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Dr. H. W. Russell
Assistant to the Director
Battelle Memorial Institute
505 King Avenue
Columbus, Ohio

Dear Dr. Russell:

As discussed at our last visit at Battelle, we will submit our monthly progress letter to you as of the 20th of each month. We feel that in this, the first letter, it will be worthwhile to review briefly for your information the work done on the bismuth distillation project to date with emphasis on the materials-of-construction phase of the problem.

Our problem is to separate the one to two parts per million of polonium formed upon irradiating high-purity bismuth in a nuclear reactor. The recovery of polonium should be as near quantitative as possible; in the order of 99.5 per cent would probably suffice. The ultimate goal of the project is to obtain polonium purities greater than 95 per cent in the product, although product concentration ratios 100 times greater than the feed would be useful and could be considered as an immediate goal if greater concentration factors could not readily be obtained.

In general, two schemes of performing the distillation are being considered. The first method approximates an ordinary batch fractionation. The active bismuth would be heated in a still pot to perhaps 800°C at low pressure and the vapors fractionated in a column to obtain relatively pure polonium at the top. The degree of concentration attained by this method could probably be increased to any desired value by increasing the complexity of the column. This, however, would be at the expense of decreasing the processing rate.

The second method would not depend on fractionation but rather on the differential or simple distillation of polonium from bismuth. Unfortunately, the relative volatility of polonium from a polonium-bismuth mixture is approximately 0.01 times the theoretical value obtained from the vapor pressures of the two substances. However if the degree of concentration need not be greater than 100, two stages of differential distillation should suffice.

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Data that have been obtained on the relative volatility of polonium compared to bismuth indicate that it is reduced from approximately 100 to 25 as the temperature increases from 600 to 800°C and that it also decreases when the percentage of bismuth that is vaporized becomes appreciable. The rate of vaporization of bismuth, however, increases approximately ten-fold with each 100°C increase in temperature. These factors are important in setting the operating temperature of the process. Additional factors which affect the design and operation of any still include necessity for and means to agitate the molten charge, method of heating, operating pressure, and many others.

It has become apparent in working on this project that, while certain of the above problems involving the actual distillation of polonium and bismuth remain to be solved, perhaps the most important single difficulty is finding a material of construction suitable for the still. The answer to this problem is needed not only for the ultimate operating still but also for the experimental units being built to obtain distillation data.

An ideal material would have the following characteristics:

1. It should be capable of containing bismuth up to 900°C or higher under reduced pressure without adding impurities to the bismuth and without being affected by the bismuth.

2. It should be capable of containing polonium under the above conditions without alloying or combining with the polonium.

3. It should be readily available and capable of being machined, formed, and welded.

4. It should have a low vapor pressure.

5. It should withstand air cooling from 1,000°C and water quenching from 600°C without deleterious changes in the material.

6. It should not be oxidized by air at 800°C to 1,000°C.

7. It should be nonmagnetic to permit electromagnetic stirring if required.

8. It should not be affected by nitric acid to allow periodic leaching.

It is extremely doubtful whether a material embodying all these qualities will ever be found, so that any material chosen will no doubt represent a compromise. Inconel was originally selected because it is resistant to high temperatures and corrosion; is machinable, weldable, and easily formed; is available in standard shapes; is nonmagnetic; and is unaffected by polonium and nitric acid. Initial tests showed no effect from molten bismuth, but it later became apparent that these tests were not sufficiently prolonged nor at high enough temperatures to be conclusive. Data published jointly by the Atomic Energy Commission and the Navy (Liquid-Metals Handbook, A.E.C. and Dept. of the Navy, June, 1950, Nevexos P - 733) and a report by the American Smelting and Refining

Company (Everhart, J.L. and Van Nuis, E.L., American Smelting and Refining Company, Research Dept., Progress Report No. 1, September 29, 1950) indicated that Inconel would probably not be satisfactory for this application. Neither publication was available in time to be of any value other than to verify our difficulty with Inconel. It might be pointed out that considerable disagreement exists between the two reports mentioned above and an Oak Ridge National Laboratory report (ORNL-754) on the effect of molten bismuth on various metals.

After the failure of the 10-inch-diameter Inconel still, a low-carbon steel unit of the same design was constructed and was operated about 50 hours at approximately 800°C. Chrome plating the outside of the still avoided external corrosion. At the end of this time the walls and bottom of the unit had been deformed by the combination of high temperature and internal vacuum to the extent that the still could no longer be used. When this still was dismantled, the interior did not show any evidence of attack by bismuth, and analysis of the bismuth showed only 0.01 per cent iron present. Several smaller test stills of varying types have also been constructed of low-carbon steel and operated for short periods of time with satisfactory results.

The testing of materials of construction at Mound Laboratory in the hope of finding an ideal material has centered on low-carbon steel or Armco Ingot Iron, some miscellaneous materials, and the alloys furnished by Battelle following our first visit there. Low-carbon steel was selected because of its ready availability and the fact that in general the references mentioned above list good resistance for iron and steel to liquid bismuth.

The results are given in Table I for a series of corrosion tests conducted by heating bismuth in low-carbon steel cylinders one foot long by 3/4 inch internal diameter. The tubes, which were mounted vertically, were heated electrically over the lower third of their length, and the upper portions were air cooled. As in all our tests the cylinders were evacuated continuously, and in this case the residual atmosphere was nitrogen. At the conclusion of each test the bismuth was poured out under an atmosphere of nitrogen. It was then pulverized and sampled and its iron content determined by the thiocyanate method.

After each pair of tests the cylinders were sectioned and examined. In all cases a brittle deposit above the heated zone was observed. The iron content of this deposit, which was magnetic, is shown in Table I under "Iron in Condensation Ring". The deposit had the appearance of an alloy, but, instead of fusing when heated to about 1,000°C, bismuth flowed out in small globules, leaving a residue of the same form and size as the original piece. X-ray diffraction analysis on this material showed only metallic iron and bismuth.

The results of some early corrosion tests in quartz containers are listed in Table II. Duriron showed excellent resistance but has not been used because of fabricating difficulties. Although not reported in this table, it was found that titanium formed pyrophoric alloys with bismuth.

The results obtained on short time tests are given in Table III for the alloys that you furnished to us. These samples were heated in bismuth in an evacuated quartz tube with residual helium atmosphere. From these results it

was felt that only the S-590, 19-9DL, and Haynes-Stellite No. 25 showed sufficient promise for further investigation. The results obtained of heating these three alloys in quartz tubes for 100 hours with a residual helium atmosphere are shown in Table IV. The S-590 and the 19-9DL continued to show excellent corrosion resistance and should be thoroughly investigated. We plan to make some tests to determine if these alloys will combine with polonium.

Very truly yours,

Malcolm M. Haring

Malcolm M. Haring
Laboratory Director

MMH:gl

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TABLE I

CORROSION OF LOW-CARBON STEEL BY MOLTEN BISMUTH

Test No.	Bismuth Charged (grams)	Temperature (°C.)	Vacuum (mm.Hg.)	Duration of test (hours)	Bismuth Recovered (grams)	Iron in Bismuth (Per Cent)	Iron in Condensation Ring (Per Cent)
1	100	980	1	24	81.	0.005	---
2	100	980	1	24	93.	0.005	28.
3	100	850	1	48	73.7	0.006	---
4	100	850	1	48	78.0	0.003	15.5
5	100	850	1	78	78.0	0.05	---
6	100	850	1	78	tube failed when weld opened		30.
7	100	850	0.7	100	80.0	0.2	---
8	100	850	0.7	100	85.8	0.24	26.**
9*	100	850	0.7	100	97.	0.08	0.04

* Sample 3.34 square centimeters in area tested in quartz tube.

** Weight of condensation ring, 17.5 grams.

TABLE II

BISMUTH CORROSION TESTS ON MISCELLANEOUS METALS

Test Metal	Temperature (°C.)	Vacuum (microns)	Time (hrs.)	Remarks
Tantalum	850	50	100	The 1 in.^{2} sheet of 6 mil Ta lost 51.5 mg. The immersed portion was very brittle.
Alnicol	850	50	24	During the test, needles appeared on the bismuth surface and solidified. A 35 degree increase in temperature permitted continuation. Ni, Fe, Cu, Co, were present in the bismuth.
Molybdenum	900	50	190	All the bismuth had condensed in the cool part of the chamber at the end of the test. A spectrographic trace of molybdenum was present in the bismuth. The immersed part of the molybdenum wire was brittle.
Duriron Test 1	850	50	72	0.002% iron was found in the bismuth pool, and 0.001 mg. in the bismuth mirror on the walls of the container.
Test 2	850	50	100	A trace of iron and molybdenum was present in the bismuth. The 25 g. test piece lost 21.5 mg.

N.B. 100 g. of bismuth was charged in each test.

TABLE III

Material of Construction Tests

Sample Name-ASM Code	Weight grams	Loss in Weight, g.	Pressure microns	Time Min.	Temp. °C
Haynes Stellite, 25	9.8431	0.0026(gain)	<1	30	960
Haynes Stellite, S-816	6.9670	0.5830	1.0	35	1000
Super Alloy, S-590	9.7126	0.0174	1.0	35	840
Super Alloy, 19-9DL	6.3391	0.0037	1.5	45	900
Vitallium	4.0553	0.3631	<1	37	980
Hastalloy, B	9.6620	—	<1	60	950
Stainless Steel, 304	12.2201	0.1473	<1	60	950
" " 347	12.0450	0.6072	<1	60	960
" " 316	5.2381	—	<1	60	950
" " 314	12.6036	0.1956	<1	60	925
" " 446	7.0940	0.0145	<1	30	900
" " 321	17.9412	0.2317	<1	60	950
" " 310	12.9649	0.1244	<1	60	950
" " 309	14.3350	0.2020	<1	60	950

TABLE III (cont.)

		Haynes Stellite		Super Alloy		Vitallium	Hast-Alloy B
		25	S-816	S-590	19-9DL		
Reported Composition of alloy %	C	*	0.35-0.45	0.4-0.5	0.25	0.22	0.05
	Mn		1.0	2.0	0.5	0.66	0.59
	Si		1.0	1.0	0.60	0.53	0.19
	Cr	*	18.5-20.5	18.5-20.5	19	27.4	
	Ni	*	19-21	19-21	9	2.8	65
	Co	*	42-44	19-21		62.2	
	Mo		3.5-4	3.5-4	1.25	5.5	28.6
	Cb		3.5-4	3.5-4	0.3		
	Ti				0.2		
	W	*	3.5-4	3.5-4	1.2		
	Fe	*	Rem.	Rem.	Rem.	0.7	4.7
Final Bismuth Comp., %	C					None	
	Mn		0.02	Trace	None	0.11	0.063
	Si			None	None	None	None
	Cr	None	0.045	None	None	0.054	0.015
	Ni	None	0.16	Trace	None	None	0.135
	Co		0.06	0.001		0.05	
	Mo				None	None	None
	Fe	None	0.04		Trace	0.10	0.33

* Amount unknown

** Approximately 100 grams of bismuth used in each test

TABLE III (cont.)

STAINLESS STEEL

		304	347	316	314	446	321	310	309
Reported Composition of alloy %	C	0.08	0.08	0.10	0.10	0.35	0.08	0.25	0.20
	Mn	2.0						2.0	2.0
	Si								
	Cr	18-20	17-19	16-18	25	23-27	17-18	24-26	22-24
	Ni	8-11	9-12	10-14	20		8-11	19-22	12-15
	Co								
	Mo			2-3					
	Cb		0.8						
	Ti						0.4		
	W								
	Fe	Rem.	Rem.	Rem.	Rem.	Rem.	Rem.	Rem.	Rem.
Final Bismuth Comp., %	C	None	None	None					
	Mn	0.05	0.04	0.05					
	Si								
	Cr	0.02	0.09	0.013	0.009	0.001	0.009	0.016	0.007
	Ni	0.014	0.077	0.074	0.06		0.02	0.02	0.03
	Co								
	Mo			None					
	Fe	1.4	7.5	0.70	0.10	0.08	0.04	0.04	0.08

** Approximately 100 grams of bismuth used in each test

TABLE IV

BISMUTH CORROSION TESTS ON HIGH TEMPERATURE ALLOYS

Alloy	Haynes Stellite No. 25		S-590		19-9-DL	
Weight before	8.7785 g.		9.3925 g.		8.8000 g.	
Weight after	8.6225		9.3947		8.7370	
Loss	0.1560 g.		0.0022 gain		0.0630 g.	
Corrosion rate	0.0678 g./in.^{2}/ 100 hrs.		(0.907 in.^{2} area)		0.0532 g./in.^{2}/ 100 hrs.	
Bismuth Charge	100 g.		100 g.		100 g.	
Temperature °C.	880		875		890	
p. in microns	12		12		12	
Appearance After test	Etched		Corners alloyed		No change	
Analysis of the bismuth for	Spectro.	Chem.	Spectro.	Chem.	Spectro.	Chem.
Fe	Not taken	trace	Weak	0.03%	Trace	none
Ni	Not taken	0.05	Weak	0.016	Trace	none
Co	Not taken	0.06	Weak			
Si			Trace		Weak	0.007%
Mn			Trace			none
Cr		0.04	Trace	0.015		none
Mo			Trace			none
W			none		none	
Nb			none		none	
Ag			Trace			
Al			Trace			
Mg			Trace			
Pb			Trace		Trace	
Ti					none	