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Dr. Herbert J. Kouts
Post Office Box 625
Brookhaven, New York 11719

Dear Dr. Kouts:

Attached is a copy of a report prepared by the USAEC Health and Safety Division, Idaho Falls, Idaho, dated September 17, 1964, and entitled "Radiochemical Analysis of the United Nuclear Incident at Wood River Junction, Rhode Island". This is one of the reports identified as outstanding on pages 25-27 of Volume 3 of our investigation report. As additional information is received it will be furnished you and members of your Committee.

Very truly yours,

Original signed by
L. D. Low

Lawrence D. Low, Director
Division of Compliance

Enclosure:
Idaho Falls Report
dated 9-17-64

cc: Dr. Wayne C. Bills
Dr. Richard L. Dean
Dr. Marvin M. Mann
Dr. Hugh C. Paxton
Dr. Warren E. Winsche

bcc: Harold L. Price, REG
Lyall Johnson, DML
Eber R. Price, SLR
Forrest Western, SS
R. W. Kirkman, CO:I, w/o encl.

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U. S. ATOMIC ENERGY COMMISSION
Health and Safety Division
Idaho Falls, Idaho

RADIOCHEMICAL ANALYSIS OF THE UNITED NUCLEAR INCIDENT AT WOOD RIVER JUNCTION, RHODE ISLAND

September 17, 1964

Report Prepared by:

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U. S. Atomic Energy Commission
Health and Safety Division
Idaho Falls, Idaho

RADIOCHEMICAL ANALYSIS OF THE UNITED NUCLEAR INCIDENT AT WOOD RIVER JUNCTION, RHODE ISLAND

I. Introduction

An uncontrolled nuclear reaction occurred at the uranium-235 scrap recovery facility operated by United Nuclear Corporation at Wood River Junction, Rhode Island. The accident occurred at 1405 July 24 when an operator poured the contents from a 5-in. diameter cylindrical bottle into an 18-in. diameter vessel. Samples of the uranium solution involved in the incident, metal objects from the vicinity of the vessel, metal objects from the film badge of the operator and coins from a person making a post entry were analyzed to determine the magnitude and spectrum of the nuclear incident and the doses received by individuals.

II. Counting Procedure

Gamma-ray spectrometry employing a standard 3 in. x 3 in. NaI (Tl) crystal coupled with a 400-channel pulse height analyzer was used to determine absolute disintegration rates. Standards obtained from the National Bureau of Standards and Nuclear Chicago Corporation with a certified counting uncertainty of less than 2% were analyzed along with samples.

III. Analytical Procedures

A. Liquid Samples

The solution from the reaction vessel was analyzed for barium 140, cerium 141, molybdenum 99, total and isotopic uranium. Cerium was separated from other radioactive isotopes by precipitation with

barium sulfate and reprecipitation as cerium peroxide [1]. Molybdenum was extracted into ethyl acetate as the alpha benzoinoxime complex and precipitated as lead molybdate for counting [2]. Barium required no chemical separation because the 1.6-mev gamma ray of the lanthanum-140 daughter of barium 140 was free of interference and the results could be calculated directly by gamma analysis of aliquots from the original solution. Uranium was determined spectrophotometrically by directly measuring the uranyl tetrapropylammonium nitrate complex extracted into hexone.[3] The uranium isotopic distribution was measured on a surface ionization mass spectrometer.

B. Metals

Metallic samples from the vicinity of the reaction vessel were examined for induced activity. Mild steel objects including the screwdriver, film badge clip, spring and rivets were analyzed for iron 59 and manganese 54 produced from the thermal Fe 58 (n, γ) Fe 59 and fast Fe 54 (n, p) Mn 54 neutron reactions. Because no other activation products deriving from alloy steel were present, the mild steel was known to have the conventional composition of about 99% iron. This conclusion was substantiated by X-ray analysis and chemical separation on the screw driver and by X-ray analysis on the film badge clip. In addition to the analyses for iron 59 and manganese 54, stainless steel objects were also analyzed for chromium 51 and cobalt 58 produced from the thermal Cr 50 (n, γ) Cr 51, and fast Ni 58 (n, p) Co 58 neutron reactions. The two pieces of hose clamp were found by X-ray fluorescence (see attachment #1) to be type-316 stainless steel with composition Fe - 71%, Cr - 17%, Ni - 11%, Mn 1 to 2%, Mo trace.

Samples were dissolved in perchloric acid. Chromium was distilled off as chromyl chloride, precipitated as barium chromate and counted. Iron was extracted into isoamyl acetate from 9 M hydrochloric acid, precipitated and ignited to the oxide for counting. Cobalt was extracted as the thiocyanate complex into methylisopropylketone, precipitated as cobaltic oxide and counted. Manganese was oxidized to permanganate with divalent silver, selectively precipitated as tetraphenylarsoniumpermanganate, and counted.

C. Chemical Composition of Yellow Solid from Reaction Vessel.

A yellow crystalline precipitate that had separated from the reaction material accompanied the liquid samples and was analyzed to determine total fissions. The material was easily soluble in dilute acid with copeous evolution of a gas, undoubtedly carbon dioxide. Later, a request was received to determine the chemical composition of this sample. Only small quantities of the solid material could be obtained from the first liquid samples received, making analysis very difficult. Since the yellow material was obviously a carbonate containing uranium as a major component, it seemed likely that the compound is moderately soluble sodium uranyl carbonate with a formula suggested to be $Na_4UO_2(CO_3)_3.2H_2O$. [4] (3) A yellow crystalline precipitate was produced in the laboratory by addition of a strong sodium carbonate solution to a solution of uranyl nitrate. As near as could be determined, this synthetic compound was identical to the unknown sample. The samples were anisotropic and had the same refractive index. X-ray diffraction studies gave inconclusive results because of the poorly-crystalline characteristics of both compounds.

IV. Constants

The constants used in the calculations are given in Tables I and II and are believed to be the best values available.

TABLE I.

Nuclide	Sample Volume	Half Life	Fission Yield	Branching Ratio	Decay Correct.	Counting Position(cm)	*1/TPA
Ba 140	0.5 ml	12.8 d	6.35%	0.88	3.03	10 cm	222
Ce 141	1.0 ml	32.5 d	6.00%	0.48	0.48	3	6.82
Mo 99	1.0 ml	66 h	6.06%	0.163	varied	3	23.0

TABLE II.

Nuclide	Sample Volume	Half Life	Cross Section	Branching Ratio	Sensitivity mgu/Abs.U	Counting Position(cm)	*1/TPA
Fe 59		45 d	1.0	0.44		3	38.7
Mu 54		314 d	0.065	1.0		3	25.7
Cr 51		27.8d	13.5	0.09		3	10.4
In 114m		50 d	56	0.035		3	22.1
Ag 110		249 d	3.2	0.72		3	27.0
U	0.1 ml				25		

* A composite factor for the gamma-ray measured containing the absolute detection efficiency, peak-to-total ratio, and correction for a 760 mg cm^{-2} polystyrene beta absorber. Values were taken from a table of gamma-ray conversion factors.(2) To calculate disintegrations per minute the following expression can be used:

$$d/m \text{ @ } To = \frac{(\text{Integrated area})(\frac{1}{\text{T.P.A.}})(\text{Decay Correction})}{(\text{Branching Ratio})}$$

V. Data

Since all samples were analyzed by gamma spectrometry, the integrated area under the index photopeak is given in Table III for the energy range listed.

For liquid samples, the total number of fissions within a given bottle varied significantly when measured by different nuclides. Considering the precipitate formation in the gallon bottles, the sample splitting and subsequent dilution, fractionation of the various fission products is most likely. Gamma spectra showing the different ratios of fission products present in two different bottles of solution are given in Attach. 2 as conclusive proof of this fractionation. However, the total fissions obtained as the sum of the fissions in each bottle shows excellent agreement among the results obtained from the three fission products analyzed. The data for the metallic samples is given in Table IV.

VI. Conclusions

A. Total Fissions

Summation of the total number of fissions found in 26 different solutions by analysis for 3 different fission product nuclides agreed among themselves to within about 2%. Assuming 40% of the solution was lost during the incident, an average of 2.1×10^{17} total fissions is obtained.

Induced activity from Fe 54 (n,p) Mn 54 reaction in the metal samples used to calculate fissions gave the following results, assuming 2.5 neutrons are released per fission, the escape probability from

TABLE III.

Bottle Number	Bottle Volume ml.	Ba 140 $\sum$ 1.73 / 1.54	Ba 140 Fissions (X 10^{+5})	Ce 141 $\sum$ 0.170 / 0.110 (X 10^{+5})	Ce 141 Fissions (X 10^{+5})	Mo. 99 Decay Corr.	Mo. 99 Chem. Yield	Mo. 99 $\sum$ 0.820 / 0.700	Mo. 99 Fission (X 10^{+15})	U Abs.	U Gms
HA1	1,874	33,486	10.4	1.67	7.8	420	0.783	602	8.6	.286	136
HA2	1,211	42,057	8.39	1.97	6.1	540	.580	577	9.2	.321	98.3
HA3	865	53,474	7.23	2.25	4.9	195	.307	1,050	8.2	.375	82.1
HA4	932	36,771	5.67	1.50	3.7	545	.527	438	6.0	.301	71.0
HA15	1,718	38,405	10.9	1.25	5.8	420	.543	218	4.1	.303	132
HA17	1,162	34,678	6.63	2.55	7.4	195	.650	980	4.8	.202	59.4
HA18	1,507	41,050	10.2	2.31	8.9	195	.650	1,205	8.1	.265	101
HA20	1,256	40,041	8.29	1.93	6.2	420	.947	773	6.1	.317	101
HA21	1,854	35,482	10.9	1.61	7.6	420	.650	595	10.0	.281	132
HA1-1C9	1,886	45,767	13.4	1.81	8.7	196	.527	1,480	15.0	.315	150
HA2-1C9	1,102	9,427	2.57	.91	2.7	545	.783	137	1.5	.117	32.6
HA3-1C9	1,403	6,736	1.56	.74	2.2	420	.413	200	4.1	.100	35.5
HA4-1C9	740	6,501	.75	.78	1.5	195	.610	245	8.2	.093	17.4
HB1	1,649	8,346	2.27	1.31	5.4	420	.650	190	2.9	.142	92.2
HB2	1,731	5,299	1.51	1.04	4.5	195	.543	125	1.1	.109	47.8
HB3	1,977	5,804	1.79	1.17	6.0	196	.703	192	1.5	.112	55.9
HB4	1,087	3,425	.58	.85	2.3	194	.853	85.4	.30	.093	25.5
HB15	1,955	6,010	1.95	1.14	5.7	540	.820	81.5	1.5	.134	66.3
HB17	2,181	1,356	.46	.102	.57	194	.670	450	4.5	.108	59.5
HB18	2,537	3,932	1.66	.78	4.8	420	.547	77.9	2.2	.136	87.3
HB20	1,516	12,286	3.06	1.30	5.0	420	.650	383	5.3	.168	64.4
HB21	2,275	7,322	2.75	.89	5.5	545	.717	89.6	2.2	.133	76.4
HB1-1C9	1,911	34,106	10.8	1.66	8.6	545	.850	611.5	11.0	.255	123
HB2-1C9	2,196	3,220	1.16	.94	5.3	194	.747	63.5	.51	.086	47.9
HB3-1C9	1,746	2,678	.78	.66	3.1	545	.683	30.2	.61	.084	37.2
Solids	(20.694 g)	11,235	.19	.39	.11	550	.753	21.9	.23	.095	2.40
Total			1.26 x 1017		1.30 x 1017				1.28 x 1017		1,934 gms

$\sum$ - events per minute in the index photopeak

U 235 - 94.75%
U 234 - 1.00%
U 236 - .26%
U 238 - 5.62%

TABLE IV.

Nuclide	Sample Wt., g	Decay Corr.	Id/Io	Σ	Determined Flux (nvt)
Screwdriver Chemically Separated					
Fe 59	8.49	1.24	0.86	54.5	3.4×10^{12}
Mn 54	8.49	1.03		200.7	1.9×10^{13}
Screwdriver Scanned as a 1" Disc					
Fe 59	9.53	1.22	0.75	3217	2.9×10^{12}
Mn 54	9.53	1.03	0.68	11468	2.0×10^{13}
Film Badge Clip					
Fe 59	1.6	1.17		9.96	1.7×10^{12}
Mn 54	1.6	1.02		19.43	5.1×10^{12}
Film Badge Spring					
Fe 59	.594	1.52		3.87	2.2×10^{12}
Mn 54	.594	1.05		9.1	6.7×10^{12}
Film Badge Pins					
Fe 59	.596	1.52		4.4	2.6×10^{12}
Mn 54	.596	1.05		9.9	7.1×10^{12}
Hose Clamp 9" from Vessel					
Fe 59	.697	1.30		6.88	2.9×10^{12}
Mu 54	.697	1.02		9.83	5.64×10^{12}
Cr 51	.178	1.53		285	2.43×10^{12}
Co 58	.110	1.16		82.3	5.17×10^{12}
Hose Clamp 174" from Vessel					
Fe 59	.708	1.34		0.5	2.0×10^{11}
Mn 54	.708	1.04		0.28	1.6×10^{11}
Cr 51	.178	1.60		14.7	2.4×10^{11}
Co 58	.110	1.19		2.54	1.6×10^{11}
Indium Foils					
#1	.212	1.15		344	1.0×10^{12}
#2	.221	1.15		372	1.0×10^{12}
Silver Coins					
dime	2.23	1.10	.88	4.68	2.2×10^{9}
dollar	17.7	1.10	.93	20.4	1.4×10^{9}

$\frac{Id}{Io}$ - absorption of gamma rays in the source

the vessel is 0.5, and the geometric change follows an inverse square relationship:

hose clamp at 174 in 5.1×10^{17} fissions;

hose clamp at 9 in. 1.5×10^{17} fissions;

and then screwdriver, 1.7×10^{17} fissions.

B. Flux Ratio

Each object containing iron was subject to thermal Fe 58 (n, γ) Fe 59 and fast Fe 54 (n,p) Mn 54 neutron reactions which are used to find the flux ratios. A thermal flux buildup is present near objects which are good moderators. Each sample analyzed was subject to unknown buildup errors due to a plastic handle, a hose, or a body. The following values of the fast-to-thermal flux ratios have not been corrected for this buildup and are therefore biased by some unknown values:

screwdriver, - 5.6;

film badge clip, -.3.0;

film badge spring,- 3.0;

film badge pins, - 2.7;

hose clamp 9" - 1.8;
and
hose clamp 174", - 0.8.

C. Dose

1. From Initial Incident

Dimensions of the critical assembly (18 in. diam. x 16 in. high) are very comparable to the Y-12 mock-up (20 in. diam. x 16 in. high) and therefore the escape spectrum should be similar The ratio of fast-to-thermal neutron flux was determined to be 2.3 and the ratio of gamma-to-neutron dose was 2.9.[5]

The induced activity found by analyzing the indium foil in the film badge resulted from a thermal flux of 1.0×10^{12} nvt. assuming a thermal reflection from the body to be 1.8 and correcting for self shielding in the indium, the incident thermal flux would be 6×10^{11} nvt. Assuming the spectrum to be like the Y-12 mock-up the total dose is calculated to be 1×10^{4} rads. The R.B.E. of the Y-12 mock-up has been postulated to be 0.98, which could be used to convert to rems if desired.

2. From Delayed Entry

Silver coins from the pocket of the person making the post entry contained induced activity from the nuclear reaction Ag 109 (n, γ) Ag 110. Values determined for the incident flux required to produce the activities observed and the calculated body dose assuming the same flux spectrum as listed above is 39 rem in addition to the gamma dose resulting from exposure to fission products.

Literature Cited

(1) Sill, C. W., Willis, C. P., New procedure being prepared for publication.

(2) Olson, D. G., U. S. Atomic Energy Commission Report, IDO-14613, January 1964.

(3) W. J. Maeck, J. E. Rein, US AEC Report IDO-14316, Supplement #4, U-Color-5, page 107.

(4) Sidgwick, N. V., "The Chemical Elements and their compounds," Vol. II, Oxford University Press, 1950, page 1078.

(5) G. S. Hurst, R. H. Ritchie, L. C. Emerson, Health Physics Vol. II, pages 121-133, 1959.

Acknowledgement

The radiochemical analyses were made by D. G. Olson, R. W. Henry, C. P. Willis, C. F. T. Ching, R. W. Stone and J. I. Anderson. Calculations were made by R. W. Henry and D. R. Davie*; and other spectrographic analyses were made by D. B. Hawkins and the Analytical Chemistry Branch of Phillips Petroleum Co.

* Research Officer, Australian AEC

Attachment #1

PHILLIPS PETROLEUM COMPANY
ATOMIC ENERGY DIVISION
CPP ANALYTICAL SECTION
SPECTRAL ANALYSIS

Method	X-ray fluorescence	Log No.	GJ-4122
Date Run	8-5-64	Codo	Hose Clamps
Record No.	7-39	Activity	
Cell		Requested By	
Solvent		Report Results To	D Olson
Reference		Sample Description	
Sample Disposal	To	Method Suggested	
Run By		Date Submitted	
Calculated By		Charge No.	GJ412
Approved	R	Determinations	X-ray

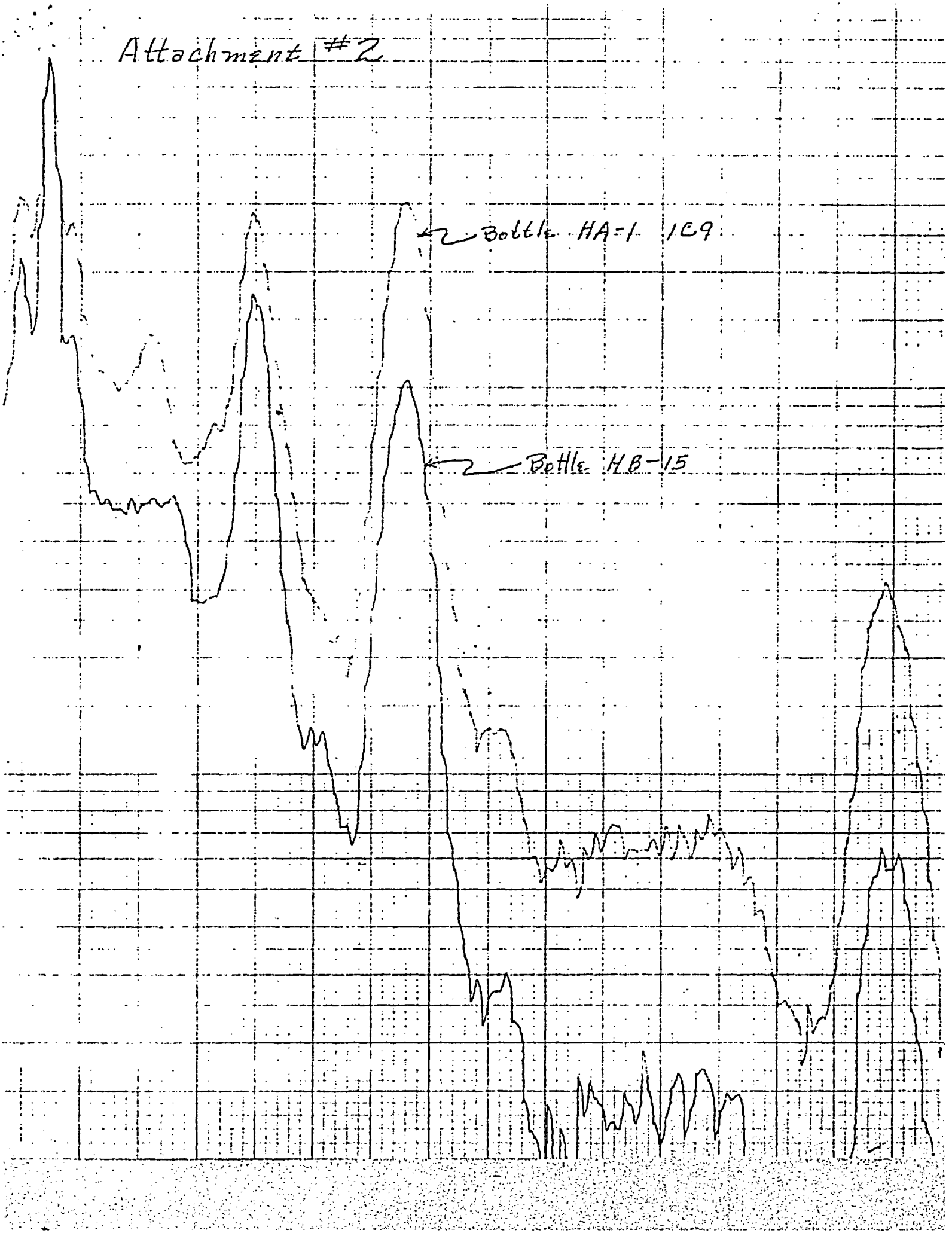
2 Sections of SS hose clamps.

Both sections are 316-type Stainless Steel.

Section #2 was analyzed for the following:

Cr = 17.1 % } ± 10% of these reported values.
Ni = 10.9 % }

Mn = 1 to 2%

Mo = detected

Attachment #2