron. Above all, the Karlsruhe researchers think of Fe-55, Na-22 and Mn-54. Tab. 13 shows the advantages and the drawbacks in producing radionuclides in a reactor or a cyclotron, respectively.

To round off the picture it must also be pointed out that the European Atomic Community (EURATOM) has started to collect a stock of radioactively labeled organic compounds not commercially available under normal circumstances which will be at the disposal of users within the Community. EURATOM has made out contracts favoring those interested in these compounds; parties to the contracts are laboratories producing these compounds for research of their own. Tab. 14 lists the samples labeled with carbon-14 and tritium as far as they are available now.

The research laboratories treat the production of these compounds only as a sideline. In case it should become evident that the demand exceeds the production facilities of the research laboratories, the EURATOM commission jointly with industry will attempt to satisfy the demand. The samples listed in the table are then stored in the producers' laboratories, and they are shipped upon advice of the EURATOM commission. The prices quoted include neither costs of shipping nor customs or other duties, if any. The samples must be ordered in all cases with EURATOM (Labeled Molecules Department) in Brussels.

The Radiochemistry Institute of the Karlsruhe Nuclear Research Center has also started to produce labeled compounds, mainly those containing C-14 and tritium. Highly specific, radiochemically pure nuclides of phosphorus-32, sulphur-35, chromium-51, iodine-131, and gold-198 were produced by special methods worked out in the Radiochemistry Institute. Many more labeled compounds are available through German industry, e.g. Farbwerke Hoechst, Frankfurt and Röhm and Haas, Darmstadt.

For instance, Farbwerke Hoechst A.G., Germany, has available for sale the following labeled compounds:

- |   |   |
|---|---|
| d, 1-Campher-(7- <sup>14</sup> C)                                   | 2,4-Dichlortoluol-5-sulfamid- <sup>35</sup> S   |
| 2,5 Diaminotoluol- <sup>14</sup> C                                  | Trevira- <sup>14</sup> C-Folie                  |
| 3,4 Dichlorbenzoesäure-(7- <sup>14</sup> C)                         | Blausäure- <sup>14</sup> C, wasserfrei          |
| Hexamethylentetramin- <sup>14</sup> C                               | Zinntetrachlorid- <sup>113</sup> Sn, wasserfrei |
| d,1-Isoborneol-(7- <sup>14</sup> C)                                 | Bis-tri-n-butylzinn-oxyd- <sup>113</sup> Sn     |
| d,1-Isobornylacetat-(7- <sup>14</sup> C)                            | Tri-n-butylzinnbenzoat- <sup>113</sup> Sn       |
| 1,1-Methylcyclohexanol-(1- <sup>14</sup> C)                         | Tetraphenylzinn- <sup>113</sup> Sn              |
| 5-Nitro-2-aminotoluol-(7- <sup>14</sup> C)                          | Triphenylzinnacetat- <sup>113</sup> Sn          |
| Phenylacetone-(carboxyl- <sup>14</sup> C)                           |   |
| Phenylthioharnstoff- <sup>14</sup> C                                |   |
| Phtalsäure-di-n-butylester- <sup>14</sup> C                         |   |
| Phtalsäure-di-cyclohexylester(-carboxyl- <sup>14</sup> C)           |   |
| Reverin- <sup>14</sup> C (Pyrrolidino-methyl-tetracyclin)           |   |
| Sorbinsäure-(1- <sup>14</sup> C)                                    |   |
| Sorbinsäure-(2- <sup>14</sup> C)                                    |   |
| Terephthalsäuredimethylester-(carboxyl- <sup>14</sup> C)            |   |
| Trichloressigsäure-(1- <sup>14</sup> C)                             |   |
| Trichloressigsäure-(2- <sup>14</sup> C)                             |   |
| Tritriakontan-(17- <sup>14</sup> C)                                 |   |
| Rastinon- <sup>35</sup> S (N-p-Toluolsulfonyl-N'-n-butyl-harnstoff) |   |
| Thiodan- <sup>35</sup> S  |   |
| Ruthenocen- <sup>106</sup> Ru (Dicyclopentadienylruthenium)         |   |
| Fluoressigsäure-(1- <sup>14</sup> C), Na-Salz                       |   |
| Fluoressigsäureäthylester-(carboxyl- <sup>14</sup> C)               |   |
| Fluoracetamid-(1- <sup>14</sup> C)                                  |   |
| Acetylpyridin-(carbonyl- <sup>14</sup> C)                           |   |
| Diphenylsulfon- <sup>35</sup> S                                     |   |
| Dimethylsulfat- <sup>14</sup> C                                     |   |
| Nicotinsäureäthylester-(carboxyl- <sup>14</sup> C)                  |   |
| Cyanessigsäure-(3- <sup>14</sup> C), krist. (Ebenso                 | -(2- <sup>14</sup> C und                        |
|   | -(1- <sup>14</sup> C)                           |

Tab.13 Comparison of Advantages and Disadvantages of Isotope Production in the Reactor and in the Cyclotron

Reactor	Cyclotron
1. Very high total activity can be produced, but with low specific activity.	1. The total activity that can be produced is much lower, but specific activity is always very high.
2. Specific activity depends upon the ratio between irradiation and halflife and upon neutron flux <sup>*</sup> . Yet, both the fission products and for instance T, C-14, P-32, S-35 and I-131 can be produced with a maximum specific activity.	2. Specific activity is independent of irradiation time, halflife, cross section and flow of projectile.
3. Number of radionuclides that can be produced is relatively small.	3. Practically all radionuclides can be produced.
4. No special requirements of the target material.	4. Target material must be highly resistant to heat and vacuum and show good thermal conductivity properties.
5. In general, no chemical processing is necessary, unless high specific activities of the nuclides shown under (2) are required.	5. Chemical processing is nearly always required, as almost in all cases a mixture of different radionuclides is produced in the target.
6. Many samples can be irradiated in a reactor at the same time.	6. In a cyclotron each target must be handled and irradiated individually.
7. During irradiation for radioisotopes the reactor can also be used for other purposes.	7. The cyclotron is practically not available for other experiments and irradiation processes.
8. Irradiation costs are relatively low not considering the reactor facility and operating costs.	8. Special irradiation costs in a cyclotron are high, even considering neither facility nor overall operating costs.
9. Per-curie price is relatively low.	9. Per-curie price is relatively high.
10. High specific activity cannot be produced for many radionuclides or is very expensive due to separation afterwards.	10. Maximum specific activity can be produced for all nuclides easily and at relatively low cost.

\* and cross-section

Radioactively labeled Molecules at the disposal of Euratom

Tab. 14

Carbon-14-labeled Molecules

Compound	Max.spec. activity, mc/mM x)	Price		Laboratory
		1 $\mu$ c x)	1 mc x)	
Conalbumin)	10	3.200 bfrs		Service de Radiobiologie du CEN (M.Ledoux) Geel, Belgium
Lysozyme ) glycocoll-	10	3.070 "		
Ovalbumin ) labeled	5	1.600 "		
Ovomucoid )	5	3.200 "		
C-14-Puryl-6-Histamine	0,01		4.000 DM	Institut für Krebsforschung (Cancer Research Institute), University of Heidelberg (Prof.Lettré), Federal Republic of Germany
C-14-Puryl-6-Lysine	0,01		4.000 DM	
Puryl-6-C-14-Histamine	0,02		8.000 DM	
Puryl-6-C-14-Lysine	0,02		14.000 DM	
C-14-Puryl-6-C-14-Histamine	0,04		25.000 DM	
C-14-Puryl-6-C-14-Lysine	0,04		50.000 DM	
C-14 (Methyl)-Nicotine	1,5		4.500 DM	Physiologisch-Chemisches Institut(Prof.K.Decker), (Physiological Chemistry Institute), University of Freiburg, Federal Republic of Germany
C-14-2-Nicotine	2		5.000 DM	

Tritium-labeled Molecules

Compound	Max.spec. activity, mc/mM x)	Price		Laboratory
		1 $\mu$ c x)	1 mc x)	
Avidin ) labeled with	5	5.200 bfrs		Service de Radiobiologie du CEN (M.Ledoux) Geel, Belgium
Conalbumin) one of these	10	6.200 "		
Lysozyme ) amino acids:	10	6.100 "		
Ovalbumin ) Arginine, Tryp-	5	1.700 "		
Ovomucoid ) tophane, Tyrosine	5	3.200 "		
3,5-diiodo thyronine				Laboratoire de Biochimie Générale et Comparée du Collège de France (Prof.Roche), Paris 5 <sup>e</sup>
H-3-alpha-beta	100		140 NF	
3,5,3-triiodo thyronine				
H-3-alpha-beta	100		1.080 NF	
3,5,3',5'-tetraiodo thyronine H-3-alpha-beta	100		1.080 NF	
Estradiol H-3-alpha-beta	190 mc/mg		11 NF	

x) Note: mM = Millimol; mc = Millicurie,  $\mu$ c = Microcurie (i.e. one thousandth or one millionth, respectively, of one curie (c). 1 c corresponds to the activity of 1 g of radium).

Tab. 15

## Use of Radioactive Substances as Sources of Radiation

Range of Utilization		Radionuclides used	Activities required	Special Utilization
Control Engineering	Filling level control	Co-60, Cs-137	1 - 100 mc	Paper, foils, metal layers
	Measurement of thickness	Co-60, Sr-90, Cs-137, C-14, Tl-204	1 - 10 mc	Gases and liquids
	Measurement of density	Co-60, Sr-90, Kr-85, Tl-204	1 - 100 mc	Ar, S, C-H-ratio
	Absorption analyses	Fe-55, Sr-90, Am-241, H-3, Sr-90 (Bremsstrahlung)	1 - 30 mc	U, heavy metals
Materials Testing	Gammagraphy	Na-24, Co-60, La-140, Cs-137, Eu-155, Ir-192, Tm-170	0,1 - 5 c	Inspection of welding seams, search for cavities
Energy Production	Sources of light	H-3, Kr-85, Sr-90	1 - 1000 mc	Luminescent dyes, glow discharge lamp, emergency lighting, 3.000 microlambert per curie,
	Electricity generation	Sr-90, Ce-144, Po-210	10 - 3000 c	batteries of SNAP-program, e.g. 2,5 W (1500 c Po), 10 W (47 000 c Sr-90), 60 W (280 000 c Sr-90)
Radiation Chemistry	Polymerization Radiolysis Synthesis Solid-state irradiation	Co-60, Cs-137 spent fuel	$10^2 - 10^5$ c	Olefins and vinyl compounds cracking of hydrocarbons phenol, chlorination, linking activation of catalyts
Radio-biology	Pest control Sterilization Genetics	Co-60, Cs-137	$10^3 - 10^5$ c -2000 c	Irradiation of insects, potatoes, mutations
Radiology	Teletherapy	Co-60, Cs-137, Eu-152, Ir-192	100 - 10 000 c	Cancer therapy

## 6. On the Use of Radionuclides

It is impossible within the scope of this paper even to touch upon the multitude of operations using radionuclides in research, medicine, agriculture, and engineering. Moreover, the general principles of application will be familiar to the reader. It is a matter of applying a radionuclide as a source of radiation, as a tracer isotope or an indicator as well as an aid in dating events and processes. Tab. 15 indicates the use of radionuclides as a source of radiation, the nuclides that can be used for this purpose and the activities required.

Most of these applications are pursued also in the Federal Republic of Germany.

Let me quote you an example of radioisotope application in engineering. This is a brief sketch of an experiment recently made at Karlsruhe. The task was to observe simultaneously the rotary motions of three piston rings in a one-cylinder diesel engine. For this purpose 2 mm long, closed silver pins containing the gamma emitter Ag-110, half life 270 days, were inserted into the gaps of the three piston rings; the pins were activated in a ratio of 1:2:3.

For scanning the intensity curve of gamma radiation along the outer wall of the cylinder a sharply focused scintillation counter rotated around the cylinder at a period of 30 seconds. In favorable positions the counting rate was of the order of 1000 pulses/sec. For determining the rotations of the piston rings on principle the intensity curve of gamma radiation was taken as it was recorded as a function of the angular position of the detector at every revolution of the detector. Evaluation was much simplified by the fact that not the intensity curves taken at every detector revolution but the angular position of the three gaps were registered directly over the time. For this purpose a potentiometer moves synchronous with the detector; its output voltage changes linearly between 0 and  $360^\circ$ . Each value of voltage corresponds to a certain angular position. The measurement of intensity is linked with the potentiometer output voltage by an electronic circuit in such a way that the voltage values of the potentiometer are



briefly tapped whenever there is a maximum in intensity. These voltage pulses are recorded over the time of the experiment by a recorder so that the angular position of the piston ring can be directly read as a function of time. By means of electronic circuits it is possible to separate the different peaks of intensity of the three piston rings and record them individually.

## 7. Future Aspects

As has been said before it has become increasingly difficult over the past few years to keep track of the use of radioisotopes in the Federal Republic. This is due to the increasing number of importers, processors, and users. It is not even easy to get a survey of the present trend of development. Even more difficult is the attempt of an extrapolation to the future.

It may be expected that the coming years will bring an increase in the consumption of radionuclides, by value as well as by number. Most probably the increase will be larger in industry than in scientific institutions.

The start of our own production in the research reactors at Karlsruhe and Jülich will give a strong impetus especially because shortlived isotopes can then be processed and even unconventional demands for irradiation can be met. Whether or not production of radioisotopes in our own country will influence the economic side of isotope use remains open to debate. I hinted our impression that at present we will not be able to produce radioisotopes at the price foreign countries offer them. Whether this is due to especially high production costs in the Federal Republic or whether foreign isotope prices are in some way government subsidized is a question only the future can answer.

Lately there has been concern about the fact that one of the large German companies up to that time active in the distribution of radionuclides seems to retreat from the business. It is unclear whether the reason is to be found in economic considerations.

Good arguments in favor of an increase in work with radioactive isotopes are the considerable research capacities created for that very purpose. In the Federal Republic there is Isotopenstudiengesellschaft (Isotope Studies Company) of Frankfurt/Main, operating a research institution of its own on the premises of the Karlsruhe Nuclear Research Center for investigating the possibilities of isotope application in engineering. The company is expanding this institute just now.

Near Munich the Federal Ministry of Scientific Research has built an "Experimental and Training Facility for Health Physics" working along the same lines in the medical field.

The Federal Ministry of Food and Agriculture starts building an institute for "Radiation Technology of Foodstuffs" in Karlsruhe which is going to work in close cooperation with the Nuclear Research Center. All these institutes have been built on a large-scale basis and are equipped with considerable funds; their aim lies not only in using radioactive isotopes themselves but in promoting and propagating their use.

These positive arguments for a future development are opposed by negative ones. Let me quote just one example: The problem of transporting radioactive substances. Shipment and transportation are subject to the regulations laid down in the first Radiation Safety Regulations. Under that law any transport of radioactive substances over public traffic routes or routes open to the public is subject to approval. No permission under the terms of the Regulation is necessary, however, if the material is shipped by railroad under the rules of the Railroad Traffic Regulations (Eisenbahnverkehrsordnung). Besides, no permission is required, if the material is shipped by airplane subject to the Air Traffic Regulations in connection with I.A.T.A. rules on "Carriage of Restricted Articles by Air". Transporting radioactive substances by sea ship is not subject to permission, if the transport is carried out under the regulations covering transport of dangerous freight goods at sea. A permit for shipping thus is only necessary for transport by road vehicles and inland ships. As a rule this permit will be granted without any difficulties if certain conditions regarding safety, above all regarding indemnification provisions are met for meeting legal claims for damages. On principle the transportation of some radioactive substance by a pedestrian is subject to permission even if the level of acti-

vity is within the so-called unrestricted levels. For shipment by post only radioactive substances and objects are permitted whose transportation does not require permission under the first Radiation Safety Regulations.

The majority of transports are carried out by airplane or railroad express. Lately transport by railroad has become increasingly difficult so that there has been the impression that the German railroads are no longer interested in shipping radioactive substances. The railroad regulations demand a reduction of the dose rate at the outer surface of the piece to be shipped to such a level<sup>+) that this requires a massive lead shield even for very low acti-</sup> vities (of the order of 1 to 10 mc), which would render the piece so heavy that it would exceed the upper limit for express freight matter (50 kgs.). That is to say, activities of more than 10 mc can no longer be shipped by express freight. Transportation by normal goods train as a rule is so tedious that it would be longer than the short halflives of some isotopes permit. This would have as a consequence, among other things, that above all in the medical field the latest methods of diagnosis and therapy could not be put to any practical use, if the isotopes would have to be procured from abroad.

Recently the German railroads have temporarily released these rigid regulations under public pressure, but every shipment has to be subjected to special formalities when handed in, which again wastes a lot of time and increases costs of shipping. At the same time the express freight rates for radioactive substances were substantially raised. Moreover, the German railroads now even require that indemnification is provided for meeting claims for damages which might arise for the railroads or their officials in transport. This indemnification must be extended to damage caused to the railroads themselves. This special third-party liability coverage is even required of large firms which are already covered for radiation damage by their regular liability insurances. It would be more sensible if the German railroads were to obtain coverage against all liability claims arising out of shipments of radioactive substances by a global insurance of their own.

Experts are afraid that these conditions of the railroad company, unless they are waived, might increase the cost of using radioactive isotopes, especially in medicine, to such an extent that this use would stop altogether to a certain degree. Moreover, claims of such an exaggerated kind must necessarily cause

+) (about 10 mr/day)

apprehension with other shipping agents about the dangerous nature of radioactive substances, thus bringing about new claims for security by far exceeding actual risks.

These difficulties in transport were quoted to illustrate that not all signs are positive for the future use of isotopes in the Federal Republic. These hardships may be called difficulties in the adjustment stage and one can only hope that they will be overcome as time passes.

### References

1. G. Böhme, W. Köhler. Kerntechnik, 4 (1962) p.580  
"Forschungslaboratorien für hochradioaktive Stoffe"
2. H. Götte. Atomwirtschaft (1961)  
"Bedeutung der radioaktiven Stoffe in der Technik".
3. K. Hogrebe. Atomwirtschaft (1958) p.311; (1961) p.483  
"Produktion und Anwendung von Radionukliden".
4. H. Hossner. Atomwirtschaft (1959) p.245  
"Die Medizin als Isotopenverbraucher".
5. K. Kollmann, D. Stegemann, W. Kaspar-Sickermann. Motortechnische Zeitung 24 (1963) p.73 "Drehbewegung von Kolbenringen".
6. K. Sauer. Atomwirtschaft (1960) p.546; (1961) p.589  
"Einfuhr und Verwendung von Radioisotopen in Deutschland".
7. W. Seelmann-Eggebert. Atomwirtschaft (1961) p.273  
"Anwendung des Zyklotrons zur Herstellung von Radionukliden".
8. W. Seelmann-Eggebert, C. Keller, G. Zundel. Bericht Karlsruhe KFK 41  
"Abtrennung kurzlebiger Nuklide".
9. K. Zimen. Atomwirtschaft (1961) p.91  
"Bedeutung der Kernchemie".