C-4542

Concentration Increases in the Isotopic Germanium Carrier
Solutions Inferred from Solution Weights

J. K. Rowley

Chemistry Department, Brookhaven National Laboratory

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AUG 14 1997

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Concentration increases in the isotopic germanium carrier solutions via evaporation would produce weight losses in the carrier solutions. Since the bottle containing each carrier is usually weighed before and after the removal of carrier for a given run, the possibility exists of finding evidence for increasing carrier concentration in this record. With few exceptions the weighings were performed on the same top-loading balance in the external chemistry lab. The primary purpose was to monitor carrier usage, not to look for evidence for increasing carrier concentration. However, in order to look for this evidence, it is necessary only to compare the weights of the closed bottles between the times of carrier removal. This comparison has been performed and is reported here. Bear in mind that there is some evidence that the bottle containing carrier was not always weighed in the same way (e.g. perhaps a plastic bag was not removed from the bottle or the bottle cap was removed before weighing). Another possible source of weighing errors is the occasional buildup of static charges, especially on dry winter days. Such problems of static electricity were easily recognized and overcome. For the most part, the resulting record agrees with the assumption that the weighings were performed consistently.

Table 1 lists sampling dates taken from the carrier analysis sheets. I have also listed synthesis lab notebook references when I have found them. Since the weight record on the carriers is not very complete before 1992, I have concentrated on the period after that. The sampling dates, 30.06.92 and 2/94, show evidence of samples being taken in the weight loss record even though there is no record of sampling at those times in the notebooks. Because of this I have assumed that samples were taken of carriers 72 and 74 on those dates with sample sizes inferred from extrapolating linear least squares fits to both sides of the jumps in weight. The inferred sample weights were:

	Carrier	72	74
Date of Sampling			
30.06.92		3.2 g	2.0 g
2/94		1.6 g	0.9 g

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No evidence of weight loss from sampling on 5/93 and 30.06.93 was found. The latter date is probably the result of a misprint and should be 30.06.92. Note that if these

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inferred weights are not subtracted. the concentration corrections for carriers 72 and 74 would be even larger after the date, 30.06.92.

After assuming a weight loss from sampling for carriers 72 and 74 on those two dates, no further assumptions were made. A table of accumulated total weight loss versus time was constructed. The times listed are those when the carriers were weighed out. The results, plotted for both carriers 72 and 74, are shown in Figures 1a and 1b, respectively. The straight lines shown and their slopes are the result of least squares fitting to the data points. Between 1992.0 and 1994.0 carrier 72 lost weight at a rate of 2.84 g per year and carrier 74 at 2.07 g per year. After 1994 carrier 72 lost 2.03 g per year while carrier 74 lost 0.774 g per year. No evidence exists for a single sudden weight loss by the 72 carrier solution. Similar plots for the other carrier solutions show no clear evidence for the continual weight loss shown by carriers 72 and 74. Carriers 72 and 74 were stored in similar one liter bottles. The other carriers were stored in larger bottles. The weight loss was probably caused by slow evaporation, presumably through screw caps which did not seal properly.

A concentration correction factor for each period of standing was next calculated. This factor is defined as:

$$\text{Correction Factor} = \text{Final Concentration} / \text{Initial Concentration}$$

Since we are dealing with very dilute solutions, the density is essentially constant and

$$\text{Correction Factor} = \text{Initial weight of solution} / \text{Final weight of solution}$$

The cumulative factor is simply the product of all of the preceding factors for individual runs. Before calculating these factors it is necessary to subtract the weight of the empty bottle so that only the weight of the solution is used. Note that I have implicitly assumed that the solution has lost weight, but not the bottle. The resulting cumulative concentration correction factors are plotted versus time in Figure 2a and 2b. The points are calculated directly from the data. The lines show smoothed results. The smoothing was done by starting with the real solution weights at a given time (675.7 g for carrier 72 before the carrier for run 42 was weighed out and 638.5 g for carrier 74 before the carrier for run 47 was weighed out. Whenever a sample of carrier was taken for a run or an analytical sample, the actual weight was subtracted, but the rates of weight loss on standing were assumed to be those given by the least squares fits in Figure 1. The appropriate least squares rate of loss from Figure 1 and the elapsed standing time between weighings were used to calculate a weight loss during standing which was then subtracted from the initial weight of the solution to give the weight at the end of the standing period. This procedure was followed for each standing period and a cumulative correction factor was then calculated. The resulting points were connected to give the smoothed correction factors shown by the lines in Figure 2.

The next step was to correct each of the measurements of the carrier concentration using the smoothed concentration correction factor applicable for the sampling date. The resulting concentration has been corrected to a single time for each carrier (92.0 for carrier 72 and 92.1 for carrier 74). The resulting values are listed in Table 2. The

corrected concentrations for each of these dates are then averaged (weighted by the number of measurements) to give the best estimate of the carrier concentration at the reference date. The resulting spread of values is larger than I expected. However, the evidence for continual weight loss by these two carriers is very strong and I can think of no better way to estimate these two important values.

Finally, a yield correction factor was calculated for each run using the two carriers. These values are listed in Tables 3a and 3b. This multiplying factor is the ratio, old carrier concentration / new carrier concentration on reference date, divided by the smoothed concentration correction factor applicable to that carrier and run.

Should these results be taken seriously? I think that they must be unless it is shown that the bottle, not the solution, is losing weight. This question can have an experimental answer from simply weighing the empty bottles again. If the weight loss is from the solution itself, then these results offer strong and direct evidence that carrier solutions 72 and 74 have gradually increased in concentration over four years. On the other hand, there is no evidence for a single sudden loss of weight from carrier 72. A sudden 2.75% increase in concentration between runs 77 and 80 implies (the solution weight was 474 g) a 13 g loss of weight during this period. Such a large weight loss would have been easy to observe. It was not seen and the sudden loss scenario must be ruled out at a high confidence level.

Acknowledgement. This research was carried out at Brookhaven National Laboratory under contract DE-AC02-76CH00016 with the U.S. Department of Energy and supported by its Office of High Energy and Nuclear Physics

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Table 1. Record of Sampling of Carriers.

Sampling Date	Notebook/Page	Sampled By	Carriers Sampled	Comments
4/91	Not Found	---	72, 74	
7/91	Not Found	---	72	
22/11/91	3/79	FXH	72, 74	
30.06.92	Not Found	KR present	72, 74	Carriers for runs were weighed out on this day Evidence for sampling in weight losses.
02.03.93	4/22	KR	72, 74	Very likely samples were taken of 73 and 76 also.
5/93	Not Found	---	72	No evidence in weight loss for sample taking.
30.06.93	Not Found	---	74	Misprint? No one present at Gran Sasso from synthesis lab.
2/94	Not Found	CS/KR present	72, 73, 74, 76	Evidence for sampling in weight losses.
26.07.94	6/161	CS	72, 73, 74, 76	
18.07.95	8/9	HR	72, 73, 74, 76	No weights of large carrier bottle.
27.08.96	9/38	HR	70, 72, 74, 76	No weights of large carrier bottle.

Table 2. Correction of Carrier Concentrations to a Reference Time.

Carrier 72Ge_210690

TABLE 2 a

Sampling Date	Number of Measurements	Ave. Conc.	Conc. Corr. Factor	Conc. Corrected to 92.00
4/91	9	0.955 micromol/ml		
7/91	2	0.937		
11/91	13	0.943	0.999	0.944
30.06.92	8	0.972	1.002	0.970
5/93	5	0.976	1.007	0.970
2/94	5	0.977	1.011	0.966
26.07.94	7	0.959	1.015	0.945
18.07.95	8	0.972	1.021	0.952
27.08.96	7	0.975	1.033	0.944

Weighted Average

0.954 micromol/ml
0.956 micromol/g

Carrier 74Ge_250690

Table 2b

Conc. Corr.
to 92.10

4/92	11	1.003 micromol/ml	1.001	1.002
30.06.92	4	1.002	1.001	1.001
3/93	5	1.010	1.004	1.006
2/94	7	1.030	1.008	1.022
26.07.94	5	0.997	1.009	0.988
18.07.95	9	0.998	1.012	0.986
27.08.96	9	1.005	1.017	0.988

Weighted Average

0.998 micromol/ml
1.000 micromol/g

72Ge-210690

Figure 1a

74Ge-250690

Figure 1b

Figure 2a

72Ge-210690

Figure 2b

74Ge-250690

Table 3a.

Carrier 72Ge_210690

Run Number	Date Carrier Weighed	YEAR	Concentration Correction Factor	Overall yield Correction Factor
42	12.12.91	91.948	1.000	0.982
45	30.01.92	92.079	1.000	0.982
48	11.03.92	92.192	1.001	0.981
51	02.05.92	92.334	1.002	0.980
54	02.06.92	92.419	1.002	0.980
57	30.06.92	92.496	1.003	0.979
60	16 09.92	92.71	1.004	0.978
63	13.10.92	92.783	1.004	0.978
66	13.11.92	92.868	1.005	0.977
69	05.01.93	93.014	1.005	0.977
73	02.03.93	93.167	1.006	0.976
77	27.04.93	93.32	1.007	0.975
80	20.06.93	93.474	1.008	0.974
83	20.07.93	93.551	1.008	0.974
86	14.09.93	93.704	1.009	0.973
89	11.10.93	93.778	1.009	0.973
92	06.12.93	93.931	1.010	0.972
95	04.01.94	94.008	1.011	0.972
98	01.03.94	94.161	1.012	0.971
101	29.03.94	94.241	1.012	0.971
105	25.05.94	94.397	1.013	0.970
110	04.07.94	94.507	1.014	0.969
114	08.08.94	94.603	1.014	0.969
119	09.10.94	94.773	1.015	0.969
123	12.12.94	94.948	1.017	0.966
127	07.03.95	95.181	1.018	0.965
131	28.05.95	95.405	1.020	0.963
135	22.08.95	95.641	1.022	0.961
140	07.10.95	95.767	1.023	0.960
144	14.01.96	96.038	1.026	0.957

The concentration correction factor is the factor by which the concentration has increased since 92.00.

The overall yield correction factor is a factor by which present yields should be multiplied. This factor includes the concentration correction factor and a change in the carrier concentration on 94.00 from 0.939 micromol/g to 0.956 micromol/g.

Table 3b.

CARRIER 74Ge_250690

Run Number	Date Carrier Was Added	YEAR	Concentration Correction Factor	Overall Yield Correction Factor
41	21.11.91	91.893	0.999	1.007
44	09.01.92	92.025	1.000	1.006
47	18.02.92	92.134	1.000	1.006
50	31.03.92	92.247	1.001	1.005
53	08.05.92	92.351	1.001	1.005
56	30.06.92	92.496	1.001	1.005
59	18.08.92	92.63	1.002	1.004
62	13.10.92	92.868	1.003	1.003
65	09.11.92	92.94	1.003	1.003
68	05.01.93	93.014	1.003	1.003
72	02.03.93	93.167	1.004	1.002
76	27.04.93	93.32	1.004	1.002
81	22.06.93	93.474	1.005	1.001
84	17.08.93	93.627	1.006	1.000
87	14.09.93	93.704	1.006	1.000
90	09.11.93	93.857	1.007	0.999
93	06.12.93	93.931	1.007	0.999
96	31.01.94	94.085	1.008	0.998
99	01.03.94	94.164	1.008	0.998
102	25.04.94	94.315	1.009	0.997
107	23.06.94	94.477	1.009	0.997
111	11.07.94	94.526	1.009	0.997
115	23.08.94	94.644	1.010	0.996
120	01.11.94	94.836	1.010	0.996
124	09.01.95	95.025	1.011	0.995
128	05.04.95	95.26	1.011	0.995
132	26.06.95	95.485	1.012	0.994
136	12.09.95	95.699	1.013	0.993
141	31.10.95	95.833	1.013	0.993
145	12/06.96	96.118	1.014	0.992
149	28.03.96	96.238	1.015	0.991
153	06.05.96	96.345	1.015	0.991
157	25.06.96	96.482	1.016	0.990

The overall yield correction factor is a factor by which the present yields should be multiplied. This factor includes the concentration correction factor and a change in the carrier concentration on 92.10 from 1.006 micromol/g to 1.000 micromol/g.