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A REPORT ON LITHIUM, SODIUM, AND LIQUID NEUTRON SOURCES

(Ad Interim Report)

Date: April 22, 1949

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INTRODUCTION

The objectives in producing neutron sources are to produce uniform products of as high efficiencies as possible from the neutron yielding elements and to do this in the safest manner. Several reports on the subject have been issued.^{1,2,3,4} The purpose of the present work was to improve the methods of preparing sources and at the same time increase the types of sources available.

Lithium, sodium, and their compounds were chosen because the neutron fluxes and energies are low, thus making the experimental work less hazardous to the operator. Also, the elements are quite reactive, so that techniques which are successful with them can be easily adapted to other materials.

Solvents for polonium, ways of mixing polonium and target, neutron yields, reproducibility, decay of the sources, means of recovering polonium, and materials for containers, were investigated.

SUMMARY AND CONCLUSIONS

Neutron sources were prepared by evaporating acid solutions of polonium from lithium salts. The evaporation was accomplished with no appreciable loss of polonium. The replacement of fluorine in lithium compounds by chlorine was not serious when the proper proportions of lithium compound and hydrochloric acid were used.

When up to one curie of polonium per gram of lithium fluoride was used, the source efficiencies averaged 81 to 83 per cent for either hydrochloric or hydrofluoric acids as solvents. With only 0.2 gram of lithium fluoride, source efficiencies were 10 per cent lower for hydrochloric acid than for hydrofluoric acid as solvent.

The data show wide variations in the efficiencies of these experimental sources. When conditions were carefully controlled and similar proportions of polonium and target were used, the efficiencies were reproduced within 5 per cent.

In using lithium hydroxide as the target in neutron sources some conversion to lithium chloride occurred, but the neutron flux was practically unaffected because of similar neutron yields of the two compounds.

Lithium fluoborate was prepared. Neutron sources were prepared from it using hydrochloric acid as solvent for polonium when the temperature did not exceed 160°C. At 200°C. there appeared to be some decomposition of the salt. Lithium fluoborate was hygroscopic, which caused a decrease in efficiency if the sources were not tightly sealed. The original neutron flux could partially be restored by heating to 150-160°C.

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Polonium-lithium metal neutron sources were prepared by inserting polonium on platinum supports in molten lithium and by volatilizing polonium onto the lithium and melting them together. The latter was the better method, but here the efficiency of the source did not exceed 65 per cent. Great difficulty was experienced in preventing thin films of oxide or nitride from forming on the lithium. This shows that a small amount of impurity can have a large effect on neutron yields.

Polonium-sodium neutron sources had efficiencies exceeding 100 per cent, indicating that absolute neutron counts are incorrect or that the theoretical yield of sodium is not known.

Methods for dissolving lithium and sodium neutron sources and recovering the activity were developed.

The neutron flux of several types of sources was followed for short periods of time and found to parallel the decay of polonium.

Polonium was not dissolved in ammonia but was soluble in a lithium ammonia solution. This indicates a compound or stable complex behaving as an acid. Sources prepared by evaporating the solution to dryness had low efficiencies. This would result if a compound such as $\text{Po}(\text{LiNH}_2)_2$ were formed.

Hydrogen sulfide water, sulfur dioxide water and carbon tetrachloride were not solvents for polonium. The application of an electric current to supported polonium in sulfur dioxide water gave finely divided polonium (probably colloidal) which could be used in preparing neutron sources.

Hydrogen bromide, sulfur monochloride and antimony pentafluoride dissolved polonium. The latter gave a neutron source which was 76 per cent efficient and showed promise of being reproducible as a standard.

EXPERIMENTAL WORK

I. Polonium-Lithium Compound Sources

A. General-Polonium-Lithium Hydroxide Sources

In approaching this work a compound had to be chosen that had the necessary chemical and physical properties, such as stability in air, water, acid, and alkaline solutions. The compound should also be easily obtainable. Lithium hydroxide was chosen as the first target material to be used. Lithium chloride hydrolyzes to form lithium hydroxide and hydrochloric acid when evaporated to dryness. This fact made it possible to utilize lithium hydroxide powder and hydrochloric acid solutions of polonium by the evaporation method.

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1. Amount of Target and Method of Preparation

A series of experiments were conducted to determine the amount of target material and solvent (hydrochloric acid) to be used to make the source. The weight of target material that appeared to be most feasible was 0.2 gram. Wetting the target material with 0.5 ml. of 0.5 N hydrochloric acid and slowly evaporating to dryness proved satisfactory. The first source LB-1 was prepared by adding 500 lambda of activity dissolved in 0.5 N hydrochloric acid to 0.2 gram of lithium hydroxide in a specially prepared glass tube. The glass tube with the source material was placed in a dessicator, containing phosphorus pentoxide, and allowed to dry overnight. In the morning the dessicator was heated, while under slight vacuum, with an electric heater until the temperature in the dessicator reached 40°C. Then a 24 inch mercury vacuum was put on the dessicator and held. The temperature within the dessicator was held at 40°C. for 24 hours. At the end of this period a count on the source indicated an 82 per cent neutron efficiency. The source was then tamped and recounted, 75 per cent neutron efficiency resulting. After careful mixing and retamping, the value remained at 75 per cent. The source contained 0.938 c. of polonium.

A second polonium-lithium hydroxide neutron source was repeated by the above described method. The source contained 1.967 c. Tamping of the source increased the efficiency slightly. A final neutron efficiency of 90 per cent was obtained on comparison with Radium-Beryllium Standard No. 49.

A polonium-lithium fluoride source was prepared by evaporation, simultaneously with the above source. The source contained 0.428 c. and had a neutron efficiency of 84 per cent. Tamping increased the efficiency somewhat, while further mixing and tamping decreased the neutron efficiency. The previous sources were made in glass containers. The sources following were prepared by evaporating the polonium solvent from a platinum can (Figure 1), the platinum can having a platinum lid to cover the source when completed. Modifications were made in the evaporation apparatus. Figure 2 shows a view of the heater block; Figure 3 shows the Evaporation Apparatus.

2. Effect of Solvent on Source Efficiency

Two polonium-lithium fluoride neutron sources were prepared with different solvents for polonium. The one source was prepared using 0.5 N hydrochloric acid as solvent, while normal hydrofluoric acid was used for the other. The purpose in using two different solvents was to determine their effect on the neutron efficiency. The sources were evaporated slowly around 90°C. with air being pulled over the top of the platinum can to carry away the vapors. Then

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BRASS HEATER BLOCK

SCALE: DOUBLE SIZE

PLATINUM CAN

Figure 2

Figure 1

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Figure 3

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EVAPORATION APPARATUS

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it was evaporated to dryness, the temperature was increased to 150-160°C. to further evaporate any solvent that might remain in the residue. A neutron count on LB-4, polonium-lithium fluoride, (hydrochloric acid solvent) indicated an efficiency greater than 100 per cent. Upon investigating the situation it was learned that the trouble existed in the neutron counter. Source LB-5, polonium-lithium fluoride, (hydrofluoric acid solvent) had an efficiency greater than 90 per cent.

Until this time it was very difficult to duplicate counts on the neutron counter. It was thought this great difference was due to the poor mixing of the activity with the target material. Upon further investigation it was learned that the great differences in neutron counts was due to inability to keep the source at the same distance from the face of the counter. A source holder was made up to hold the sources in a fixed position from the counter. The standard source was counted in the same fixed position. Henceforth, the difference between counts checked within the error of the counter. Several of the previously prepared sources were recounted with the result that polonium-lithium fluoride sources prepared with hydrochloric acid as solvent for polonium averaged about 10 per cent lower in efficiency than lithium fluoride sources prepared with hydrofluoric acid as solvent.

The next step was to investigate the 10 per cent efficiency difference between hydrochloric acid and hydrofluoric acid as the solvent. The theoretical neutron flux was calculated for lithium fluoride with varied amounts of lithium hydroxide (replacement of fluorine by chlorine and hydrolysis of lithium chloride yields the hydroxide) as impurity by the "attenburg Formula for mixed targets. This was done in order to determine the cause of the difference in neutron efficiencies when hydrochloric acid is used as solvent for polonium versus the use of hydrofluoric acid as the solvent. In the theoretical calculation, it was found that 3 per cent lithium hydroxide in lithium fluoride reduced the neutron efficiency by 2.1 per cent; 5 per cent lithium hydroxide - 3.6 per cent, and 10 per cent lithium hydroxide - 7.0 per cent. The next step was to set up a test with lithium fluoride and hydrochloric acid, evaporating it to dryness and then determining the amount of chloride left after evaporation. Three duplicate samples of 1.0 gram of lithium fluoride plus 2.5 ml. 0.5 N hydrochloric acid (five times the amount of target material and solvent used in preparing sources to cut down errors) were evaporated in the oven to dryness at 100-110°C. One set of duplicate samples was further dried at 150-160°C., and another set of duplicate samples was further dried at 200-210°C. A gravimetric chloride test was run on all of the samples to determine what per cent lithium chloride remained after evaporation. The amount of lithium chloride formed at 100-110°C. (3.5 per cent) checked the amount formed at 150-160°C. (3.5 per cent); however, the amount of lithium chloride formed at 200-210°C. was lower (2.8 per cent).

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The actual amount of lithium chloride formed does not coincide with the theoretical* (5.2 per cent) probably due to hydrolysis; however, the hydrolysis appears to be more critical somewhere between 160-210°C. All of the sources prepared were evaporated and finally heated at 150-160°C. Calculating by the Wattenburg Formula (neutron flux for mixed target), 3.5 per cent lithium chloride affected the neutron flux of lithium fluoride by 3.3 per cent.

In a series of experiments it was found that the actual lithium fluoride converted to lithium chloride varies directly with the hydrochloric acid concentration, (Figure 4.) With small portions of hydrochloric acid (1 ml. N) the amount of lithium fluoride in a one gram sample converted to lithium chloride is less than 2 per cent. In Table I is shown the per cent actual lithium fluoride converted to lithium chloride with varying amounts of hydrochloric acid.

Table I

CONVERSION OF LiF TO LiCl

<u>ML. N HCl</u>	<u>LiF Converted</u>	<u>Theoretical* LiF Conversion</u>
1.0	1.7%	2.6%
1.2	1.8%	3.2%
5.0	5.6%	13.0%
20.0	21.8%	52.0%

* Assuming all hydrochloric acid being used to convert lithium fluoride to lithium chloride.

The work was undertaken to determine the use of hydrochloric acid as a solvent for polonium in the preparation of lithium fluoride sources by the evaporation method. It is desirable to use hydrochloric acid instead of hydrofluoric because only a few materials are not attacked by hydrofluoric acid. The data show that as much as five milliliters of N hydrochloric acid may be used per gram of lithium fluoride without greatly affecting the neutron

* Theoretical lithium chloride in this report is the amount of lithium chloride in the lithium fluoride formed by the complete reaction with all the available hydrochloric acid (excess lithium fluoride). The percentage is based on the original weight of lithium fluoride in the experiment.

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Figure 4

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yield. This amount of acid is sufficient to prepare sources containing at least 40 curies of polonium.

A similar experiment was carried on with lithium hydroxide and hydrochloric acid. Five times the amount of each component was used as is normally used in making a source; this was evaporated to dryness. A quantitative determination of chloride was made. It was obvious that the hydroxide would be replaced with chloride ion; however, it was not known if the lithium chloride formed would all hydrolyze to form lithium hydroxide upon drying as was expected, theoretically. Actually, the theoretical amount of lithium chloride formed (5.2 per cent) checked the actual amount of lithium chloride (5.2 per cent) by experiment. Again calculating by the Mattenburg Formula, 5.2 per cent lithium chloride only affects the neutron flux of lithium hydroxide by 0.03 per cent. This small percentage was due to the neutron flux for lithium chloride and lithium hydroxide being so nearly the same.

A considerable number of polonium-lithium fluoride neutron sources were prepared using hydrochloric and hydrofluoric acids as solvents for polonium. The sources were all prepared by the evaporation method using 0.2 gram of lithium fluoride. The purpose in preparing these sources was to see what effect the two solvents had on the neutron efficiency. In Table II are listed data for the different solvents.

Table II

EFFECT OF SOLVENT ON SOURCE EFFICIENCY

0.5 N HCl as Solvent for Polonium

<u>Ident.</u>	<u>Quantity</u>	<u>Neutron Flux</u>	<u>Efficiency</u>
LB-4	0.316 c.	6.83×10^4 n./sec.	69%
LB-6	0.378 c.	8.66×10^4 n./sec.	73%
LB-7	0.378 c.	8.34×10^4 n./sec.	70%
LB-10	0.997 c.	2.39×10^5 n./sec.	76%
LB-11	0.117 c.	1.86×10^4 n./sec.	50%*
		Average	72%

1. N HF as Solvent for Polonium

LB-5	0.515 c.	1.26×10^5 n./sec.	78%
LB-8	0.343 c.	9.60×10^4 n./sec.	69%
LB-9	0.409 c.	1.07×10^5 n./sec.	83%
LB-12	0.983 c.	2.54×10^5 n./sec.	82%
LB-13	0.189 c.	4.27×10^4 n./sec.	72%*
		Average	83%

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* The efficiency drop in this region is probably due to adsorption of polonium on the walls of the source container, since the amount adsorbed is a higher percentage of the total as the polonium is decreased. These efficiency figures were not averaged in with the other figures.

As can be seen from the above data, the average neutron efficiency is approximately 11 per cent higher with hydrofluoric acid as the solvent for polonium. The trend of the sources on a graph plotting neutron flux against curies of polonium can be seen in Figure 5.

In a series of cold experiments with normal hydrochloric acid and lithium fluoride, it was learned that the amount of lithium fluoride converted to lithium chloride was relatively small on small amounts (1-2 grams). On increasing the weight of the target material from 0.2 grams to 1.0 gram, the efficiency using 0.5 N hydrochloric acid as solvent increased to the efficiencies of sources with 0.2 grams of lithium fluoride using normal hydrofluoric acid as the solvent for polonium. In Table III are listed the data for the different solvents using 1.0 gram of lithium fluoride.

Table III

EFFECT OF SOLVENT ON EFFICIENCY WITH ONE GRAM OF TARGET

0.5 N HCl as Solvent for Polonium			
<u>Ident.</u>	<u>Quantity</u>	<u>Neutron Flux</u>	<u>Efficiency</u>
LB-17	0.906 c.	2.34×10^5 n./sec.	82%
LB-18	0.301 c.	8.01×10^4 n./sec.	85%
Average			83%
N HF as Solvent for Polonium			
LB-19	0.431 c.	1.11×10^5 n./sec.	82%
LB-20	0.802 c.	1.97×10^5 n./sec.	78%
LB-21	0.506 c.	1.40×10^5 n./sec.	88%
LB-22	0.667 c.	1.64×10^5 n./sec.	78%
LB-23	0.630 c.	1.49×10^5 n./sec.	75%
LB-24	0.765 c.	1.99×10^5 n./sec.	83%
Average			81%

Averaged efficiency of polonium-lithium fluoride (0.5 N HCl as solvent).

0.2 gm. LiF
72%*

1.0 gm. LiF
83%*

* Average of four neutron sources.

** Average of two neutron sources.

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The average efficiency for polonium-lithium fluoride neutron sources using 0.5 N hydrochloric acid on 1.0 gram of lithium fluoride (83 per cent) has increased to the average of polonium-lithium fluoride sources using normal hydrofluoric acid on 0.2 grams lithium fluoride (81 per cent). This increase in efficiency was probably due to the increased ratio of target to polonium and the consequent decrease in the ratio of lithium chloride to lithium fluoride (hydrochloric acid concentration being the same). This comparison in graph form can be seen in Figure 5.

The next step was to prepare some polonium-lithium fluoride neutron sources using normal hydrofluoric acid as solvent on 1.0 gram of lithium fluoride. Increasing the weight of the target material should keep the efficiency the same or slightly better. Results of these sources can be seen in Table III. The efficiencies of these sources are inconsistent, this inconsistency could have been due to creeping of the lithium fluoride on the walls of the source container. This appeared to be more noticeable with hydrofluoric acid than with hydrochloric acid. The volume of hydrofluoric acid evaporated was twice that of hydrochloric acid, this could increase the chances of creeping, thus causing inconsistent efficiencies.

Suggestions for decreasing these inconsistent results are:

1. Increase concentration of polonium by decreasing volume of solvent.
2. Evaporate the hydrofluoric acid more slowly over an extended period of time.

Three polonium-lithium hydroxide sources were prepared by using 0.5 N hydrochloric acid as solvent in order to ascertain comparable data (Figure 5) with sources of other target materials. Since the first three lithium-hydroxide sources were made, quite a number of refinements in technique were developed and it was felt that data on previous polonium-lithium hydroxide sources (LB-1 to LB-3 inclusive) could not be compared with later neutron sources. Data on these sources are listed in Table IV.

Table IV

0.5 N HCL AS SOLVENT FOR POLONIUM ON 0.2 GRAM LIOH

<u>Ident.</u>	<u>Quantity</u>	<u>Neutron Flux</u>	<u>Efficiency</u>
LB-14	1.548 c.	3.72×10^4 n./sec.	67%
LB-15	1.186 c.	2.16×10^4 n./sec.	51.3*
LB-16	0.758 c.	1.81×10^4 n./sec.	67%

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* The low efficiency of this source is probably due to an error of adding a half portion of concentrated hydrochloric acid instead of half normal acid to the target material.

B. Polonium-Lithium Fluoborate Neutron Sources

To further investigate the usefulness of lithium compounds for making sources, it was decided to try lithium fluoborate. This compound was unavailable commercially, so the production became our first problem. The problem of preparation was handled by W. J. Schrantz, Jr. A thorough search of the literature gave very little reference to this compound; however, it was expected that a correlation could be drawn between the preparation of potassium fluoborate and lithium fluoborate. The method utilized in the preparation of potassium fluoborate consisted of the preparation of fluoboric acid from boric acid and hydrofluoric acid. To this was then added a solution of potassium hydroxide with the resulting precipitating potassium fluoborate. The first problem faced in the preparation of lithium fluoborate was the relative insolubility of lithium hydroxide. A large volume of water was necessary and this plus the water formed in the formation of the fluoboric acid was enough to hold a large percentage of the lithium fluoborate in solution. On running this preparation with calculated equivalent weights, which should have given yields of 46.9 grams, the experimental yield was only 2.3 grams.

In order to check the purity of the product, an analysis of the lithium fluoborate was attempted. Here, too, trouble was encountered because of the stable fluoborate radical. The precipitation of fluorine as lead fluo-chloride was unsuccessful. Heating of the compound with sulfuric acid to give lithium sulfate was also unsuccessful at this time because the heating temperature was too low.

A reference to preparation of fluoborate (Z. Anor. Allgem. Chem. 155, 327-32 (1926)) made use of the carbonate of the metal instead of the hydroxide. A series of four experiments was then planned.

I. Preparation of lithium fluoborate in an ice bath with lithium hydroxide in the solid form.

II. Preparation of lithium fluoborate in an ice bath with lithium carbonate in the solid form.

III. Preparation of lithium fluoborate at room temperature with lithium hydroxide in the solid form.

IV. Preparation of lithium fluoborate at room temperature with lithium carbonate in solid form.

All these experiments were designed to reduce the reaction. The preparation using lithium hydroxide

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with much lithium hydroxide remaining in the precipitate. The results with lithium carbonate in both cases were much more gratifying. The yields were 23.2 grams against a theoretical yield of 25.0 grams. A carbonate test was run on the sample and it was found that the sample contained 0.7 per cent carbon dioxide. To reduce impurities, about 12 grams were dissolved in boiling water and 7.5 grams of this precipitated. A sample from the supposedly pure lithium fluoborate was sent for X-ray and spectrographic analyses. The spectrographic report showed lithium, boron and fluorine present as major constituents. Lead and silica were present as minor constituents. The X-ray report showed traces of lithium fluoride plus new lines which were probably those of lithium fluoborate.

At this time R. E. Watrous suggested a preparation by utilizing a double decomposition between fluoboric acid and lithium chloride. Along with this it was decided to run another preparation using lithium hydroxide. Both the lithium chloride and the lithium hydroxide were added in solution and the mixture was then evaporated to dryness three times. Both gave the same crystal pattern on reaching dryness. The yields were as follows:

- I. With lithium chloride - 20.2 grams, or 86 per cent yield.
- II. With lithium hydroxide - 20.8 grams, or 93 per cent yield.

Spectrographic reports on both of these samples gave for Number I, lithium, fluorine, and boron present as major constituents and copper, calcium, silica, magnesium, and lead as minor constituents (less than 0.1 per cent). For Number II, the analysis showed the same impurities present.

The X-ray analysis showed a new system of lines as the major constituent. The absence of known lithium compounds that could have been present indicated that it must be lithium fluoborate. An unidentified minor constituent was also present. The sample prepared with lithium chloride seemed to have greater purity of the major constituents.

An analysis of the lithium as lithium sulfate gave a difference of 0.7 per cent between the actual and theoretical yields. This difference is probably due to impurities shown to be present in the spectrographic analysis.

Now, since some relatively pure lithium fluoborate had been prepared, it was necessary to simulate a lithium fluoborate source preparation in order to find any difficulties that might be encountered in preparing such a source. To begin with 0.5 N hydrochloric acid would be used as the polonium solvent. To 0.2 gram of lithium fluoborate, in a platinum can (Figure 1) were added 0.5 ml. 0.5 N hydrochloric acid. The acid was slowly evaporated to dryness in the evaporation apparatus (Figure 3) and then heated at 150-160°C. to further dry. Then the residue was taken up with water and quantitative chloride test

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was run to determine if any of the fluoborate radical had been replaced with the chloride ion. The sample did not show any positive test for chloride. The hydrochloric acid had evaporated completely.

The first two polonium-lithium fluoborate neutron sources LB-32 and LB-33 prepared by the evaporation method, had efficiencies of 72 and 64 per cent respectively. A source prepared using fluoboric acid as the solvent for polonium (LB-35) gave a much lower efficiency (54 per cent). Three additional lithium fluoborate sources were prepared using different solvents for polonium and different amounts of target material. In Table V are listed for comparison all the polonium-lithium fluoborate sources prepared.

Table V

DATA ON LITHIUM FLUOBORATE NEUTRON SOURCES

<u>Ident.</u>	<u>Wt. of Target</u>	<u>Solvent</u>	<u>Quantity</u>	<u>Neutron Flux</u>	<u>Efficiency</u>
LB-32	0.2	0.5 N HCl	1.219 c.	4.02×10^5 n./sec.	72%
LB-33	0.2	0.5 N HCl	0.354 c.	1.03×10^5 n./sec.	64%
LB-35	0.2	2.6 N HBF ₄	0.342 c.	8.46×10^4 n./sec.	54%
LB-38	0.2	N HF	0.915 c.	2.38×10^5 n./sec.	57%
LB-39	0.2	0.5 N HCl	1.180 c.	3.34×10^5 n./sec.	62%
LB-40	1.0	0.5 N HCl	0.270 c.	6.44×10^4 n./sec.	52%

The neutron efficiencies were much lower than expected; it was thought that the sources should be counted at the same time in order to rule out any errors due to the counter being off at the time the sources were prepared. In Table VI is listed the sources, dates counted, and neutron efficiencies.

Table VI

CHANGE IN EFFICIENCY OF FLUOBORATE NEUTRON SOURCES

<u>LB-32</u>	<u>LB-33</u>	<u>LB-35</u>	<u>LB-38</u>	<u>LB-39</u>	<u>LB-40</u>
3/26 72%	3/31 63%	5/3 54%	5/19 57%	5/26 62%	5/28 52%
5/17 58%	5/17 55%	5/17 51%			
6/18 52%	6/18 49%	6/18 47%	6/18 50%		

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As can be seen in each case, the neutron efficiency dropped. At first it was thought that the drop was probably due to an error in the neutron count at the time the source was prepared. Later (6/18) when all the sources were counted again, the sources all dropped in efficiency. This drop could not be due to counter error since two previously prepared polonium-lithium fluoride sources were used as checks on the counter. The efficiency of these sources checked previous values well within the probable error. Since lithium fluoborate is quite hygroscopic, absorption of moisture was probable.

In order to determine if moisture was causing the neutron efficiencies to drop off, a definite plan of drying neutron counting and calorimetry was set up. The purpose of calorimetry was to see that no activity was lost during drying and handling of the source (LB-39). Results of these operations are listed in Table VII.

Table VII

EFFECT OF DRYING ON SOURCE EFFICIENCY (LB-39)

<u>Remarks</u>	<u>Curies</u>	<u>Neutron Value</u>	<u>Efficiency</u>
Calorimeter Value When Made	1.168 c. 5/28/48	3.34×10^5 5/26/48	62%
Calorimeter Value 7/28/48	0.861 c. 7/28/48	2.11×10^5 7/28/48	54%
By Decay from 5/28/48	0.860 c. 7/28/48		54%
<u>Drying 48 hours 150-160°C.</u>			
Calorimeter Value 8/10/48	0.820 c. 8/6/48	2.24×10^5 8/6/48	60%
By Decay from 7/28/48	0.823 c. 8/6/48		60%
<u>Drying 24 hours 200-210°C.</u>			
Calorimeter Value 8/13/48	0.795 c. 8/12/48	1.87×10^5 8/12/48	52%
By Decay from 8/10/48	0.796 c. 8/12/48		52%
<u>Drying 18 hours 150-160°C.</u>			
By Decay from 8/13/48	0.775 c. 8/17/48	1.82×10^5 8/17/48	51%

Each time the source was calorimetered the decay value from the previous calorimeter value was calculated and tabulated for purpose of comparing. In each case the decayed value checked the calorimeter value. The efficiency increased after being dried 48 hours at 150-160°C., this was probably due to losing moisture since lithium fluoborate is very hygroscopic. The decrease in efficiency after being heated 24 hours at 200-210°C. may be due to decomposition of lithium

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fluoborate. It was thought that the lost neutron count might have been in error, a second neutron count, five days later, checked the efficiency of the previous count. From the above data it would appear that no activity is lost by drying sources at 200-210°C.

II. Polonium-Lithium Metal Sources

A. General Polonium-Lithium Metal Sources

The purpose in preparing a polonium-lithium metal neutron source was to see how the actual neutrons per second per curie (efficiency) compared with the theoretical figure, also to learn the techniques in the handling of lithium metal sources. To begin with, lithium is a metal belonging to the alkali metal group, it has a density of 0.53 at 20°C., melting point of 186°C. and boiling point 1336 ± 5°C. It decomposes in water and oxidizes rapidly in air. It is soluble in acid and ammonia. It also fuses very readily. From these few properties it can readily be seen that exceptional care would have to be exercised in handling this element. Since the element has such a low melting point, the first approach was to melt the lithium metal in an inert atmosphere with polonium on a microfoil and then to mix well. The lithium (Baker and Amend C.P. Grade) was stored in mineral oil to prevent oxidation. The mineral oil was removed by washing with petroleum ether (benzine). The lithium was cut up under petroleum ether to prevent further oxidation and to prepare small pieces for experimental purposes. The corrosion rate of various materials was tested by melting lithium with them. Stainless steel and nickel appeared to stand the test satisfactorily.

All the work on lithium metal was done in a tight drybox under an inert atmosphere. By flushing the drybox with helium the oxygen content was brought down to less than 0.5 per cent; however, the oxygen increased rapidly when the drybox was no longer flushed. Phosphorus pentoxide was used to absorb the moisture. The gas for flushing was predried. Indicating "Drierite" was used as a quantitative (length of time to change color) method of checking the moisture content. It was found that, when the oxygen content was less than one per cent, there was very little action on the scraped lithium metal; however, as the temperature increased, slight darkening occurred, probably due to lithium nitride. Lithium nitride is a black to gray cubic crystal and forms readily at 100°C. or above. Experiments were conducted at temperatures just above 186°C., the melting point of lithium and above the melting point of polonium (250-260°C.). Only a slight darkening of the surface of the lithium metal occurred between 250-300°C. Carbon dioxide was used also to flush the drybox and satisfactory results were obtained. It was thought that carbon dioxide might be easier to keep in the drybox since it was heavier than air.

Experiments were tried, sandwiching a platinum foil between two layers of lithium metal, melting, then scraping off the molten lithium metal from the platinum foil and observing the foil. No apparent effects on the platinum foil

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were observed. A polonium-lithium neutron source was prepared by this method. The source, LB-35, was prepared by heating lithium and polonium above their melting points and mixing well by tapping the closed nickel cylinder (Figure 6) vigorously on the bottom of the heater. The experiment was performed in a drybox with a carbon dioxide atmosphere and with moisture and oxygen content less than one per cent by volume. Lithium, cleaned by scraping off the oxide, was cut into small pieces and transferred to the nickel cylinder. The cylinder was flushed well with carbon dioxide, closed with the screw cap and placed in the heater to heat above the melting point of lithium. After heating several minutes, the nickel cylinder was tapped vigorously, removed from the heater, cooled and the contents examined. A layer of lithium covered the bottom area of the cylinder. The surface of the microfoil with the polonium was placed on the layer of lithium and more "cleaned" lithium metal cuttings were added on top of the microfoil with polonium. The cylinder was capped and transferred to the heater to melt the lithium. After heating to 220°C., the nickel cylinder was removed and prepared for neutron counting. The source, LB-35, 0.214 c., had a neutron efficiency of 23 per cent. The source was again heated several minutes at 300°C., mixed and neutron counted. The efficiency had increased to 29 per cent. Upon heating the source to 300°C. and mixing a third time, the efficiency dropped to 25 per cent. Upon removing the screw cap, the lithium appeared to be rather dark, probably lithium nitride formation, a compound having a 74 per cent lower neutron flux. To prepare a lithium neutron source and have its efficiency remain constant, it will be necessary to keep the source in a dry, tightly closed nitrogen-and-oxygen-free container.

A second polonium-lithium neutron source was prepared in the same way. The temperature of the final heating of polonium and lithium was increased to 360°C. The period of heating was lengthened and the temperature of the nickel cylinder was more accurately determined. The efficiency of LB-26, 0.164 c., increased to 59 per cent, which was more than double that of the first source.

B. Volatilizing Polonium to Prepare a Source

Two polonium-lithium neutron sources were prepared by plating the polonium on a platinum spiral coil and then volatilizing the polonium onto the inside surface of cold nickel cylinders. The polonium-coated nickel cylinders were then transferred to a carbon dioxide atmosphere in a drybox where the lithium metal was added. Neutron source LB-27, 0.337 c., had a low efficiency of 26 per cent and gradually reached, with heating and mixing, a peak efficiency of 60 per cent. Neutron source LB-28, 0.297 c., had a low efficiency of 12 per cent and with heating and mixing a peak of 32 per cent.

The platinum spiral coil (Figure 6) consisted of 22 gauge wire approximately 8 inches long with its lower lead going up through a quartz tube on the inside of the coil, the quartz acting as an insulator. The coil was made just small enough to clear inside of the nickel cylinder. Two small tips of quartz, just longer than the width of the coil, were fastened onto the quartz insulator

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Figure 6

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VOLATILIZATION APPARATUS

—SCALE: ACTUAL SIZE—

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to prevent the bottom of the coil from touching the cylinder. A platinum hemisphere was fitted over the coil to act as a shield and guide. The surface area of the coil (cathode) was approximately 4 cm.² An anode of platinum, 1 x 2 cm., was made up to be used with the cathode.

A simple plating control, consisting of a voltmeter, milliammeter, variable resistor and two dry cell batteries, was used for the plating of polonium from 8 ml. of 1.5 N nitric acid. A magnetic stirrer was used to agitate the plating bath. The amperage was under strict control, 8 milliamperes, equivalent to a current density of 2 milliamperes per sq. cm. The voltage was allowed to vary. Samples of the plating bath were taken during the plating operation and alpha counted in order to determine how the operation was proceeding. When a sufficient amount of polonium had been plated out, the coil was removed (current on) from the bath, rinsed in water and acetone. The coil was then placed in the nickel cylinder in the carbon dioxide filled volatilizing chamber and flushed with carbon dioxide. The volatilizing chamber containing the nickel cylinder was immersed in an alcohol dry ice bath to cool the nickel cylinder prior to volatilizing the polonium from the coil. A 5 volt, 60 ampere, transformer was used to heat the coil for the volatilization. The coil was heated for 10 seconds to flash the polonium. After 10-15 minutes the coil was heated again for 10 seconds to flash any polonium that might have remained on the coil from the first flashing. The nickel cylinder with polonium was calorimetered to determine how much activity was volatilized and condensed. Small pieces of lithium were added to the polonium in the nickel cylinder and then heated to just above the melting point of polonium. After mixing well the source was neutron counted. The procedure of heating, mixing and neutron counting was continued until the efficiency reached a maximum. Temperatures of the heater for melting and mixing the sources ranged up to 425°C. The nitrogen in the drybox was eliminated by trimming the lithium metal and having the nitrogen react with the lithium to form the black nitride. When the trimmed metal no longer turned black, it was believed that the free nitrogen had been removed.

Another polonium-lithium neutron source (LB-29, 0.260 c.) was made by volatilizing polonium from a platinum spiral coil and condensing the vaporized polonium onto a cold surface of lithium metal in a carbon dioxide atmosphere. The efficiency of the source just after condensing the polonium on the lithium was 51 per cent. After melting without mixing, the efficiency dropped to 27 per cent. Then with melting and mixing the efficiency rose to 54 per cent and finally after several meltings and mixings the efficiency rounded off at 65 per cent. The procedure used to prepare the source was nearly the same as that described in LB-27 and LB-28. The length of platinum wire in the spiral coil was reduced from 8 inches to 4 inches and was wound tightly around the quartz tube in order to fit freely within the lithium coated nickel cylinder. The lithium coated nickel cylinder was made by filling the cylinder with molten lithium, cooling and then drilling a hole in the lithium with a 1/4 inch drill. A brass rod 1 3/8 inches in diameter was drilled and fitted as a holder and

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guide for drilling the hole in the lithium so the platinum coil would fit into it without touching.

The source was checked to see if the neutron flux dropped off proportionally with the decay of the polonium over a period of three days. The average neutron efficiency before and after checked within 0.2 per cent, indicating the flux dropping off proportionally with the decay of polonium. The drop in efficiency upon melting the lithium was probably due to localized concentration of polonium. The 65 per cent efficiency was the highest attained to date for a polonium-lithium neutron source.

A polonium-lithium neutron source, LB-30, 0.227 c., was prepared by dropping a platinum microfoil with polonium, into a lithium coated cylinder. The source had an efficiency of 12 per cent just after dropping the foil into the lithium coated cylinder. Melting down the lithium onto the foil increased the efficiency only slightly, however, with heating and mixing well by tapping, the efficiency reached a maximum of 42 per cent.

Another polonium-lithium neutron source, LB-31, 0.380 c., prepared by volatilizing polonium onto a lithium coated cylinder with mixing by heating and placing the source in a mechanical vibrator attained an immediate maximum efficiency of 65 per cent. In Table VIII is listed for comparison all the polonium-lithium neutron sources.

Table VIII

DATA ON POLONIUM-LITHIUM NEUTRON SOURCES

<u>Ident.</u>	<u>Po</u>	<u>Neutron Flux</u>	<u>Efficiency</u>	<u>How Prepared</u>
LB-25	0.214 c.	6.17×10^3 n./sec.	29%	Adding foil with Po to Li
LB-26	0.164 c.	9.50×10^3 n./sec.	59%	Adding foil with Po to Li
LB-27	0.337 c.	1.96×10^4 n./sec.	60%	Volatilizing Po onto Ni cylinder
LB-28	0.297 c.	9.50×10^3 n./sec.	32%	Volatilizing Po onto Ni cylinder
LB-29	0.260 c.	1.64×10^4 n./sec.	65%	Volatilizing Po onto Li metal
LB-30	0.227 c.	9.52×10^3 n./sec.	42%	Adding foil with Po to Li
LB-31	0.380 c.	2.48×10^4 n./sec.	65%	Volatilizing Po onto Li

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C. Reclaiming Polonium from Lithium Sources

Considerable difficulty had been encountered in dissolving and accounting for the activity used in polonium-lithium neutron sources. At first the sources were dissolved in an open beaker with methyl alcohol. When the reaction had stopped, dilute hydrochloric acid was added to dissolve the polonium. Alpha counts indicated only 61 per cent accounted for. A closed system, with glass wool traps to catch entrained acid, was designed. With this apparatus it was found that more than 90 per cent of the polonium was recovered if the lithium was first dissolved in water and then leached in aqua regia.

D. Polonium-Lithium Ammonia Complex Sources

For some time the idea of finding some solvent for lithium that would not rapidly change into a compound of lithium has been worked upon. It was hoped to be able to find a solvent that would put lithium in solution and then vaporize the solvent without changing the chemical properties of the lithium. Investigations of the chemical literature revealed liquid ammonia to be such a solvent. It was hoped that it would be possible to dissolve polonium in liquid ammonia and dissolve lithium in the polonium-ammonia solution, then vaporize the liquid ammonia and thus have a very finely divided polonium-lithium mixture which should give a rather high neutron efficiency. Results from a solubility experiment of polonium in liquid ammonia indicated that it was practically insoluble. The solubility of polonium in a lithium-ammonia solution was investigated. The work had to be done in a drybox in an inert atmosphere. With a carbon dioxide atmosphere the liquid ammonia readily formed ammonium carbonate with very little lithium being dissolved. Then a dry nitrogen atmosphere was tried. More than 60 per cent of the activity on a microfoil was dissolved in a lithium-ammonia solution in a three minute interval. This indicated good possibilities for preparing a neutron source. A small polonium-lithium source, LB-34, 0.079 c., was prepared by this method with an average efficiency of 24 per cent. Two other sources, LB-36, 0.072 c., and LB-37, 0.197 c., had efficiencies of 21 and 27 per cent respectively. A fourth source, LB-44, 0.169 c., had an efficiency of 43 per cent, the highest for this type of source. The source was prepared in the same manner as the previous polonium-ammonia sources, with exception that after the liquid ammonia was vaporized at room temperature, it was further dried by heating at 100°C. plus for one hour with cover removed. Then it was heated (capped) at 300°C. plus and mixed three different times, at one minute intervals, on the mechanical vibrator. All the operations were done over a period of two hours, which reduced the chance of the lithium reacting to form a compound of a lower flux. This method of preparing sources is not as satisfactory as melting lithium and polonium together. In Table IX is listed for comparison sources prepared by dissolving polonium in a lithium-ammonia solution.

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Table IX

DATA ON POLONIUM-LITHIUM-AMMONIA NEUTRON SOURCES

<u>Ident.</u>	<u>Quantity</u>	<u>Neutron Flux</u>	<u>Efficiency</u>
LB-34	0.079 c.	1.87×10^3 n./sec.	24%
LB-36	0.072 c.	1.50×10^3 n./sec.	21%
LB-37	0.197 c.	2.19×10^3 n./sec.	26%
LB-44	0.169 c.	7.35×10^3 n./sec.	43%

III. Polonium-Sodium Metal SourcesA. General-Polonium-Sodium Metal Sources

The purpose in preparing a polonium-sodium metal neutron source was to obtain data such as, neutron flux, efficiency, techniques, etc. Sodium is a cubic, silvery metal with a density of 0.97, melting point of 97.5°C . and boiling point of 880°C . It decomposes in alcohol and is insoluble in benzene and ether. It is one of the alkali metals and tarnishes rapidly on exposure to air; however, it is not acted on by dry air or oxygen. It decomposes water at ordinary temperature forming hydrogen and the hydroxide. Dry nitrogen and dry carbon dioxide are inert to sodium below 300°C . Gaseous ammonia reacts vigorously with sodium and liquid ammonia forms blue solution - the so-called alkali ammoniums. Since sodium has such a low melting point, the idea of inserting a microfoil with polonium into molten sodium was tried. Two polonium sodium sources, LB-41, 0.207 c. and LB-42, 0.194 c., had efficiencies of 144 and 88 per cent, respectively. The sources were prepared by melting sodium in a cylinder (carbon dioxide atmosphere) and then inserting an active foil with polonium into the molten sodium. The sodium and polonium were heated above the melting point of polonium and mixed on a mechanical vibrator. Another source, LB-43, 0.029 c., was prepared in order to find out the reason for the two previous high efficiencies. It was thought that some of the neutrons might have come from impurities in the source container. The microfoil was counted in the glass vial and found to yield 8.31×10^2 n./sec. Then it was transferred to the brass cylinder and neutron counted with an atmosphere of air. No neutrons could be detected above that of the background. The brass cylinder was then opened in a carbon dioxide atmosphere and filled with carbon dioxide. Again no neutrons could be detected above that of the background.

The foil in the glass vial yielded neutrons, probably due to the action of the alphas on the boron in the glass. Then the cylinder was filled with sodium on top on the foil and heated above the melting point of sodium. A neutron count then resulted in just a little more than a background. Heating the source

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from 260°C. to 300°C. and mixing on the vibrator yielded the 24 per cent efficiency. The low efficiency might be attributed to (1) high adsorption of activity on container wall, lessening that on the target and (2) poor mixing of polonium with target material, which could have been brought about by putting the solid sodium on top of the active foil.

Two additional polonium-sodium neutron sources, LB-45, 0.390 c. and LB-47, 0.403 c., were prepared to see how they would compare in efficiency with the previous three sources. They were prepared by inserting a microfoil with polonium into a stainless steel cylinder of molten sodium. Stainless steel instead of brass was used because of the corrosive action of metallic sodium on brass. The efficiency of LB-45 was 112 per cent, while that of LB-47 was 120 per cent. Both sources were prepared in an atmosphere of dry nitrogen. The active foil of LB-47 was neutron counted in the stainless steel cylinder in order to see if any neutrons were originating from the alpha-neutron reaction on the impurities in the stainless steel. The count was 5.27×10^2 neutrons per second, two per cent of the total neutron count. There are uncertainties in neutron emissions of the elements and in counting which can account for high efficiencies.

B. Polonium-Sodium-Ammonia Complex Sources

Two polonium-sodium neutron sources were prepared, LB-46, 0.012 c., and LB-48, 0.028 c., by dissolving the polonium in a sodium-liquid ammonia solution. The active foil of LB-46 was inserted in the sodium-liquid ammonia solution about one minute and then removed. The liquid ammonia was vaporized slowly until all of it was volatilized, then the source was capped, heated to 300°C. plus and mixed on the mechanical vibrator. The neutron count was equivalent only to a background. Source LB-48 was prepared in the same manner as LB-46 with the exception that the active foil was inserted into the ammonia-sodium complex and permitted to remain in the source. The efficiency of this source was 55 per cent.

C. Reclaiming Polonium from Sodium Metal Sources

The sodium was dissolved by adding absolute methyl alcohol to the opened source in a closed, vented vessel. The reaction of the sodium with methyl alcohol was very mild. When the reaction stopped, a very small amount of water was added. Then a small amount of normal hydrochloric acid was added. After each addition of liquid (water or acid) a lapse of time of at least one hour was allowed to permit the reaction, if any to go to completion. All the activity from the source was recovered.

D. Polonium-Sodium Compound Sources

A polonium-sodium bromide neutron source was prepared (LB-49, 0.394 c., on 0.2 gram NaBr) by dissolving the polonium in 1.4 N hydrobromic acid and then

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evaporating the acid from the sodium bromide. The source was prepared the same way as the polonium-lithium compound sources, using the same evaporation apparatus (Figure 3). The neutron count was 7.04×10^3 n./sec. for an efficiency of 91 per cent. Since bromine does not yield neutrons and sodium bromide can be obtained in pure form, this offers a means of obtaining a pure sodium neutron spectrum.

Preparation of a sodium compound neutron source by volatilizing polonium onto a sodium compound in a quartz capsule was suspended. The idea was given up because of the danger of extreme pressure developing while heating the capsule to approximately 800°C . to vaporize the polonium. Calculations from data on cold experiments indicated excessive pressures on the capsule.

An effort was made to find some solution that would dissolve polonium and not react with certain target materials to form other impurities. The solutions tried were water gassed with hydrogen sulfide, water gassed with sulfur dioxide and water gassed with carbon dioxide. In each case, a platinum microfoil with polonium was dropped into one milliliter of the solution and allowed to remain. The progress of the solubility of the polonium was followed by preparing slides to be counted on the Logac at regular intervals.

In the first experiment, a microfoil with 0.302 c. of polonium was dropped into one milliliter of freshly gassed hydrogen sulfide water at 25°C . When the foil was first dropped into the hydrogen sulfide water, the portion of the foil containing the polonium turned black immediately and after a short time the hydrogen sulfide water was darkened. After one and three quarter hours, approximately 0.053 c. had gone into solution. Twenty hours later approximately 0.040 c. remained in solution; however, the solution had cleared up and a dark precipitate had formed. The precipitate would not go into solution upon mixing with the pipette tip. Some black residue from the foil seemed to flake off and remain on the bottom of the dissolving vessel. The one milliliter solution was mixed well, and the liquid and black residue were transferred with a 500 lambda pipette to 0.2 gram of sodium sulfide in a platinum can. The sodium sulfide water was evaporated and the source was heated to 150°C . a few hours to thoroughly dry. The source, LB-51, 0.204 c., had an efficiency of 89 per cent. This would indicate that quite a sizeable amount of polonium had been suspended in the solution and that it had to be rather finely divided in order to have such a high neutron efficiency.

Hydrogen sulfide is more than twice as soluble in water at 0°C . than at 40°C ., hence it was thought that water gassed with hydrogen sulfide at 0°C . and held at that temperature might have a greater solubility for polonium than at room temperature. An experiment was set up and after an hour and ten minutes less than one millicurie had gone into solution.

A polonium-sodium sulfide source, LB-52, 0.145 c., had an efficiency of 103 per cent. The source was prepared by the evaporation method. The polonium

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was dissolved in 0.09 N hydrochloric acid and the solution transferred to 0.2 gram of target material in a platinum can. The rate of solubility of the polonium in one milliliter of 0.09 N hydrochloric acid was very slow. After six days only 1.130 c. had gone into solution.

A third polonium-sodium sulfide source, LB-50, 0.234 c., had an efficiency of 89 per cent. The polonium was placed in one ml. of hydrogen sulfide water to dissolve, after four hours only seven millicuries had gone into solution. The solution was darkened and after standing over the week-end, the solution cleared up leaving a precipitate. After two drops of 0.09 N hydrochloric acid and one drop 6 N hydrochloric acid was added to try to put the precipitate into solution, a slide count indicated 0.048 c. to be in solution. After 24 hours, 0.169 c. had gone into solution and the black precipitate still remained. Thereupon the one ml. of solution was transferred to 0.2 gram sodium sulfide in a platinum cylinder to be evaporated to dryness. From the above account of the amount of polonium in solution at the time of transfer (0.169 c.) and the calorimetered source value, 0.234 c., it would indicate that much of the activity had been in suspension.

An experiment was conducted on the solubility of polonium in freshly gassed sulfur dioxide water. A platinum foil with polonium was inserted into one ml. of the solution. After 1 1/4 hours, 0.052 c. had gone into solution, however, after eighteen hours only 0.021 c. remained in solution. This might indicate the presence of colloidal or finely divided polonium. Conducting the same type of experiment using carbon dioxide, the solubility after one hour and a half was less than one millicurie; and after twenty hours the solubility was still less than one millicurie.

Several experiments have been conducted for the purpose of deplating polonium by means of an electric current from an electrode into a conducting solution such as sulfurous acid. In the experiments as much as 0.09 c. was put into solution. However, the solubility decreased upon standing, probably due to colloidal polonium settling out. A source, LB-53, 0.130 c., was prepared by deplating polonium at 2.4 milliamperes (current density - 2 milliamperes/sq. cm.) into one ml. solution of freshly gassed sulfur dioxide water. The above solution with the deplated activity was transferred to 0.2 gram of sodium sulfide in a platinum cylinder. The liquid was slowly evaporated to dryness and then further dried at 150-160°C. A slide test at the time the activity was transferred to the target material indicated approximately 0.02 c. in solution. From the above calorimeter value of the source 0.130 c. and the 0.020 c. in solution at the time of transfer, it was indicated that approximately 85 per cent of the activity had been in suspension. The source had a neutron efficiency of 72 per cent.

A polonium-sodium sulfide source, LB-54, 0.107 c., was prepared by deplating polonium into one ml. water solution of 0.2 gram of sodium sulfide.

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After deplating for three hours, the source was heated to evaporate to dryness in the usual way. The source had an efficiency of 67 per cent. In Table X are listed for comparison all sodium sulfide sources.

Table XDATA ON POLONIUM-SODIUM SULFIDE NEUTRON SOURCES

<u>Ident.</u>	<u>Quantity</u>	<u>Neutron Flux</u>	<u>Efficiency</u>	<u>Remarks</u>
LB-50	0.234 c.	7.31×10^3 n./sec.	89%	H ₂ S water-HCl solution
LB-51	0.204 c.	6.36×10^3 n./sec.	89%	H ₂ S water
LB-52	0.145 c.	5.21×10^3 n./sec.	103%	0.1 N HCl
LB-53	0.130 c.	3.34×10^3 n./sec.	72%	SO ₂ water
LB-54	0.107 c.	2.49×10^3 n./sec.	67%	Na ₂ S solution

IV. Polonium Liquid Neutron SourcesA. Polonium-Carbon Tetrachloride Sources

In the work on liquid neutron sources, it was hoped to be able to find a liquid compound with a relatively high neutron flux which would dissolve polonium. If polonium was soluble in the liquid, chances of having a homogeneous mixture would be much better than with any dry compound source. The compound should not have any elements that act as moderators, but it should consist of elements that have a high neutron flux.

Polonium-carbon tetrachloride neutron sources prepared by adding a microfoil with polonium to about one ml. carbon tetrachloride in a brass cylinder had efficiencies up to 65 per cent. After several days the efficiencies started a downward trend. In order to determine if the activity had been dissolved from the foil, the foil was removed from a source and the source calorimetered. The calorimeter value of the source only dropped 0.01 c. with the removal of the foil. This would indicate that the polonium had been dissolved from the platinum foil. Upon removing the supernatant liquid with a pipette, the efficiency was 5 per cent above that of the previous day. Checking the supernatant liquid for polonium by slide test indicated less than one-half millicurie. This indicates that the activity either flaked off the platinum foil and dropped to the bottom of the cylinder or the activity went into solution and plated out on the surface of the cylinder. Figure 7 shows the trend of the efficiencies of liquid neutron sources from day to day. Data are listed in Table XI.

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Figure 7

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EFFICIENCY vs. AGE OF SOURCE IN DAYS

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LB-57 SBF5
LB-35
LB-36
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LB-96
LB-97
LB-98
LB-99
LB-100

REMOVE SOIL FROM SOURCE

EGC-1 REMOVED FROM SOURCE

DAYS

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Table XI

DATA ON POLONIUM CARBON TETRACHLORIDE NEUTRON SOURCES

<u>Ident.</u>	<u>Quantity</u>	<u>Neutron Flux and Efficiency When Source was Made</u>	<u>Highest Neutron Flux and Efficiency</u>
LB-55	0.103 c.	2.13×10^2 n./sec. - 49%	2.78×10^2 n./sec. - 65%
LB-58	0.198 c.	3.66×10^2 n./sec. - 46%	4.04×10^2 n./sec. - 51%
LB-59	0.596 c.	1.48×10^3 n./sec. - 61%	1.48×10^3 n./sec. - 61%

B. Polonium-Sulfur Monochloride Source

A polonium-sulfur monochloride source, LB-56, 0.185 c., had an efficiency of 69 per cent initially and a final count of 3.59×10^2 n./sec., or efficiency of 79 per cent. The source was prepared by adding a microfoil with polonium to a stainless steel cylinder nearly filled with sulfur monochloride. After several days the source was uncapped to check the liquid level, the liquid level was approximately the same as when it was prepared. Figure 7 shows the trend of efficiencies from day to day.

C. Polonium-Antimony Pentachloride Sources

A polonium-antimony pentafluoride source, LB-57, 0.215 c., had a top efficiency of 76 per cent (4.83×10^4 n./sec.) at the time the source was prepared. The source was prepared by adding a microfoil with polonium to a stainless steel cylinder nearly filled with antimony pentafluoride. The neutron efficiency rounded off at 74 per cent and remained the same on six different days. The efficiency, at the time the polonium was reclaimed, was 76 per cent. As can be seen from Figure 7 of day to day trend of efficiencies on liquid sources, the efficiencies remained constant. Of the three liquid sources experimented on, antimony pentafluoride appears to be most favorable from a standpoint of constant efficiency.

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