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# Radiation Laboratory

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Radiation Laboratory

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MONTHLY PROGRESS REPORT NO. 60 FOR APRIL 1948

Berkeley, California

## UNIVERSITY OF CALIFORNIA, RADIATION LABORATORY

MONTHLY PROGRESS REPORT NO. 60 FOR APRIL 1948

1. 184-inch Cyclotron Program

The cyclotron was used for research experiments ninety-two per cent of the 483 hours that the crew was on duty. Work on mesons and delayed neutrons used the bulk of this time.

A magnetic deflector was installed to enable mesons, produced inside the cyclotron tank, to emerge from the vacuum system of the cyclotron.

During this month, the power situation improved sufficiently to permit normal operation of the 184-inch cyclotron (8:00 a.m. through 12:00 midnight, seven days a week).

2. 60-inch Cyclotron Program

Power conservation did not curtail activities to any great extent. Efficiency of operation was relatively high throughout the month. Considerable time was devoted to research on ion sources.

3. Synchrotron Program

The magnetic testing program has been continued during the past month and it is believed that the field is now satisfactory for operation. The duration of the "betatron period" is still not as long as desired, and it may be necessary to turn on the oscillator at a time somewhat earlier than that corresponding to 2 Mev. Since this is an operating variable, the best conditions will be determined during operation.

A considerable time was spent adjusting the phase variation of zero field. Variations of about 6 gauss were observed which occurred as a high harmonic of the orbital frequency. It is believed that these large variations will do no damage so long as the order of the harmonic is high. Their presence, however, has made the investigation of the low order harmonics (first, second, and third) very difficult. The design tolerance on the first harmonic amplitude is 1/10 gauss, and it has been necessary to make a Fourier analysis of the field versus azimuth after each correction has been added to the shading coils. A method has been developed for quickly making this analysis. The first three harmonics have been reduced to permissible amplitudes.

The excitation equipment still does not perform satisfactorily. An engineer from the manufacturing company has been conducting tests on the equipment with the view to preventing mis-firing and arc-backs as well as to incorporating protective devices which will prevent damage to the tubes in case either of these events occurs. These tests are still in progress, and it is not yet possible to determine what changes will be necessary.

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4. Linear Accelerator Program

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Van de Graaff Generator. During the month of April the Van de Graaff generator delivered a 4 million volt beam for a total of 55 hours. This is somewhat less than the preceding month, due principally to a leak in the accelerating tube assembly, a bearing failure on the main belt pulley, and the necessity for replacing the charging belt.

Comparative tests between litton and silicone oils in the refrigerated baffle test setup mentioned last month showed that litton oil is superior. The silicone oil in the pump was replaced with litton oil and a definite improvement in operation was noted. Circulation of ice water through the heat exchanger is still necessary for prolonged operation. To find a satisfactory permanent solution to the problem, tests of an activated charcoal trap are being made and preliminary design work on a direct driven freon compressor has been started.

The ion source pulser was operated successfully for the first time on April 19. The output of the linear accelerator was increased from about  $10^{-11}$  amps to  $3 \times 10^{-10}$ . Rough measurements indicate total pulse currents of about 100 $\mu$ a from the Van de Graaff generator. At the present time it is not possible to take advantage of the full output of the ion source assembly because the overall focusing of the system at 4 million volts is not quite right when the ion source voltages are set to give maximum input into the tube. Under these conditions the beam is between one and two inches in diameter when it emerges from the accelerating tube which means that only a fraction of it can be accelerated by the linear accelerator.

In an attempt to correct this focusing difficulty, the strength of the first lens in the accelerating tube is being decreased by installation of a grid on the positive side. This grid has just been installed and its effect has not yet been tested.

Linear Accelerator. During the available beam time a total of 25 bombardments were made, exclusive of beam measurements. Of these, about 80 per cent were for short half life experiments, 15 per cent on inelastic scattering, and 5 per cent on activation curves.

5. Experimental Physics

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Film Program and Meson Experiments. Experiments on the artificially produced mesons previously reported continue. Strenuous efforts are being made to reduce background and some progress has been made in this direction. It is found that positive mesons can be observed in the plane of the circulating ion beam in the case of those mesons emitted in the backward direction with relation to the ion beam. These mesons can thus be observed on the same plate as the negative mesons by appropriate changes of the shielding.

Mass determinations have been made by the grain count method in addition to the  $H\alpha$ -range determinations previously reported. The results confirm the assignment of masses.

Of 206 heavy mesons studied, 85 per cent give visible stars with number of prongs ranging up to 6 but with a maximum at 1. Of 39 light mesons, 40 per cent give stars of up to 3 prongs.

Work on bringing the mesons clear of the main vacuum chamber by means of a magnetic shield has been successful in preliminary experiments.

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Counter Development. In agreement with results reported by R. Bell of Clinton Laboratories, it is found that anthracene is markedly superior to naphthalene as a "moth-ball" counter material. This is sufficient to obviate the use of liquid air or other coolants for the photomultiplier tube. Moreover over-voltaging the later stages of the tube results in much larger pulses (up to 20 volts from the photomultiplier alone) than are normally available.

Cloud Chamber. Work continues on the neutron-proton scattering studies. A run has been made to estimate the energy of the neutrons emitted from  $O^{17}$  following the decay of  $N^{17}$ ; the results are not in contradiction with those otherwise obtained. Preparations are being made for meson studies of the deflected mesons which it is hoped will be shortly available as indicated above.

Scattering of 90 Mev Neutrons. Experiments to resolve the difference between the differential scattering cross sections as measured with either carbon detector or coincidence proportional counters and those predicted by diffraction from nuclei treated as opaque obstacles continued. Measurements with either of the detectors thus far show intensities in the scattering angles investigated ( $0^\circ - 10^\circ$ ) higher than simple diffraction theory would indicate.

Other Experimental Work . Assembly and component testing of the proton-proton equipment for the photographic method and for the proportional counter method are progressing. Both sets of equipment may be ready to use some time in the coming month. Bombardments have been made with the linear accelerator for an inelastic scattering experiment of protons on aluminum foil, using photographic plates as detectors.

Bombardments for short half life measurements have continued. Proton bombardments of  $Mg^{24}$ ,  $Mg^{25}$  and  $Mg^{26}$  show that the half life of  $Al^{26}$  is approximately 8 seconds, rather than the 7.0 seconds according to the literature, and that the half life of  $Al^{25}$  is approximately 6 seconds and that there is an activity of approximately 20-25 seconds half life which can be assigned to  $Na^{21}$ . Several other light elements are being investigated for new activities. Activation curves for  $C^{12}(p,pn)C^{11}$ , and for  $B^{11}(p,n)C^{11}$  have been obtained.

The n-p scattering measurements have been extended to  $71.6^\circ$  with emphasis on the range  $45^\circ$  to  $71.6^\circ$ . The measurements indicate that in the center of mass system the intensity of scattered protons increases in the backward direction, implying that there is some "ordinary" scattering in addition to the "exchange" scattering already found.

Experiments have continued on delayed neutrons, on total cross section measurements with bismuth fission counters, and on the detection of mesons in an ionization chamber containing plates of fissionable material.

6. Theoretical Physics

A fairly good fit to the observed n-p scattering has been achieved by using a long-range tensor force. Further investigation of such a force is continuing. Calculations have been made of the energy and angular dependence of mesons produced in encounters between nucleons. Estimates have also been made of the production of mesons by the synchrotron, and it appears that the background due to pair formation should not be too bad. Calculations are in progress on the yields of various nuclear reactions, e.g., the reactions leading to  $N^{17}$ .

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7. Chemistry~~SECRET~~

## Part A

Radioactive Properties of Heavy Isotopes. The new collateral chain of the uranium series starting with Pa<sup>226</sup> and joining the uranium series at RaE has been somewhat further characterized. The decay chain is governed by the 38 minute half-life of the alpha emitter Pa<sup>226</sup>. The daughter of Pa<sup>226</sup>, Ac<sup>222</sup>, has been separated and found to have a 3 minute half-life. The two long range activities, presumably due to Fr<sup>218</sup> and At<sup>214</sup> are apparently too short to measure by chemical separation. A small amount of orbital electron branching (0.5 - 3 per cent) of the Pa<sup>226</sup> was noted by removing the daughter product Th<sup>226</sup>.

The above activities were made in the bombardment of thorium with deuterons in the energy range 60-70 Mev. At the same time there is produced an alpha emitter of about 22 hour half life attributed to Pa<sup>228</sup>. This activity has been found to decay principally by orbital electron capture as determined by comparing the new alpha activity at 6.09 Mev with the amount of Th<sup>228</sup> which grows into the sample. The alpha branching is probably in the range 1 - 5 per cent.

Following the bombardment of Am<sup>241</sup> with 38 Mev helium ions a new alpha emitter of energy 5.8 Mev was found and since it appeared in the Cm fraction from the adsorption column separation it has been tentatively assigned to Cm<sup>243</sup>. If it is assumed that the yield in the formation of this isotope is the same as that to form the 5 month Cm<sup>242</sup>, the half life becomes about ten years.

A long lived isomer of Am<sup>242</sup> has been previously reported following the neutron irradiation of Am<sup>241</sup>. It is known that it decays by alpha emission since Np<sup>238</sup> grows into it. It also has associated with it considerable amount of  $\beta^-$  activity. The growth of Cm<sup>242</sup> from the  $\beta^-$ -decay process has now been observed.

Fission of Bismuth with High Energy Particles. The yields of three bismuth products have been determined following the irradiation of bismuth with 100 and 200 Mev deuterons and 400 Mev helium ions. The three isotopes chosen for analysis correspond to positions on the light wing, heavy wing, and peak of the fission product distribution curve. The data are summarized in the following table:

Fission of Bi with Particles of Different Energies

	100 Mev D+	200 Mev D+	400 Mev He++
	$\sigma$ (mb)	$\sigma$ (mb)	$\sigma$ (mb)
Cu <sup>67</sup>	0.3	0.6	2.3
Mo <sup>99</sup>	8.	8.	18.
Ba <sup>133</sup>	0.1	0.3	1.

It may be seen that there are no great differences in the yields over this wide energy range. The only trend discernible is that the yields of the isotopes on the wings appear to increase somewhat relative to that at the peak as the energy increases.

According to the proposed mechanism for the fission of bismuth in which it is assumed that the compound nucleus loses many neutrons before arriving at a light nucleus in which the fission reaction predominates, it would be expected that the threshold for the fission of Pb<sup>204</sup> would be lower than that for Pb<sup>208</sup>. Samples of isotopically enriched Pb<sup>204</sup> and Pb<sup>208</sup> were irradiated with 100 Mev deuterons and

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75 Mev deuterons and the relative fission yields of Cu<sup>67</sup>, Mo<sup>99</sup> and Ba<sup>133</sup> were determined. It was found in all three isotopes that the yields were 2-1/2 - 4 times higher for the lead target which contained 27 per cent Pb<sup>204</sup> than one which contained 90 per cent Pb<sup>208</sup>. If reasonable guesses are made for the contributions made by the other lead isotopes present (Pb<sup>206</sup> and Pb<sup>207</sup>), it is found that the cross section for fission of Pb<sup>204</sup> at these energies is 5 - 10 times higher than that of Pb<sup>208</sup>.

Chemistry

Part B

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Synthetic and Experimental Chemistry. Studies on the synthesis of labeled choline  $[(CH_3)_3NCH_2CH_2OH]$  OH, methylene-labeled malonic acid,  $CH_2(CO_2Et)_2$ , and ring-labeled phthalic acid are beginning this month. The study of the synthesis of anthranilic acid, hydroxyanthranilic acid, and stilbamidine is being continued.

Malonic acid, synthesized from active KCN and dead chloroacetic acid, has been decarboxylated with the hope of noting an isotope effect; such an effect was detected. The carboxyl-labeled malonic acid was decomposed by heating at its melting point. The experiments were carried out on a 1-2.2 millimole scale. The experiments show that out of one hundred decarboxylation events approximately forty-five involve rupture of a C<sup>12</sup> - C<sup>14</sup> bond. Thus, the relative frequencies of C<sup>12</sup> - C<sup>12</sup> and C<sup>12</sup> - C<sup>14</sup> rupture are 55/45 or 1.22 instead of 50/50 or 1.00 which would be the case if rupture of either bond was equally probable. The figures just quoted are based on the ratio of specific activity of carbon dioxide and malonic acid for four experiments. When the calculation is based on the ratio of carbon dioxide and acetic acid activities, the frequency ratio is 47/53 or 1.13. These experiments are to be repeated on a larger scale in order that the difference between the acetate basis and carbon dioxide basis values can be resolved. Experiments are also to be carried out on various substituted malonic acids.

Biological Chemistry. The studies on the metabolism of carbon 14 labeled dibenzanthracene, tyrosine, tryptophane, and dihydroxyphenylalanine are continuing. Preliminary results on the metabolites produced after injection of labeled dibenzanthracene into mice have been inconclusive, and effort this month has mostly been directed toward the building up of an adequate supply of these compounds so that further work on their identification may continue. Radioactive kynurenic acid has been isolated in the urine of dogs fed labeled tryptophane, and this compound is being degraded to determine the positions of carbon labeling.

In the continuation of the study of the metabolism of labeled tyrosine, it has been found that the uptake of DL-tyrosine by tissues is extremely rapid. This extremely rapid appearance of activity and the high specific activity of the material suggests that much of the initial output may be unchanged tyrosine, possibly predominantly the D form. Of the specimens examined, the intestines, kidneys, plasma, tumor, and lymph nodes showed the highest specific activity. Comparatively little (2 per cent) of the administered dose was expired in this interval.

Photosynthetic Chemistry. In the fixation of carbon dioxide by photosynthesizing green algae it has been shown that aspartic acid is the major (greater than 70 per cent) amino acid produced in the first thirty-second period of photosynthesis. The oxidative degradation procedure which has been developed in this laboratory for labeled malic acid has been adapted to degrade this labeled aspartic acid. With aspartic acid synthesized during thirty seconds by *Scenedesmus*, 4 per cent of the activity was found in the central carbon atoms and 96 per cent in the carboxyl groups. This distribution of radioactivity corresponds closely to that observed in the laboratory in the case of malic acid synthesized in the same experiments.

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The effect of changing the nitrogen source from a nitrate to ammonium ion in the determination of the growth rates of carbon dioxide reduced by *Chlorella* has been investigated. There was no marked influence on the rate from the use of the more reducing form of nitrogen.

Investigations were undertaken to determine the dependence of the rate of dark fixation of carbon dioxide by pre-illuminated *Scenedesmus* on the partial pressure of the labeled carbon dioxide. The result is strikingly similar to the dependence of photosynthesis steady state rate on carbon dioxide partial pressure.

Studies on the metabolism of carboxyl-labeled acetate and *Scenedesmus* were continued. The major part of the activity in the acetate after forty minutes illumination in a Warburg vessel is present in very complex plant material. After twenty-four hours hydrolysis in 6N sulfuric acid solution, the activity is still in the residual material, and only after alkaline hydrolysis is the labeled material extractable in ethanol.

A study is being continued on the relation of deuterium to chlorophyll photosynthesis and on the oxidative degradation of methyl glucoside which is part of the program for the complete determination of the distribution of activity in plant synthesized glucose.

Chemistry~~SECRET~~

Part C      Subproject 48 B

Metals and High Temperature Thermodynamics. Work is in progress on the following problems:

1. Thermodynamics of CN.
2. Thermodynamics of gaseous molybdenum and copper halides.
3. Absorption coefficients of species in sun.
4. Low melting metal alloys.
5. Refractory studies.
6. Heats of formation of Na-Sn and Li-Sn alloys.
7. Theory of the solid state.
8. Report on thermodynamics of the oxides.

Basic Chemistry. Solvent Extraction. The following problems are under investigation

1. The aqueous chemistry of zirconium.
2. Hydrolysis of uranyl ion and the formation of a uranyl- TTA chelate in aqueous solutions.
3. Complex ion formation of lanthanum.
4. Exchange reaction between iodine and iodate ion.
5. Solvent extraction of uranyl salts.

8. Medical Physics~~RESTRICTED~~

Part A Project 48 A-I

Radioautographic studies with radio-yttrium are being continued. A detailed analysis of previously studied elements is being made and time has been spent in teaching a new assistant certain of the radioautographic techniques.

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During the month of April, metabolic tracer studies in rats have been set up with radio-europium. Additional studies involving the mechanisms by which radio-cadmium is eliminated from the liver have been initiated. Work is continuing on the metabolism of  $U^{230}$  in rats. The 64 day intramuscular studies with radio-zirconium have been completed showing the typical Zr deposition in animals as reported earlier. Carrier-free radio-vanadium studies have been set up. The 1 and 4 day intramuscular studies are complete and show that the radio-vanadium is eliminated primarily and rather rapidly by the urine. Also measurable amounts of activity are to be found in large organs such as bone, muscle, liver, and kidney. Less than one-tenth of one per cent of radio-vanadium is absorbed after stomach tube administration.

Studies showing the comparison of the deposition in rats of radio-lanthanum in the carrier-free state and with lanthanum carrier, as well as lanthanum in the carrier-free state complexed with citrate, and in addition with carrier-complexed with citrate have been completed. Citrate allows the body to mobilize carrier-free lanthanum more readily than when it is administered without a complexing agent. This effect is not nearly so great when lanthanum carrier is added.

Kinetic studies of the uptake of radioactive strontium by the skeleton are being extended from the initial 1 hour period up to 48 hours. Radioautographs are being prepared to determine whether any shifts in skeletal distribution occurs during this initial period.

A comparison of the metabolism of carrier-free radioactive calcium and strontium indicates a great similarity in the biological behavior of these two elements, with a somewhat higher bone retention and a lower excretion in both urine and feces in the case of calcium.

Studies are being continued on the rickets produced by severe phosphorus deficiency, and on that produced by addition of aluminum hydroxide to the diet. The effect of bone resorption coupled with zirconium citrate treatment is being investigated to determine whether this will increase the excretion of radioactive yttrium and plutonium.

Two thorium targets were received having had several hours of bombardment with deuterons. Protoactinium was separated by a solvent extraction method elsewhere reported. Approximately 25 millicuries were obtained and set aside for future separation of  $U^{233}$  from the  $Pa^{233}$ . Europium received in February from Oak Ridge was prepared for animal injection. Radioactive columbium, previously separated from zirconium, was prepared for injection. Radio-lanthanum was separated from barium and prepared for the biology section.

#### Medical Physics

#### Part B Project 48A-II

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Biological Effects Due to the Deuteron Beam of the 184-inch Cyclotron. During the past month our dosage measurements were compared with those of Dr. Failla and his group who have been visiting in Berkeley. For the first time a number of mice with mammary carcinoma were exposed to the deuteron beam with an effort to produce selective irradiation effects on these tumors. The work is being continued.

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Trace Analysis by Induced Radioactivity. The radioactive silver and mercury fractions in a set of neutron irradiated tissue ash samples were separated and analyzed. Further work is being done on the iron, cobalt, and zinc fractions.

Theory of Biological Tracers. Some detailed calculations were made in connection with the rate determinations of "steady state" biochemical reactions by means of radioactive tracers.

9. Health Physics and Chemistry

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Routine monitoring was carried out as usual during the month. In addition, on March 31, April 6, April 12, and April 16 one hundred eighty various boxes of equipment stored at Warehouse 2001 were dumped at sea. On April 21 twenty-three barrels and three packages from the 184" area were dumped at sea.

Research and development work has progressed on the following items:

- a. Beta-gamma lead-shielded gloved box essentially complete; also three gloved hoods.
- b. Alternate target assembly for 60-inch cyclotron designed and built and ready for trial dummy run on May 3. Original assembly ready ca. May 15.
- c. New style target built with jig for mounting platinum foil, for Ralph James target on 184-inch cyclotron.
- d. Plans for suitable equipment for gloved hood for yttrium colloid work by Dr. John Gofman under way.
- e. Special setup for shielding tests practically completed.
- f. Shielding and handling equipment for milling machine being designed. Mill being overhauled.
- g. Conway arc source: two new mock-ups being built for trial.
- h. A graphic catalog of remote control equipment being assembled, showing drawing, print numbers, rough dimensions, and available status of each piece.
- i. Four gloved boxes ready for shipment to Los Alamos. The special box for A. Ghiorso now ready.
- j. Overseeing of construction of tray for catch basin for horizontal column in Gilman Hall.
- k. Electric stirring apparatuses being assembled and modified according to request.

The Health Chemistry Section has also been engaged during April in organizing its supplies and equipment in the newly finished Room 101, Bldg. 5 and in the assigned space in the Old Radiation Laboratory.

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April, 1948  
APPROXIMATE DISTRIBUTION OF EFFORT

<u>PROGRAM</u>	<u>SUBDIVISION</u>	<u>MAN-MONTHS EFFORT</u>	<u>COMMENTS</u>
1. 184-inch Cyclotron	Operation	12.2	
2. 60-inch Cyclotron	---	---	Non-Project
3. Synchrotron	R.f. System	2.3	
	General	.3	
	Injection	2.0	
	Miscellaneous Equipment	1.3	
	Magnet Tests and Operation	4.3	
4. Linear Accelerator	Linear Accelerator-General	7.0	
	Van de Graaff General	7.0	
	General, Development, etc.	0.5	
5. Experimental Physics	Cloud Chamber	5.0	
	Film Program	3.7	
	Ionization Chamber and Crystal Counter	0.8	
	Neutron-proton Scattering	2.0	
	Proton-proton Scattering	2.0	
	Neutron Diffraction	0.5	
	Delayed Neutrons	2.0	
	Meson Counting	1.0	
	Absolute Cross Section Measurements	1.0	
	Neutron Half Life	1.0	
	Bevatron Design Studies	0.5	
	General Physics Research	6.1	
	Magnetic Measuring Equipment	0.7	
	Bevatron Magnet	3.4	
	6. Theoretical Physics	Synchrotron	0.3
Bevatron		1.3	
Cyclotron		0.5	
Linear Accelerator		0.3	
General Physics Research		8.7	

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<u>PROGRAM</u>	<u>SUBDIVISION</u>	<u>MAN-MONTHS EFFORT</u>	<u>COMMENTS</u>
7. Chemistry. Part A	Chemistry of Transuranic Elements	5.0	
	Nuclear Properties of Transuranium Elements	3.5	
	Transmutations with the 184-inch Cyclotron	6.0	
	Transmutations with the 60-inch Cyclotron	1.0	
	Analytical and Service	14.5	
	Chemistry of Astatine	1.5	
Chemistry. Part B	Synthetic and Experimental Organic Chemistry	6.6	
	Biological Chemistry	5.1	
	Photosynthetic Chemistry	6.9	
Chemistry. Part C	Metals and High Temperature Thermodynamics	2.5	
	Basic Chemistry, including Metal Chelates	4.5	
	General	2.0	
8. Medical Physics. Part A	Evaluation of Metabolic Properties of plutonium and Allied Materials in Animal and Man	11.0	
	Decontamination Studies	7.0	
	Radiochemistry	1.0	
	Radioautography	2.0	
Medical Physics. Part B (Project 48A-11)	Uranium Research	1.5	2.0 Consultant Man-Month
	Tumor Metabolism	0.3	0.5 "
	Special x-ray Studies, Radioactive Measurements, etc.	0.5	--
	Radioactive Carbon Studies	0.3	--
	Fundamental Medical Research	0.5	1.0
	Hematology	--	0.5
	Medical Work with 184-inch Cyclotron	0.5	0.5
9. Health Physics and Chemistry	Monitoring and Special Problems	6.6	
	Salvage, Decontamination, Disposal, etc.	3.7	
	Research and Development	8.4	

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