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Loss of Radioactivity from Barium Carbonate Samples

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Publication Date

2010-02-02

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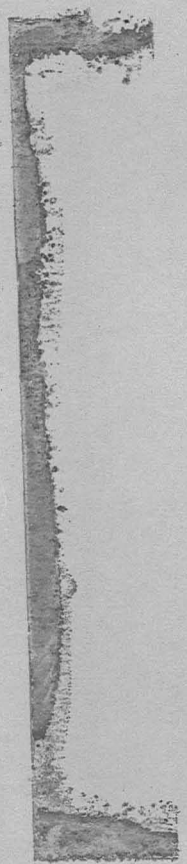

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LOSS OF RADIOACTIVITY FROM BARIUM CARBONATE SAMPLES

By

Peter E. Yankwich

9 January 1948

Special Review of Declassified Reports
Authorized by USDOE JK Bratton
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Abstract

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LOSS OF RADIOACTIVITY FROM BARIUM CARBONATE SAMPLES

By

Peter E. Yankwich

Department of Chemistry
and Radiation Laboratory,
University of California, Berkeley.

9 January 1948

ABSTRACT

The exchange loss of radioactivity from solid samples of barium carbonate through the agency of carbonic acid and its ions has been investigated under a variety of conditions. It is concluded that the losses observed are greatly dependent upon the method of sample preparation, in particular the nature of any heat treatment which the sample materials undergo.

This paper is based upon work performed under Contract No. W-7405-Eng.-48 with the Atomic Energy Commission in connection with the Radiation Laboratory, University of California, Berkeley.

For publication in "Science"

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LOSS OF RADIOACTIVITY FROM BARIUM CARBONATE SAMPLES

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9 January 1948

Several investigators have reported observations which lead them to conclude that barium carbonate samples containing carbon fourteen can lose rapidly rather significant fractions of their activity by exchange with atmospheric carbon dioxide through the agency of moisture. In many experiments it is desirable to store counting samples for possible checking after weeks or months, and any rapid loss of activity by exchange or any other process would make such a practice hazardous insofar as checks of experimental accuracy are involved; other consequences of rapid exchange loss are legion and need not be detailed here.

Only two papers are available which report the results of purposeful investigations of this exchange phenomenon, other observations having been reported during the course of various national meetings. One of these reports (1) deals only with exchange loss when solutions of sodium bicarbonate (pH 8.8) are evaporated to dryness in a variety of atmospheres. It was observed that even the small concentration of carbon dioxide in lab-

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(*) This paper is based on work performed under contract No. W-7405-Eng. -48 with the Atomic Energy Commission in connection with the Radiation Laboratory, University of California, Berkeley.

(1) W, B. Leslie, Experimental Use of C^{14}, Atomic Energy Commission, MDDC 674

oratory air caused losses as high as 60 percent in a single evaporation.

Most radioactivity determinations on solid samples are made on barium carbonate and it is to be expected that this solid can lose activity by exchange with carbon dioxide through carbonic acid and its ions, though the rate of this exchange should be inappreciable in the absence of water because of the high decomposition temperature of barium carbonate. Several estimates have been made as to the rate of loss of activity from moist precipitates of barium carbonate, but only one paper has been published which contains the results of experimentation designed to explore the problem(2).

In that study, thick deposits of barium carbonate were prepared by filtering a slurry of freshly precipitated solid through a retentive filter paper; the samples were dried by drawing air through the filter after washing the mass with acetone and ether. All samples used in the investigation were of "infinite-thickness" with regard to the carbon fourteen beta radiations. Several samples thus prepared were subjected to atmospheres of 1) moist carbon dioxide, 2) moist carbon dioxide-free nitrogen and 3) dry carbon dioxide. The experiments were carried out in a dessicator, the bottom of which was covered with a layer of acidified water during the "moist" experiments.

Exchange was observed only in the first case; after 65 hours, 37.4 percent of the activity had disappeared from the solid and was recoverable from the gas phase.

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(2) W. D. Armstrong and J. Schubert, Science, 106, 403 (1947).

Somewhat different results have been obtained in these laboratories with samples prepared by evaporation-settling technique (3). Samples are prepared by the evaporation of 95 percent alcohol from an alcohol-barium carbonate slurry; after the evaporation is complete the samples are dried for a few seconds at 130 °C. (It must be mentioned that the barium carbonate used in these experiments was dried at 115° after precipitation from an ammonium nitrate-sodium hydroxide solution; washing on the filter was carried out with distilled water only.)

Four different types of experiment were performed, the first of which was a repeat of part of the work just described. Two plates (deposits of solid on aluminum), 3.7 and 5.6 mg/cm^2 thick, were stored in a sealed desiccator, the bottom of which was covered with distilled water; the gas space was swept with a moderate stream of tank carbon dioxide for 15 minutes before the vessel was sealed. After 172 hours the plates were removed and counted; the activity loss in both cases was 0.9 ± 1.0 percent. Concentrated hydrochloric acid was added to the water layer in an amount necessary to produce an approximately 3N solution; the samples were put back into the vessel, which was swept again with carbon dioxide and sealed. The samples were removed after 150 hours and their radioactivities determined; the lighter sample lost an additional 1.2 ± 1.0 percent of its original activity, while the heavier plate apparently gained 0.2 ± 1.0 percent. The atmosphere during the second part of this experiment was sufficiently acid to etch the exposed portions of the aluminum sample mounts.

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(3) P. E. Yankwich, G. K. Rollefson and T. H. Norris, J. Chem. Phys. 14, 131 (1946); W. G. Dauben, J. C. Reid and P. E. Yankwich, Anal. Chem. 19, 828 (1947). See also: R. H. Hendricks, L. C. Bryner, M. D. Thomas and O. Ivie, J. Phys. Chem. 47, 469 (1943); P. E. Yankwich, Report CC-3567.

An experiment was performed to give information on the exchange rate when the solid was in contact with two solvent media. Two plates, both 4.65 mg/cm^2 thick, were carefully freed of loose particles and placed in shallow dishes; one was covered with 95 percent ethanol saturated with carbon dioxide and the other was covered with distilled water, also saturated with carbon dioxide (this gas is about three times as soluble in ethanol as in water). After 28 hours the samples were carefully removed from the dishes and dried in an air bath at 60°. Radioactivity determinations showed that both samples lost only 0.3 ± 1.0 percent of their activity during this time.

In a third test, small portions of carbon dioxide-saturated ethanol and water, and carbon dioxide-free ethanol and water were evaporated repeatedly from barium carbonate plates of various thicknesses. The plate was freed of loose particles and wet with 0.25 ml of the solvent or solution to be investigated, then dried in an air bath at 60° . The plate was then cooled, another portion of liquid added, and the cycle repeated; at suitable intervals the sample radioactivities were measured. The results are summarized in Table 1. The small difference in the rate of loss between the two aqueous media probably is due to very rapid loss of dissolved carbon dioxide from the evaporating solution of carbon dioxide in water; the rate of loss with this solution probably would be considerably higher if the evaporation were carried out under a constant pressure of carbon dioxide.

In one of the papers cited (2) a loss of 2.7 percent in 6 days is reported for a sample stored in a covered Petri dish. A fourth set of experiments has been performed to shed more light on this long-term exchanges loss. Pairs of barium carbonate plates of different thicknesses

Table 1

Activity Loss from $BaCO_3$ Plates - Repeated Liquid Evaporation

Liquid	Typical Result			Dependence on sample thickness
	Thickness, mg/cm^2	Liquid Vol. ml.	Activity Loss %	
95%-ethanol	4.65	11.0	0.0 ± 0.6	none observed
dist. H_2O	0.42	15.0	0.8 ± 0.1 per ml. liquid	loss/ml decreses with increasing thickness
95%-ethanol + CO_2	4.82	21.0	0.2 ± 0.5	none observed
dist. H_2O + CO_2	0.41	9.1	0.9 ± 0.1 per ml. liquid	loss/ml decrease with increasing thickness

were stored over a period of 9 weeks in different controlled and uncontrollable atmospheres. At intervals of 10 to 14 days, the plates were collected and their radioactive strengths determined; at the end of the test all samples were reweighed, but no significant weight changes were observed. The sensitivity of the counter was checked with standard of uranium glass and of shellac-bound barium carbonate. The data obtained are collected in Table 2.

It is apparent from these data that barium carbonate samples, at least those of this particular variety, can be stored in loosely closed containers for rather long periods without serious loss of activity These data do not mean that exchange will be a trivial matter with samples prepared by other methods, because of particle size distribution, etc; each circumstance at least requires observation of active samples for long periods.

It is certain that barium carbonate-carbon dioxide exchange can occur rapidly only when the solid is in contact with a medium in which both carbon dioxide and barium carbonate are appreciably soluble; this is demonstrated by the failure to observe exchange when alcoholic solutions are used, and the simplest explanation is that barium carbonate is much less soluble in ethanol than in water. There is also a surface effect involved, for under the same conditions precipitates which had not been warmed exchanged much more rapidly than those which had been subjected to heating; such effects are well known and are responsible for the solubility denaturation of such precipitates as aluminum and ferric hydroxides.

Table 2

Long-term Loss of Activity from $BaCO_3$ Samples

Conditions of Storage	Plate thicknesses mg/cm^2	Mean percent loss (9 wks) Error includes counting error and sensitivity loss	
Uranium glass	-	1.2	1.2
$BaCO_3$-shellac bound	-	1.0	1.2
covered Petri dishes	1.65, 2.20	2.9	2.0
office-type room	0.53, 3.06	2.6	1.2
outdoors (protected only from rain)	1.07, 2.90	15.2	4.2
stream of wet outdoor air	0.63, 3.29	3.6	1.3
drying oven at 130° (air)	1.35, 2.14	3.6	1.4
stream of wet CO_2	1.75, 2.87	8.0	1.4
organic synthesis laboratory low CO_2, relatively acid air	1.03, 2.77	6.0	1.3
combustion room, high CO_2, relatively acid air	0.66, 1.88	12.1	1.6
sample storage book; somewhat better sealed than Petri dishes	1.25, 2.86, 3.10	1.7	1.3 (20 wks)