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PROJECT GASBUGGY GAS QUALITY ANALYSIS AND EVALUATION PROGRAM TABULATION OF RADIOCHEMICAL AND CHEMICAL ANALYTICAL RESULTS

CONF-700101--6

Charles F. Smith, Jr.

November 17, 1969

This report supersedes report UCRL-50635 dated April 22, 1969.

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Lawrence Radiation Laboratory
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UCRL-50635
Rev. 1

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Foreword to Revision 1

Since publication of the initial tabulation in April, 1969, significant new information has become available. In particular, Rev. 1 contains results through sample No. 75, corresponding to the end of August 1969. The data added for samples No. 59 through 75 in this revision cover the first 6 mo of the long-term production test that began in February, 1969. Production testing continued through October 1969, when we shut-in the chimney, following, a rapid drawdown of the well. When we have collected the data from this series of tests, we will issue a second revision of this tabulation.

Much new information has also been added to the early data, the most significant of which may be the concentrations of ^{14}C in methane and CO. Some minor corrections have been made in a few of the previously published data.

This tabulation provides a listing of data that will form the basis for a discussion of the Gasbuggy Gas Quality Program to presented at the Symposium on Engineering with Nuclear Explosives—4th Plowshare Symposium at Las Vegas in January 1970 (proceedings to be published).

Contents

PROJECT GASBUGGY
GAS QUALITY ANALYSIS AND EVALUATION PROGRAM TABULATION OF RADIOCHEMICAL AND CHEMICAL ANALYTICAL RESULTS

Abstract

In the past, analytical results for samples of Project Gasbuggy chimney gas have been distributed to project participants and the interested public in various ways. This tabulation is an attempt to summarize all of the previous results with a consistent up-to-date listing of samples, radionuclide concentrations, and chemical analysis results and, at the same time, provide a vehicle for distributing these results. The listing includes the description and the disposition of all samples obtained for analysis at the Lawrence Radiation Laboratory following the Gasbuggy detonation. Radiochemical results expressed as concentrations in chimney or formation gas are given for ^{85}Kr, HT, HTO, CH_3T, C_2H_5T, C_3H_7T, $^{14}CO_2$, ^{37}Ar and ^{39}Ar. Upper limits for the concentrations of several nongaseous or semivolatile nuclides are presented. The tabulation also includes the percentages of N_2, CO_2, CO, H_2, CH_4, C_2H_6 and C_3H_8 in the sample gas as determined by mass spectrometry.

We intend to update this report as additional results and samples are obtained, and to reissue it periodically as a continuing series of revisions bearing the same report number.

Introduction

The Gas Quality Analysis and Evaluation program for Project Gasbuggy is an active investigation of both the degree of radionuclide contamination and the post-detonation chemistry of the gas in the nuclear chimney. A significant body of information has already been amassed. The program participants are presently evaluating these data, as well as continuously analyzing additional samples. The distribution of these results to project participants and interested public has been accomplished by various means in the past; however, none of these ways have been completely satisfactory. This tabulation, then, has two primary purposes. First, it assembles the analytical results in a single document with enough additional information to permit interpretation of their significance. And second, it provides a means for general distribution of the results in advance of the final program reports.

By the nature of the Gasbuggy experiment, there can be no definitive conclusion

of the project in the near future. Production testing and chimney flaring experiments are scheduled for at least the next several months. Therefore, this tabulation will be up-dated periodically to reflect this continuing program and to provide a current listing of the results of analyses for samples yet to be obtained. In addition, samples presently on-hand may be re-analyzed or additional determinations may be made as required by the evaluation program. The results of such analyses will appear in forthcoming revised editions of this tabulation.

This report consists of four sections. The first section lists all samples obtained for analysis at the Lawrence Radiation Laboratory and provides information pertaining to their disposition and significance in the general Gasbuggy program. Radiochemical results for krypton-85, tritium and carbon-14 appear next, followed by results of chemical analyses for major constituents. The last section summarizes a number of results which are important to the gas quality program but do not fit the tabulations of results presented in other sections.

Sample Log

The majority of samples were obtained simply by connecting an evacuated sample bottle to the existing blow-down equipment at the well head. Sufficient gas was released to flush the well pipe before the sample was snapped. Typical sample pressures ranged from 15 to 25 psig in either 8-liter or 0.8-liter sample bulbs. Early in the postshot program several samples were obtained by lowering an evacuated sampling system on a wire rope to depths of 3820 ± 70 ft.[1] The bottle was allowed to reach ambient temperature, remotely opened, reclosed, and then withdrawn. Sample pressures were on the order of formation pressure, total sample volumes ranged between 2 and 3 ft^3.

A sufficient number of samples have been analyzed to satisfy the requirements of the program's major goals.[2] An approximately equal number of samples have been collected and stored for possible analyses or to serve as historical specimens. Portions of the analyzed samples have been similarly stored. These historical samples will either be retained or analyzed at some later time to satisfy the requirements of the evaluation program.

In addition to determining the radionuclide concentrations and chemical composition of the gas, several samples were passed through charcoal filters. These were subjected to gamma spectroscopy in an attempt to identify nongaseous nuclides being carried by the gas. This topic is discussed later.

Results from samples 1 through 34 comprised the information upon which the Gas Quality Status Report was based.[3] This status report, which also contained a limited interpretation of the results, was based upon the averages of samples taken at approximately the same time. For completeness, the individual results and sample description upon which these averages are based, are included in the tabulation of this report. Most results for samples through No. 58

were given limited public distribution at the Gasbuggy work shop held at LRL on February 26, 1969.

Although most entries in Table I are self-explanatory, the following definitions are included to avoid confusion.

Sample No.:

Samples are numbered in the order in which they were received at LRL. This produces chronological sequencing in most instances.

Acquisition:

Date and prevailing local time that the sample was collected.

Sample type:

Well: GB-E — grouted emplacement hole

GB-ER — reentry of emplacement hole to the nuclear chimney

GS-2RS — reentry of the GB-2 preshot investigation well

Location:

D — down-hole snap sample

S — surface snap sample

Total flow:

Approximate cumulative total of gas produced from GB-ER. Units of 10^6 ft^3

Laboratory analysis:

RC — Radiochemistry

* "minimal" analysis

** "normal" analysis

*** "complete" analysis

MS — Mass Spectrometry - complete analysis

γ — All or a portion of the sample passed through charcoal for gamma-counting.

Comments:

BHP — Bottom hole pressure

Table I. Post-detonation log for Project Gasbuggy samples.

Sample No.	Acquisition Date	Acquisition Time	Sample type Well	Sample type Location	Total flow (10^6 ft^3)	Laboratory analysis RC	Laboratory analysis MS	Laboratory analysis γ	Comments
1	12-10-67	2130	GB-E	S	0.		*	*	Cable leak to sealed well-head annulus — charcoal filtered
2	12-11-67	1120	GB-E	S	0.	**	*		Cable leak to sealed well-head annulus
3	12-11-67	1115	GB-E	S	0.	**	*		Cable leak to sealed well-head annulus
4	12-10-67	2130	GB-E	S	0.		*	*	Cable leak to sealed well-head annulus — charcoal filtered
5	1-13-68		GB-ER	D	0.	**	*	*	4-in.-diam sampling system
6	1-13-68		GB-ER	D	0.	**	*	*	4-in.-diam sampling system

Table I. (Continued)

Sample No.	Acquisition Date	Acquisition Time	Sample type Well	Sample type Location	Total flow (10^6 ft^3)	Laboratory analysis RC	Laboratory analysis MS	Laboratory analysis γ	Comments
7	1-13-68	0200	GB-ER	S	0.	***	*		Surface 8-liter snap sampler
8	1-17-68	0930	GB-ER	S	0.3		*		Surface 8-liter snap sampler
9	1-17-68		GB-ER	D	0.3		*		2-in.-diam sampling system
10	1-17-68		GB-ER	D	0.3	**	*		2-in.-diam sampling system
11	1-17-68	1555	GB-ER	S	0.3	**	*		Surface 8-liter snap sampler
12	2-27-68	1024	GB-ER	D	0.3		*		2-in.-diam sampling system
13	2-27-68	1335	GB-ER	S	0.3		*		Surface 8-liter snap sampler
14	2-27-68	1340	GB-ER	S	0.34	**	*		Surface 8-liter snap sampler
15	2-27-68	1350	GB-ER	S	0.34			*	Sample vented through charcoal filter
16	2-27-68	1425	GB-ER	D	0.34	***	*		2-in.-diam sampling system
17	4-22-68	1030	GB-ER	S	0.35	*	*		Surface 8-liter snap sampler
18	4-22-68	1030	GB-ER	S	0.35	***	*		Surface 8-liter snap sampler
19	6-28-68	1320	GB-ER	S	0.36	*	*		Start 5×10^6 ft^3 /day flow test
20	6-29-68	1315	GB-ER	S	5.4	***	*		[Surface 8-liter snap sampler]
21	6-30-68	1315	GB-ER	S	10.4		*		
22	7-1-68	1300	GB-ER	S	15.4				
23	7-2-68	1300	GB-ER	S	20.4	**	*		
24	7-3-68	1330	GB-ER	S	25.4				
25	7-4-68	1306	GB-ER	S	30.4	**	*		
26	7-7-68	1306	GB-ER	S	40.4	**	*		
27	7-8-68	1300	GB-ER	S	45.4				
28	7-9-68	1300	GB-ER	S	50.4				
29	7-10-68	1300	GB-ER	S	55.4	**	*		End 5×10^6 ft^3/day flow test
30	7-11-68	1200	GB-ER	S	56.1				Start 0.75×10^6 ft^3/day flow test
31	7-12-68	1300	GB-ER	S	56.9	***	*		[Surface 0.8-liter snap sampler]

Table I. (Continued)

Sample No.	Acquisition Date	Acquisition Time	Sample type Well	Sample type Location	Total flow (10^6 ft^3)	Laboratory analysis RC	Laboratory analysis MS	Laboratory analysis γ	Comments
32	7-13-68	1300	GB-ER	S	57.6				
33	7-14-68	1300	GB-ER	S	58.4	**	*		End 0.75×10^6 ft^3/day flow test
34	7-22-68	1900	GB-2RS	S		**	*		Surface 8-liter snap sampler
35	7-22-68	1915	GB-2RS	S					
36	9-18-68	1255	GB-ER	S	58.5	**	*		
37	9-18-68	1250	GB-ER	S	58.5				
38	11-7-68	1100	GB-ER	S	68.1				Start constant bottom hole pressure test
39	11-8-68	1200	GB-ER	S	68.7	**	*		BHP about 860 psig
40	11-15-68	1200	GB-ER	S	73.0				
41	11-22-68	1245	GB-ER	S	75.9	**	*		flow about 0.35×10^6 ft^3/day
42	11-29-68	1200	GB-ER	S	78.2				
43	12-6-68	1400	GB-ER	S	81.0				Surface 0.8-liter snap sampler
44	12-7-68	1100	GB-ER	S	81.4	**	*		End test
45	12-10-68	1200	GB-ER	S	95.6	**	*		
46	12-13-68	1445	GB-ER	S	104.5	**	*		Start constant bottom hole pressure test
47	12-12-68	1200	GB-ER	S	103.8				
48	12-19-68	1000	GB-ER	S	109.3				BHP about 710 psig
49	12-26-68	1215	GB-ER	S	114.1	**	*		flow about 0.6×10^6 ft^3/day
50	1-2-69	1345	GB-ER	S	118.0				Surface 0.8-liter snap sampler
51	1-11-69	1400	GB-ER	S	122.5	**	*		End test
52	1-14-69	1200	GB-ER	S	140.0	**	*		
53	1-17-69	1115	GB-ER	S	~149.0	**	*		Start constant bottom hole pressure test
54	1-27-69	1520	GB-ER	S	~156.0				
55	2-3-69	1100	GB-ER	S	~161.0	**	*		BHP about 500 psig
56	2-10-69	1130	GB-ER	S	~165.0				flow about 0.6×10^6 ft^3/day
									Surface 0.8-liter snap sampler
57	2-18-69	1245	GB-ER	S	~170.0	**	*		End test
58	2-25-69	1150	GB-ER	S	~186.0	**	*		Start chimney flushing—rapid drawdown
59	3-5-69	1300	GB-ER	S	~204	*	*		BHP = 380 psig or less flow not greater than 2.3×10^6 ft^3/day Surface 0.8-liter snap sampler

Table I. (Continued)

Sample No.	Acquisition Date	Acquisition Time	Sample type Well	Sample type Location	Total flow (10^6 ft^3)	Laboratory analysis RC	Laboratory analysis MS	Laboratory analysis γ	Comments
60	4-11-69	1500	GB-ER	S	~218	**	*		
61	4-18-69	1800	GB-ER	S	~222	*	*		
62	4-25-69	1605	GB-ER	S	~224	**	*		
63	5-9-69	1530	GB-ER	S	~228	*	*		
64	5-16-69	1630	GB-ER	S	~232	**	*		
65	6-6-69	0900	GB-ER	S	~237	*	*		
66	6-12-69	1600	GB-ER	S	~238	**	*		[Continuing long term production test BHP ≈ 260 psig]
67	6-27-69	1300	GB-ER	S	~241	*	*		
68	7-4-69	1700	GB-ER	S	~243	**	*		
69	7-11-69	1725	GB-ER	S	~244	*	*		
70	7-18-69	1800	GB-ER	S	~245	**	*		
71	8-1-69	1500	GB-ER	S	~247	*	*		
72	8-8-69	2030	GB-ER	S	~248	**	*		
73	8-15-69	1745	GB-ER	S	~250	*	*		
74	8-21-69	2000	GB-ER	S	~251	**	*		
75	8-29-69	1330	GB-ER	S	~252	*	*		

Radionuclide Concentrations

Concentrations of various principal long-lived radionuclides found in the Gasbuggy chimney are listed in Table II. All species have been related to chimney gas after the air (based on oxygen content) had been removed. All concentrations are listed in units of picocuries per standard cubic centimeter. Table II also lists the precision of the measurements expressed as percent standard deviation of the mean of replicate measurements. This quantity indicates agreement between replicate determinations for a given radionuclide. The absolute accuracy of these measurements is unknown, but the uncertainty is probably less than ±10 percent for the species listed. The determination is complete for all the values listed. Systematic errors are improbable, but their occurrence cannot be completely ignored. The evaluation program has caused us to suspect that a few individual determinations lie outside the quoted deviations. For this reason, these determinations have been, and will be, repeated. However, because of the nature of a tabulation such as this, and the attempt to up-date it as new information becomes available, the reader is cautioned that such outliers are both statistically and operationally possible. One of the several goals of the program is to minimize their occurrence.

Radiochemical analysis of a Gasbuggy sample begins with separation and purification of the desired components by elution chromatography. Carrier (e.g., stable Kr gas) is added to aid in recovery of the trace components. The purified fractions are then placed in appropriate counters for radio assay. Krypton-85 is determined in quadruplicate by thin-window beta proportional counting. Compounds containing tritium and ^{14}C are determined by internal proportional counting of duplicate or quadruplicate fractions. Appropriate corrections for counting efficiency, geometry, chemical purity, background, and sample absorption are made where necessary and the disintegration rate per unit volume of active gas is obtained. In the case of trace components this is directly the concentration of the species in the original sample. For components of the chimney gas this is a specific activity which is converted to a concentration in the sample by application of the fractional abundance as determined by mass spectrometry. Concentrations per unit volume of sample, thus obtained, are converted to the quantities appearing in Table II by application of the appropriate air corrections (discussed later) and conversion factors. This tabulation, therefore, is not raw data but is, in fact, fully reduced to consistent, meaningful, final results.

For intercomparison of the radionuclide concentrations, all data have been corrected for decay to the time of detonation. True concentrations at any given time can, therefore, be found by application of the appropriate decay correction. Due to the half lives involved, such a correction is small (< 12 percent) for the species listed in Table II.

Table II. Radionuclide concentrations for Project Gasbuggy samples in picocuries per standard cubic centimeter of chimney gas.[a,b]

Sample No.	Date	^{85}Kr	HT	CH_3T	C_2H_5T	C_3H_7T	$^{14}CO_2$	$^{14}CH_4$	^{14}CO
1	12-10-67								
2	12-11-67	110.0 (1.0)	3690.0 (1.7)	319.0 (1.7)	17.2 (0.5)				
3	12-11-67	118.0 (0.4)	3360.0 (0.7)	346.0 (1.3)	17.8 (0.7)				
4	12-10-67								
5	1-13-68	124.0 (0.4)	225.0 (1.6)	457.5 (2.6)	79.6 (1.0)				0.343 (4.0)
6	1-13-68	125.0 (1.0)	124.0 (1.0)	430.0 (4.2)	76.3 (1.8)				0.201 (9.4)
7	1-13-68	119.0 (0.5)	189.0 (1.3)	454.0 (0.7)	79.8 (0.6)	34.0 (1.8)	2.11 (0.4)	0.256 (1.2)	0.216 (2.3)
8	1-17-68								
9	1-17-68								
10	1-17-68	124.0 (0.7)	125.0 (0.5)	458.0 (1.2)	80.6 (1.5)				0.310 (6.8)
11	1-17-68	123.0 (0.3)	175.0 (1.0)	450.0 (0.9)	77.8 (2.2)		1.34 (0.4)		0.218 (1.0)
12	2-27-68								
13	2-27-68								
14	2-27-68	124.0 (0.3)	97.1 (0.8)	461.0 (2.0)	83.4 (1.1)			0.370 (4.1)	0.129 (1.8)
15	2-27-68								
16	2-27-68	115.0 (1.7)	128.0 (0.3)	470.0 (0.4)	81.6 (1.0)	15.2 (1.7)	2.48 (1.4)	0.290 (1.6)	0.125 (1.1)
17	4-22-68	111.0 (1.2)							
18	4-22-68	115.0 (0.5)	90.3 (1.4)	523.0 (1.9)	90.4 (0.7)	13.8 (2.5)	1.77 (1.2)	0.493 (7.4)	0.107 (4.5)
19	6-28-68	10.8 (0.7)		45.2 (4.0)	9.40 (0.6)				
20	6-29-68	112.0 (1.1)	63.0 (0.7)	537.0 (0.4)	92.8 (1.1)	13.9 (1.0)	2.15 (1.9)		
21	6-30-68								
22	7-1-68								

[a] Values stated in parentheses indicate the precision of the measurement in percent standard deviation.

[b] Values enclosed in brackets are determined by direct counting without pretreatment.

[c] Values given for Gross β are determined by direct counting without pretreatment.

Table II. (Continued)

Sample No.	Date	^{85}Kr	HT	CH_3T	C_2H_5T	C_3H_7T	$^{14}CO_2$	$^{14}CH_4$	Gross β^c
23	7-2-68	92.0 (1.0)	42.5 (0.4)	472.0 (0.9)	86.8 (3.2)				
24	7-3-68								
25	7-4-68	99.2 (2.4)	44.9 (0.4)	468.0 (1.7)	87.1 (1.1)				
26	7-7-68	90.4 (1.0)	54.1 (0.7)	483.0 (0.4)	76.9 (2.3)				
27	7-8-68								
28	7-9-68								
29	7-10-68	84.1 (1.4)	41.3 (0.8)	436.0 (0.4)	79.8 (1.5)				
30	7-11-68	41.3 (4.2)		212.0 (4.1)	31.4 (1.0)				
31	7-12-68	66.4 (1.1)	32.0 (2.7)	262.0 (0.8)	42.3 (4.2)		0.91 (1.0)		
32	7-13-68								
33	7-14-68	48.4 (0.7)	29.9 (0.4)	309.0 (1.0)	55.1 (1.2)				
34	7-22-68	3.98 (0.7)	0.11 (12)	14.6 (0.9)					
35	7-22-68								
36	9-18-68	33.0 (1.4)	15.1 (0.8)	191.0 (0.6)	29.3 (0.9)	6.78 (5.6)	0.80 (4.7)	0.54 (1.3)	
37	9-18-68								
38	11-7-68								
39	11-8-68	63.6 (1.2)	41.2 (0.6)	292.0 (0.9)	49.6 (3.2)		1.15 (2.9)	0.36 (1.8)	406 (4.4)
40	11-15-68								
41	11-22-68	56.5 (0.4)	32.2 (10.9)	309.0 (3.5)	55.8 (0.8)		1.35 (1.8)		504 (1.4)
42	11-29-68								
43	12-6-68								
44	12-7-68	54.6 (0.5)	31.0 (1.2)	311.0 (0.4)	35.0 (2.1)		1.08 (9.4)		488 (2.4)
45	12-10-68	64.2 (0.3)	30.0 (1.0)	299.0 (1.5)	51.2 (0.4)		1.31 (0.3)		
46	12-13-68	36.8 (0.7)	33.2 (1.0)	178.0 (1.5)	31.8 (2.3)		0.70 (0.5)		310 (5.7)
47	12-12-68	54.8 (1.3)	27.4 (0.9)	276.0 (1.7)		7.85 (0.4)	1.11 (3.0)		

Table II. (Continued)

Sample No.	Date	^{85}Kr	HT	CH_3T	C_2H_5T	C_3H_7T	$^{14}CO_2$	$^{14}CH_4$	Gross β^c
48	12-19-68								
49	12-26-68	41.4 (0.8)	22.5 (4.1)	242.0 (8.0)	42.1 (3.9)		1.30 (0.8)		352 (5.7)
50	1-2-69								
51	1-11-69	48.9 (1.9)	19.9 (4.6)	254.0 (6.3)	37.5 (3.7)		0.92 (0.7)		369 (3.0)
52	1-14-69	50.8 (0.5)	18.5 (15)	244.0 (0.7)	44.8 (0.4)		0.88 (0.9)		403 (0.8)
53	1-17-69	49.0 (0.6)	41.0 (4.1)	236.0 (1.7)	44.5 (6.0)		1.28 (1.2)		374 (1.9)
54	1-27-69								
55	2-3-69	36.7 (0.5)	19.0 (1.3)	171.0 (0.8)	30.1 (0.9)		0.62 (0.6)		285 (6.4)
56	2-10-69								
57	2-18-69	32.2 (0.8)	18.6 (14)	137.0 (1.6)	26.1 (0.4)		0.73 (0.6)		232 (0.6)
58	2-25-69	37.6 (2.2)	19.6 (4.0)	168.0 (1.0)	32.7 (0.4)	3.18 (0.4)	0.74 (36)	0.45 (5.3)*	294 (8.0)
59	3-5-69	[35.3 (6.0)]							234 (0.4)
60	4-11-69	17.7 (0.4)	18.5 (4.0)	73.0 (6.4)	13.9 (1.6)	2.91 (6.2)	0.49 (5.1)	0.49 (7.1)	127 (3.4)
61	4-18-69	[16.6 (0.7)]							116 (0.6)
62	4-25-69	17.0 (1.0)	16.3 (4.0)	63.8 (1.1)	9.83 (3.3)		0.68 (3.1)		112 (0.6)
63	5-9-69	[15.3 (3.7)]							103 (1.5)
64	5-16-69	14.0 (2.0)	6.90 (4.0)	54.6 (1.8)	11.7 (2.0)				96.1 (0.4)
65	6-6-69	[12.1 (1.7)]							82.4 (0.4)
66	6-12-69	12.0 (1.3)		46.4 (2.0)	11.2 (6.6)	2.09 (2.1)		0.673 (9.1)	78.2 (0.4)
67	6-27-69	[11.5 (1.1)]							80.7 (0.7)
68	7-4-69	13.1 (0.9)	6.52 (6.9)	43.1 (0.4)	9.29 (0.8)	1.77 (1.3)	0.56 (5.5)		71.7 (0.4)
69	7-11-69	[10.7 (3.2)]							73.2 (0.7)
70	7-18-69	8.83 (1.7)		39.5 (2.8)	9.53 (1.0)	2.34 (2.5)			70.9 (0.1)
71	8-1-69	[10.1 (2.0)]							71.9 (1.5)
72	8-8-69	8.69 (3.4)		37.0 (0.4)	8.86 (1.3)		0.33 (4.7)		68.2 (0.4)
73	8-15-69	[9.10 (1.9)]							66.1 (1.1)
74	8-21-69	8.76 (0.5)	4.50 (24)	34.5 (0.4)	7.09 (5.1)			0.614 (12.)	62.1 (0.2)
75	8-29-69	[8.56 (2.2)]							63.2 (5.4)

Chemical Composition

Chemical analysis of Gasbuggy samples was performed by mass spectrometry. Results of these analyses are listed in Table III for chimney (or formation) gas after air (based on oxygen) had been removed from the sample. The probable accuracy of the reported results is 1 to 5 units in the last reported figure or less than a few percent for the major constituents listed. Included as a group under the heading "other" in Table III are the hydrocarbon fractions C_4 and higher. The method of sampling was not conducive to a meaningful determination of their concentration in the nuclear chimney.

As a consequence of the method of routine snap-sampling, a small quantity of air is collected along with the chimney gas. In some cases the sample bottle leaked prior to sampling and, in the case of the cable samples (samples 1 through 4) the sample was predominately air. To make meaningful comparisons between samples, the concentrations of the chimney gas components have been re-normalized to 100 percent. Assuming the standard composition for the air impurity of

N_2 = 78.03%
O_2 = 20.99%
Ar = 0.94% ,

it becomes apparent that the fraction of a given sample which is truly air based on oxygen is

$$f(\text{air}) = f(O_2)\left[1 + \frac{78.03}{20.99} + \frac{0.94}{20.99}\right].$$

Subtracting $\left\{\frac{78.03}{20.99}\right\} \cdot f(O_2)$ from the N_2 determination and performing a similar operation for the Ar produces an un-normalized distribution of compositions for the chimney gas. Normalization is accomplished by multiplying this distribution by an air correction obtained from the relation

$$\text{Air correction} = \frac{1}{1 - f(\text{air})} .$$

Values of the air correction obtained in this manner are listed in Table III. Significant corrections have been made for only a few samples. Uncertainties for these samples should also be multiplied by the air correction factor. Excess nitrogen appears in nearly all samples indicating that it is probably a true component of the gas and not an artifact of these manipulations. O_2 and Ar are removed quantitatively. The extremely large corrections applied to samples 1 through 4 made an exception to the above treatment necessary. For samples 2 and 3, the air correction described above yielded gas compositions which included some 20-percent excess nitrogen. Because this value probably resulted from the correction process, the N_2 was subtracted and the residual compositions were re-normalized to yield the composition listed in Table III. This latter re-normalization was not applied to the data in Table I of the Gas Quality Status Report.[3] The values listed here are, therefore, upper limits for the concentrations of the components of chimney gas in the cable leak samples. Samples 1 and 4 were about 99 percent air. No useful information would result from re-normalization. Only the percentage nitrogen in the sample gas is indicated in Table III.

Table III. Chemical composition of LRL Project Gasbuggy samples in vol % as determined by mass spectrometric analysis.

Sample No.	Date	Air correction	N_2	CO_2	CO	H_2	CH_4	C_2H_6	C_3H_8	Other
1	12-10-67	large	(78.4)							
2	12-11-67	7.187		9.87	21.7	23.1	42.6	1.73		0.96
3	12-11-67	5.652		9.92	21.7	22.6	43.5	1.75		0.47
4	12-10-67	large	(78.2)							
5	1-13-68	1.045	0.64	36.2	3.9	16.7	36.4	3.61	1.21	1.15
6	1-13-68	1.016	0.99	36.1	3.7	16.9	36.5	3.63	1.15	1.09
7	1-13-68	1.010	0.22	35.0	4.1	17.4	36.5	3.52	1.25	1.97
8	1-17-68	1.013	0.90	34.6	4.0	18.0	37.2	3.42	1.01	0.77
9	1-17-68	1.002		35.8	4.5	16.8	37.5	3.63	1.14	0.79
10	1-17-68	1.018	0.53	36.0	3.9	16.7	37.2	3.68	1.27	0.79
11	1-17-68	1.012	0.61	35.6	3.9	17.0	37.7	3.56	0.97	0.63
12	2-27-68	1.018	0.71	34.8	3.1	14.4	40.0	4.10	1.13	1.78
13	2-27-68	1.001		35.6	3.3	14.8	40.4	4.10	1.23	0.67
14	2-27-68	1.010	0.62	35.7	2.6	14.8	39.9	4.12	1.21	0.98
15	2-27-68									
16	2-27-68	1.002	0.45	35.6	2.6	15.0	40.2	4.12	1.17	0.84
17	4-22-68	1.001	—	35.4	2.2	13.1	43.2	4.57	1.05	0.53
18	4-22-68	1.000	—	35.1	2.2	13.3	43.3	4.50	0.98	0.61
19	6-28-68	1.008	0.31	4.39	—	0.27	82.5	7.31	2.71	1.87
20	6-29-68	1.002	1.32	36.2	—	12.0	44.2	4.70	0.97	0.61
21	6-30-68	1.006	—	34.0	1.5	12.2	45.9	4.73	1.09	0.74
22	7-1-68									
23	7-2-68	1.002	1.13	33.5	—	11.1	46.8	4.78	1.18	0.74
24	7-3-68									
25	7-4-68	1.002	1.05	33.1	—	10.8	47.8	4.92	1.31	1.01

Table III. (Continued)

Sample No.	Date	Air correction	N_2	CO_2	CO	H_2	CH_4	C_2H_6	C_3H_8	Other
26	7-7-68	1.000	1.10	32.2	—	10.5	48.9	4.97	1.37	0.93
27	7-8-68									
28	7-9-68									
29	7-10-68	1.013	1.01	30.9	—	9.9	49.8	5.06	1.58	1.08
30	7-11-68		0.13	14.9		5.5	67.7	5.96	2.47	1.37
31	7-12-68	1.000	—	18.9	0.7	6.80	64.0	5.80	2.38	1.38
32	7-13-68									
33	7-14-68	1.001	0.79	21.7	—	6.72	60.9	5.77	2.38	1.77
34	7-22-68	1.001	0.49	0.70	—	0.20	83.4	7.94	4.42	2.83
35	7-22-68									
36	9-18-68	1.000	0.42	13.0	—	5.8	69.8	6.07	2.98	1.96
37	9-18-68									
38	11-7-68									
39	11-8-68	1.000	0.43	22.0	—	7.92	59.7	5.66	2.52	1.72
40	11-15-68									
41	11-22-68	1.001	0.49	23.1	—	8.11	58.6	5.50	2.24	1.88
42	11-29-68									
43	12-6-68									
44	12-7-68	1.000	0.42	22.7	—	8.10	59.9	5.50	2.18	1.21
45	12-10-68	1.072	1.16	22.5	—	7.70	59.2	5.62	2.40	1.44
46	12-13-68	1.000	0.33	14.1	—	4.62	70.0	6.24	3.02	1.67
47	12-12-68		0.4	26.6		9.25	74.8	6.94	3.19	2.75
48	12-19-68									
49	12-26-68	1.709	2.85	16.7	—	5.81	64.5	5.94	2.89	1.26
50	1-2-69									

Table III. (Continued)

Sample No.	Date	Air correction	N_2	CO_2	CO	H_2	CH_4	C_2H_6	C_3H_8	Other
51	1-11-69	1.079	0.60	18.8	—	6.37	64.2	5.96	2.73	1.38
52	1-14-69	1.001	0.29	20.1	—	6.72	63.0	5.86	2.60	1.39
53	1-17-69	1.000	0.40	19.0	—	6.20	63.9	6.00	2.80	1.64
54	1-27-69									
55	2-3-69	1.000	0.40	14.2	—	4.67	69.8	6.31	3.09	1.50
56	2-10-69									
57	2-18-69	1.000	0.46	13.0	—	4.33	71.1	6.51	3.05	1.50
58	2-25-69	1.000	0.50	16.1	—	5.02	66.9	6.44	3.14	1.91
59	3-5-69	1.032	0.39	14.6	—	4.25	68.3	6.67	3.35	2.44
60	4-11-69	1.001	—	9.90	—	2.60	73.9	7.10	3.90	2.72
61	4-18-69	1.002	0.56	9.42	—	2.54	73.7	7.01	3.97	2.81
62	4-25-69	1.014	0.42	9.44	—	2.55	74.3	6.96	3.91	2.45
63	5-9-69	1.133	2.15	9.29	—	2.46	73.2	6.90	3.98	2.04
64	5-16-69	1.034	0.72	8.60	—	2.28	74.7	6.94	3.94	2.86
65	6-6-69	1.001	—	7.92	—	2.06	76.0	7.07	4.09	2.82
66	6-12-69	1.020	0.81	7.97	—	2.15	75.4	6.95	3.98	2.74
67	6-27-69	1.000	0.40	8.10	—	2.10	75.0	7.0	4.0	3.4
68	7-4-69	1.003	0.40	7.73	—	1.96	76.1	7.08	4.07	2.67
69	7-11-69	1.034	0.77	7.55	—	1.98	75.9	6.97	4.07	2.76
70	7-18-69	1.002	0.34	7.72	—	1.96	76.1	7.05	4.12	2.70
71	8-1-69	1.000	0.3	7.56	—	2.01	76.2	7.02	4.11	2.80
72	8-8-69	1.003	0.40	7.48	—	1.98	76.2	7.03	4.13	2.73
73	8-15-69	1.001	0.40	7.4	—	1.97	76.3	7.02	4.14	2.77
74	8-21-69	1.018	0.45	7.23	—	1.90	76.4	7.06	4.18	2.76
75	8-29-69	1.002	0.4	7.1	—	1.86	76.4	7.05	4.18	3.0

Miscellaneous Determinations

Many investigations of the gas quality of Gasbuggy chimney gas do not lend themselves to a generalized, tabulated report such as that preceeding this section. These topics, of general interest to the Gas Quality Program, will be presented here. No particular order of topics is anticipated, and additional topics may be added as results become available.

RARE GAS ACTIVATION PRODUCTS

Neutrons escaping the nuclear explosive at the time of detonation interact with components of the rock to produce activation products. Two of these products are isotopes of argon and are, therefore, mixed with the chimney gases. ^{37}Ar produced by $^{40}Ca(n, \alpha)$ ^{37}Ar and ^{39}Ar produced by $^{39}K(n, p)$ ^{39}Ar have been identified in the Gasbuggy chimney. Because of their half lives (35 days and 270 years respectively), neither isotope poses a gas quality problem comparable to the tritium and ^{85}Kr. Both species were determined by thin-window beta-proportional counting, in a manner comparable to ^{85}Kr. ^{37}Ar was observed through bremsstrahlung following electron capture, the beta of ^{39}Ar was counted directly, assuming a counting efficiency comparable to that for ^{85}Kr (10 percent). All results are extrapolated to the time of detonation, and are listed in Table IV.

^{37}Ar and ^{39}Ar exhibit a somewhat random variation or "bounce" comparable to that observed for tritiated hydrogen. Although this observation cannot be explained clearly, it does appear to be real. Because of the assumption of 10-percent counting efficiency for ^{39}Ar, these results are considered less accurate than others listed here, but are probably within 20 percent of the correct values, with the deviation being toward the low side.

Table IV. Rare gas activation product concentrations for Project Gasbuggy in picocuries/cc STP extrapolated to the time of detonation.[a]

Sample No.	Date	^{37}Ar		^{39}Ar	
2	12-11-67	2340	(4.2)	0.0516	(11)
3	12-11-67	7660	(3.1)	0.0868	(5.0)
5	1-13-68	6202	(12)	0.0938	(5.4)
6	1-13-68	4380	(21)	0.0945	(1.0)
7	1-13-68	3420	(1.9)	0.0772	(3.4)
10	1-17-68	4410	(1.5)	0.0903	(0.9)
11	1-17-68	3920	(2.0)	0.0771	(0.8)
14	2-27-68	4990	(0.9)	0.0948	(1.6)
16	2-27-68	4260	(1.4)	0.0829	(1.5)
18	4-22-68	5622	(1.4)	0.0945	(3.1)
Average		4720	(10%)	0.0844	(14%)

[a]Values in parentheses indicate the precision of measurement in percent standard deviation.

Table V. Tritiated water results—Project Gasbuggy.

Sample No.	Date	Sample type	Total pCi HTO	pCi HTO / cc H_2O	pCi HTO / liter (STP) sample
2	12-11-67	S	0.344	18.5	0.22
3	12-11-67	S	0.150	16.7	0.10
5	1-16-68	D	2,610.0	1,710.0	43.9
6	1-16-68	D	381.0	297.0	6.94
7	1-13-68	S	6.89	58.6	0.75
8	1-17-68	S	19,500.0	703.0	2,170.0
9	1-18-68	D	126,000.0	49,500.0	2,170.0
10	1-18-68	D	44,100.0	24,300.0	747.0
11	1-17-68	S	11.5	135.0	0.77
12	2-27-68	D	6,570.0	15,500.0	110.0
13	2-27-68	S	0.321	18.9	0.02
14	2-27-68	S	12.3	135.0	0.61
16	2-27-68	D	76,900.0	118,000.0	1,220.0

TRITIATED WATER

The problem of obtaining meaningful samples of water from a nuclear chimney has no easy solution. Surface samplers are clearly inaccurate due to the interposition of some thousands of feet of pipe between source and samplers. Downhole samplers present a more acceptable means of obtaining water samples from the chimney. Unfortunately, condensation of non-chimney water on the surfaces of the sampler, and the engineering difficulties in physically entering the chimney with the sample bottle, prevented proper sampling. As the result of dilution of the chimney water so obtained, the concentrations of tritiated water determined in a gas sample have little significance and specific activities of the water so recovered are lower limits. Table V lists the results in units of total picocuries tritium as HTO recovered, picocuries per cc of water recovered, and picocuries per liter (STP) of sample. No correlation between sample volume, sample type, amount of water and amount of activity is evident. For this reason water sampling with the gas has been discontinued.

The establishment of a continued flow from the chimney region results in a kind of equilibrium being established within the sample pipe thereby permitting water of more nearly the specific activity of the chimney water to reach the surface. Dilution is, of course, still a potential problem and concentrations of tritiated water must still be considered as lower limits. Analysis of water removed at the surface during production testing has shown specific activities of more than 1 μCi/cc of water. The Eberline Instrument Corporation has been performing routine analyses of tritiated water collected in this manner

during production testing. Their reports should be consulted for details and analytical results.

A few such water samples, obtained during production testing of GB-ER, have been analyzed for tritium and for gamma-ray emitting radionuclides. The samples were collected from the liquid removed from the produced gas at the wellhead by the knock-out system, and were provided by C. Bowman of the El Paso Natural Gas Company.

Tritium concentration was determined by liquid scintillation counting of an aliquot of the sample as provided. The results of these determinations are tabulated in Table VI.

A search for the presence of gamma-ray-emitting radionuclides in these samples has been made. We examined the 10 samples (about 15 ml each) over the energy range of 0 to 2.5 MeV, using a 256-channel analyzer and a cylindrical NaI crystal 3 in. in diameter and 3 in. deep. No significant activity above background was observed. An upper limit of 0.03 pCi/ml was established for the presence of these nuclides.

NONGASEOUS RADIONUCLIDES

A cursory examination of some early gas samples was made in an attempt to detect various potentially volatile nuclides which might have been in the gas. As expected, no indication of the presence of these species was found on or in downhole sample bottles, or in gas withdrawn from the well. Upper limits were established for their concentration by gamma counting. It was assumed that 0.1 times the background in the seven channels surrounding the photopeak energy for the nuclides

Table VI. Project Gasbuggy GB-ER production testing of water samples.

Sample No.	Date	Time	Tritium Concentration[a] μCi/ml
1	7-29-68[b]	1:30	1.16
2	7-6-68		1.15
3	1-17-69		0.264
4	1-31-69	3:00	0.196
5	2-18-69	1:30	0.125
6	2-27-69	3:00	0.124
7	3-10-69	3:00	0.049
8	3-17-69	9:30	0.104
9	3-27-69		0.124
10	4-2-69	2:30	0.018

[a]Absolute uncertainty less than ±10% of number given.

[b]Probably 6-29-68.

Table VII. Nongaseous radionuclides for Project Gasbuggy, upper limits of pCi/cc STP extrapolated to time of detonation.

Nuclide	$t_{1/2}$ (days)	pCi/cc STP (maximum)
^{127}Sb	3.9	2.3
^{131}I	8.05	0.04
^{125}Sn	9.4	4.5
^{129m}Te	33	1×10^{-3}
^{103}Ru	41	8×10^{-5}
^{125m}Te	58	0.23
^{123}Sn	131	2×10^{-2}
^{106}Ru	367	1×10^{-4}
^{125}Sb	985	2×10^{-4}
^{137}Cs	10^4	2×10^{-5}

would have produced a detectable peak. Details of this investigation are to be published.[4] Field investigations performed by Eberline Instrument Corporation did not detect the presence of any gamma-emitting nuclides other than rare gases.[5] Table VII lists the lowest upper limits obtained by gamma-spectrometry in the LRL investigation. The most volatile of these species, ^{131}I, was present in concentrations of less than 0.04 pCi/cc STP. The number of ^{131}I atoms/cc may present a more probable upper limit than those listed which exceed this concentration.

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The Gas Quality Analysis and Evaluation Program for Project Gasbuggy has drawn upon the talents of a large number of people to produce the results tabulated here. In particular, the efforts of the following groups and individuals deserve recognition.

Mass spectrometry for both chemical analysis of samples and chemical purity of separated fractions was performed by Richard W. Crawford and the Analytical Chemistry Section of the LRL General Chemistry Division.

Sample procurement was a combined effort of many LRL Chemical Engineering, LRL Hazard Control, and Eberline Instrument Corporation personnel.

Radiochemistry was performed by the Gas Analysis Group of the LRL Radiochemistry Division under the immediate supervision of Dr. Floyd F. Momyer.

In addition, the continued interest of the project participants, El Paso Natural Gas, U.S. Bureau of Mines, and the Atomic Energy Commission, as well as valuable discussions with their representatives and with colleagues at LRL, has been of great help to the continuing effort to obtain and evaluate these results.

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