Title
THE EFFECT OF GASES ON THE WETTING OF STEELS BY LIQUID BISMUTH

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Publication Date
1951-01-17

UCRL-1094

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THE EFFECT OF GASES ON THE WETTING OF STEELS BY LIQUID BISMUTH

R. A. Heckman

January 17, 1951

Berkeley, California

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The Effect of Gases on the Wetting of Steels by Liquid Bismuth

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January 17, 1951

Abstract

The effect of air, nitrogen, helium, argon, and a mixture of twenty volume percent of hydrogen in argon on the wetting of a stainless steel (Type 446), a two percent chromium-one half percent molybdenum type steel, and a plain carbon type steel by liquid bismuth was investigated.

Irreversible wetting may be obtained by preheating the steel to 1500°F in a helium atmosphere and then immersing into molten bismuth at 1000°F. Reversible wetting (i.e., de-wetting takes place) may be obtained by preheating the steel to 1500°F in an argon atmosphere and then immersing into liquid bismuth.

The problem of obtaining wetted surfaces is of unusual importance in liquid-metal heat transfer. Published data[1] indicated one method by which metal surfaces could be wetted, but the technique required a high vacuum. A solution to the problem was attempted through the use of either an inert atmosphere or a reducing atmosphere, at normal barometric pressure. The results obtained are primarily of engineering importance for a limited range of steel compositions.

Apparatus

The apparatus consisted of a three inch diameter pyrex pipe tee, with the side arm extended, around which an R.F. heating coil was wound (See Fig. 1). Power was supplied from a two kilowatt input electronic radio-frequency generator, of one-half megacycle frequency. The R.F. coil was of water-cooled, one-quarter inch nominal diameter, copper tubing. The flange brackets for the pyrex tee were constructed of bakelite to eliminate excessive heating by the R.F. field generated. Wilson seals were inserted into the bakelite brackets to allow a direct means for mechanical immersion of the samples into the liquid bismuth, this mechanical apparatus also being constructed of bakelite (See Fig. 2). A Cenco-Megavac Pump was used to evacuate the system. All gases were dried by passing through a drying tube containing indicating Drierite. Temperatures of the samples were measured by means of a Leeds and Northrup optical pyrometer.

Three methods of sample immersion into the liquid bismuth were used. In the first method, the sample of steel was inserted into a piece of transite, 1.0" x 0.15" x 0.25", through a 0.13" diameter hole. The dimensions of the steel samples were 1.3" x 0.13" x 0.03". The transite-held sample was then placed in the mechanical immerision apparatus, and after preheating, the steel was lowered into the liquid bismuth, held in a two milliliter pyrex beaker. The second technique used was that of placing the steel sample in a pyrex beaker, packing small pieces of solid bismuth around it, and then melting the bismuth while in contact with the steel sample. The third technique made use of a small beaker with a vertical side arm. The steel sample was placed in the side arm, and small pieces of solid bismuth were packed into the beaker. (See Fig. 3). When placed in the R.F. field, the steel would preheat to the desired temperature, and then the melting bismuth, rising into the side arm, would come in contact with the sample. Three of these beakers were stacked into the pyrex tee at one time, using pyrex mounting stands. (See Fig. 4).

The sample and the bismuth container were placed into the tee through the side arm, and the bakelite flange bracket rebolted on. The tee was evacuated and flushed three times with the gas under consideration, and then a slow stream of the gas was passed through the tee. The R. F. field was then turned on, the temperature of the preheated steel noted with the optical pyrometer, and then the sample immersed into the liquid bismuth. The R.F. field was turned off, the sample allowed to cool to room temperature, removed from the apparatus, and then observed to see if wetting had taken place.

Experimental Results

A total of thirty-four runs were made under the varying conditions of oxidizing, reducing, and inert atmospheres. The gases utilized for these conditions were air, both at one atmosphere and 4 mm. Hg pressure, nitrogen, helium, argon, and a mixture of twenty volume percent of hydrogen in in argon, all at one atmosphere pressure. After immersion in liquid bismuth and cooling to room temperature, the samples were observed macroscopically, under 3X magnification, to determine if wetting had taken place. The results of the tests are given in Table I.

The results with air are to be expected, since both the steel and bismuth exhibited surface oxidation, even at a pressure of 4 mm. Hg where the oxygen partial pressure is approximately 0.8 mm. Hg.

When heated to 1500°F in a nitrogen atmosphere, all the steel samples were coated with a black material. Submission of a sample of this black coating to an X-ray diffraction test revealed that the coating was amorphous. A summary of the diffraction report is given in the appendix. It is quite probable that this was an iron nitride coating, that prevented any wetting from occurring.

In a helium atmosphere, if the steel is preheated to 1500°F and then immersed into molten bismuth at 1000°F, irreversible wetting takes place, i.e., dewetting does not take place on cooling down to room temperature. No wetting was ever obtained with the Type 446 stainless steel because the magnetic susceptibility of the steel was so low, due to its austenetic structure, that the R.F. field was never able to produce a temperature in excess of 1000°F. The data obtained indicates that a preheating temperature of at least 1500°F is necessary to obtain wetting. This agrees quite well with published results[1].

Table I

Effect of Gases on Wetting of Steels by Liquid Bismuth at 600°F

Type of Steel	Air	Nitrogen	Helium	Argon	Argon + Hydrogen
plain carbon	no wetting	no wetting	irreversible wetting*	reversible wetting and irreversible wetting*	no wetting
2.0 chromium + 0.5 molybdenum	no wetting	no wetting	irreversible wetting*	reversible wetting	no wetting
stainless steel type 446	no wetting	no wetting	no wetting	no wetting	no wetting

* Liquid Bismuth at 1000°F.

In an argon atmosphere, a plain carbon steel can be wet irreversibly by preheating to 1500°F and then immersing into liquid bismuth at 1000°F. Reversible wetting can be obtained by preheating the steel to 1500°F and then immersing into liquid bismuth. The term reversible wetting implies that the steel is wetted by liquid bismuth at higher temperatures, but dewets on cooling to room temperatures. The occurrence of reversible wetting is strongly indicated by three experimental observations. First, the contact angle of the meniscus between liquid bismuth and steels at elevated temperatures indicated that wetting did take place. Secondly, small pieces of bismuth still adhered strongly to the steel after cooling to room temperature. Thirdly, examination of the bismuth surface from which the steel had broken showed the presence in spots of a very thin non-metallic coating; the surface of the bismuth was a mirror image of the steel, showing file marks and surface irregularities. The thin non-metallic coating, mentioned above, usually was more pronounced near the surface of the bismuth.

All materials used were of commercial grade, with no attempt to further purify them. The bismuth was 99.4 percent pure. The presence of gaseous impurities in the gases tested, and especially of oxygen, could cause erroneous results. The presence of dissolved gases in the bismuth metal[1], could explain, in part, the dewetting observed in some of the experimental runs. Since the average time of a single run in these experiments was three minutes, a longer immersion time might give wetting, and thus alter some of the negative results obtained in these tests.

Appendix

1. Photographs of Apparatus
2. Nominal Compositions of Steel Samples
3. Summary of the X-ray Diffraction Report
4. Explanation of Data Sheets
5. Summary of Experimental Data
6. Bibliography

Figure Captions

Fig. 1. Glass Tee Apparatus. Note rf coil wound on side arm.

Fig. 2. Mechanical Immersion Apparatus. The immersion unit is attached to a bakelite rod inserted through a Wilson seal. The system is purged by means of the needle valve. The Wilson seal and needle valve are both attached to the bakelite flange bracket.

Fig. 3. Beaker with Vertical Side Arm. The steel sample is placed in the side arm and pieces of bismuth are packed into the beaker, as shown in the right hand beaker. After melting, the bismuth rises into the side arm and immerses the sample, as shown in the left hand beaker.

Fig. 3a. Wetted and Unwetted Samples. Note wetted portion on bottom of left-hand sample as compared with unwetted sample on right.

Fig. 4. Pyrex Mounting Stands.

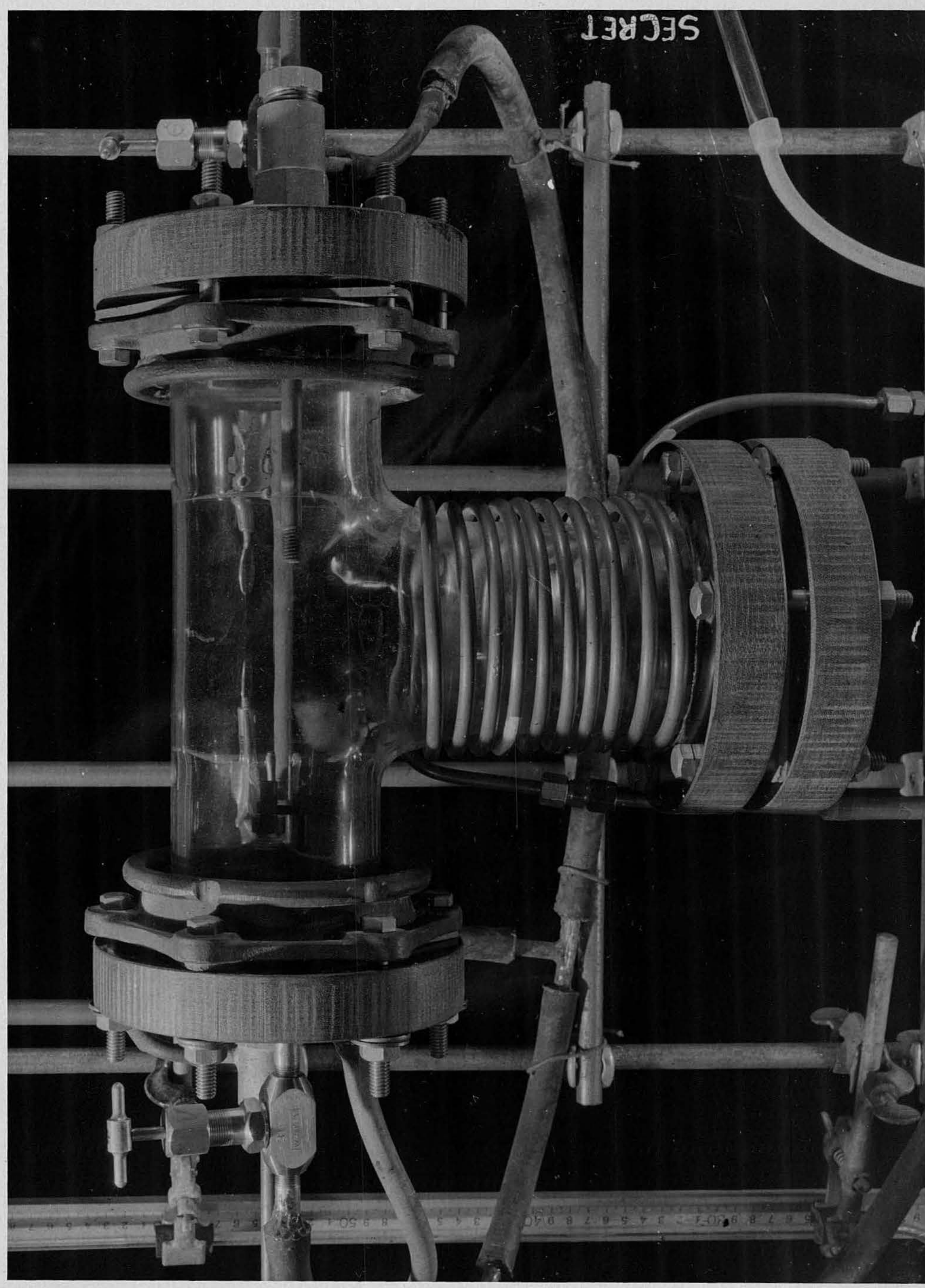

FIG. 1

OZ 1088

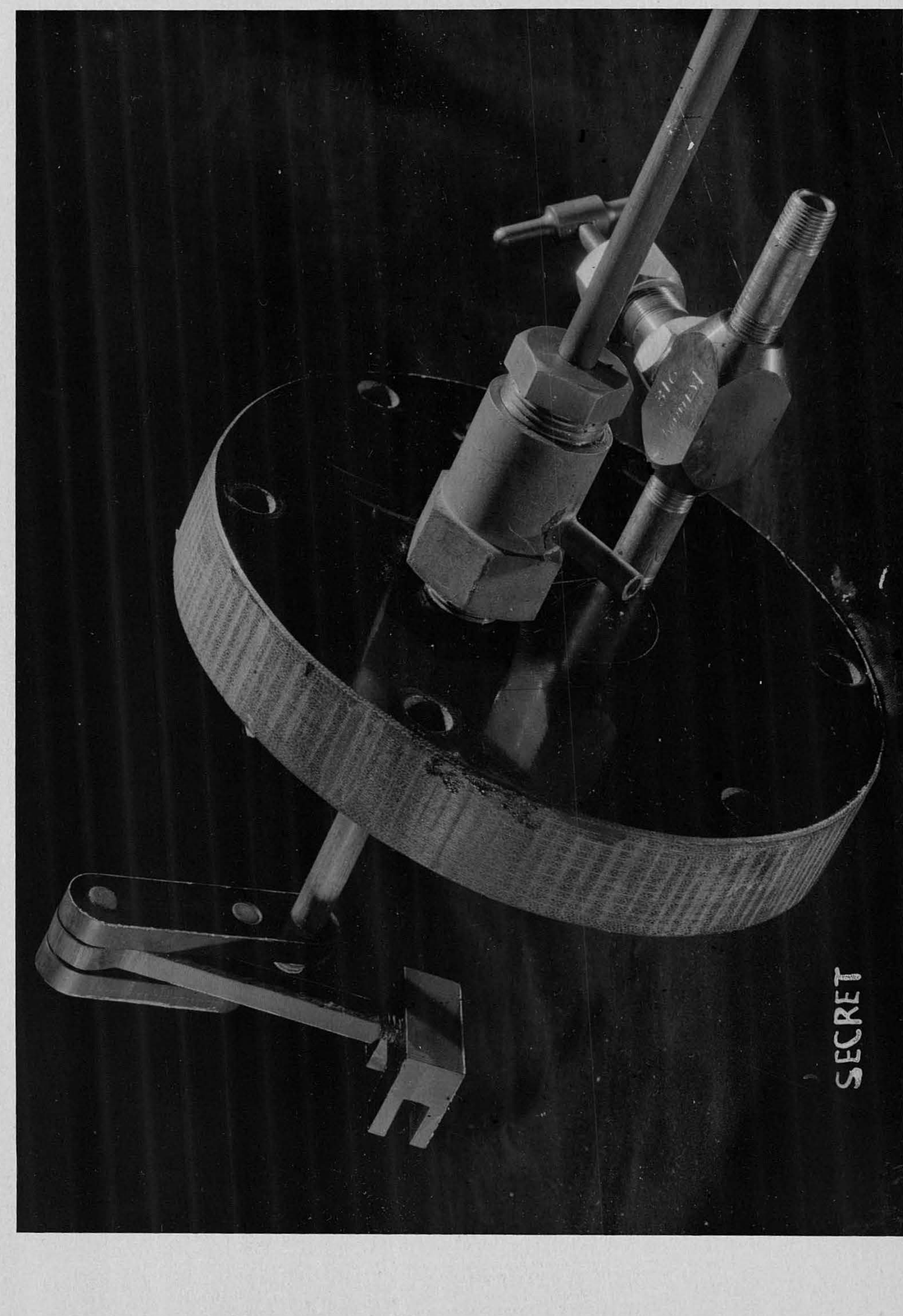

FIG. 2

OZ 1090

FIG. 3

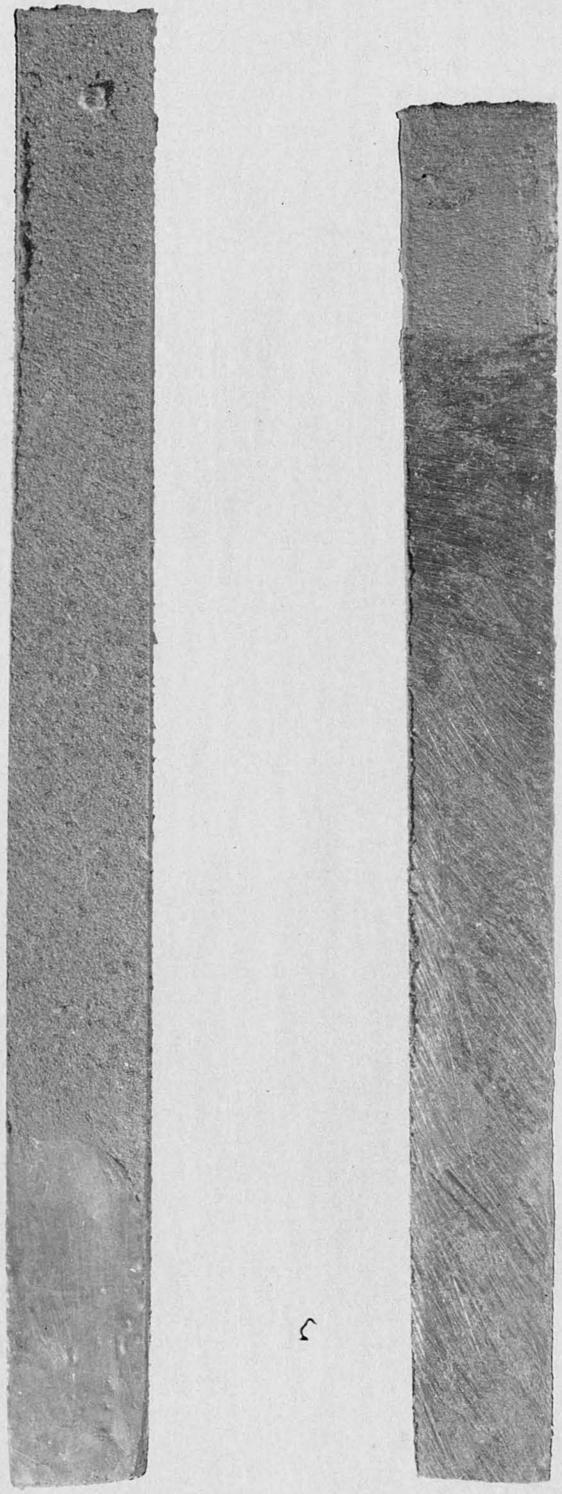

FIG. 3A

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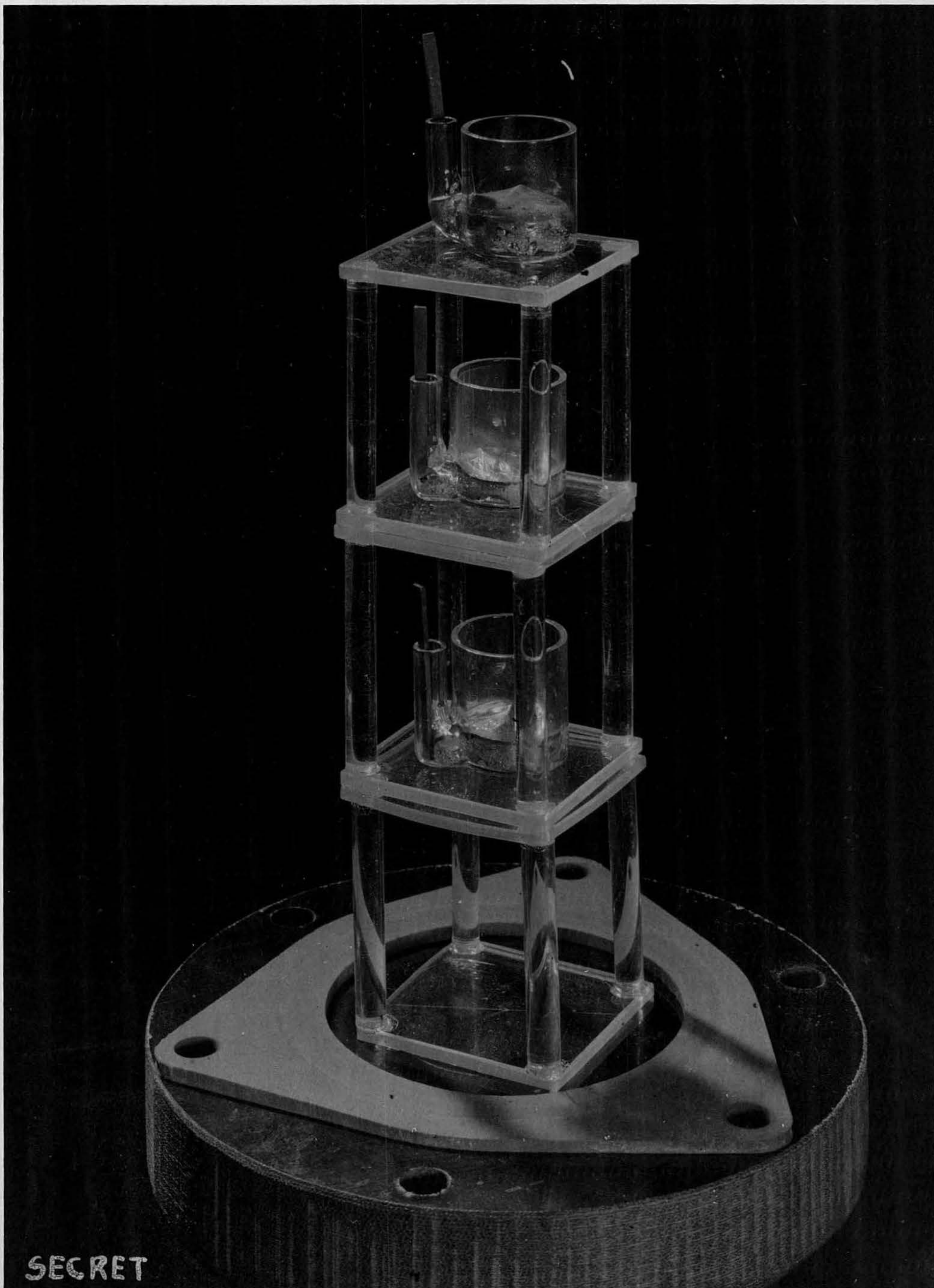

FIG. 4

Nominal Compositions of the Steel Samples[2]

Type		
1	Plain carbon steel	SAE 1010
2	Chromium-Molybdenum Steel	2.0 percent Chromium 0.5 percent Molybdenum
3	Stainless Steel Type 446	0.35 max Carbon 23.00 - 30.00 Chromium

Summary of X-ray Diffraction Report

Sample Number: RH 1 Material: Steel

Description: Pyrex cap

Film Numbers: 3398, 3399, 3402

Intensity strong

Quality sharp

Remarks: The coating was not crystalline, so did not show in the picture.

Explanation of Abbreviations and Column Headings Used on Data Sheets

1. Steel type: Refers to nominal composition of steel sample. For full explanation of numbering system, reader is referred to preceding section in Appendix entitled "Nominal Compositions of Steel Samples."

2. Surface Pretreatment:

   A. Physical: This connotation means that all excess oxidation products were filed off the samples leaving a bright, shiny surface.

   B. Chemical: Refers to a chemical process of removing further oxidation products from sample surface and involves the following steps:

      1. Wash sample in six molar hydrochloric acid.
      2. Rinse in distilled water.
      3. Rinse a second time in distilled water.
      4. Rinse in carbon tetrachloride.
      5. Dry under slight vacuum.

3. Temperatures of liquid bismuth and steel samples: All temperatures given are in degrees Fahrenheit. Those temperatures followed by a small letter m were actually measured by means of the optical pyrometer and have been corrected by an emissivity value of 0.37 for iron and the correction chart given in Kehl[3], page 326. All other temperatures were estimated.

Explanation of Abbreviations and Column Headings

Used on Data Sheets (cont.).

4. Experimental Handling Techniques: This column indicates the method of sample immersion into the liquid bismuth. Mech. Imm. means that the sample was mechanically lowered into the liquid bismuth. In contact with Melt. Bi. means the sample was packed in a beaker with solid bismuth, and was in contact with the bismuth as it melted. Imm. by rising level means that the sample was placed in the side arm beaker and immersion was obtained by the molten bismuth rising into the side arm. For more details the reader is referred to the second paragraph of the section entitled "Apparatus" in the body of the report.

5. Wetting: Yes refers to obtained irreversible wetting. Rev. means irreversible wetting was observed. No means that no wetting was obtained.

Summary of Experimental Data

Run	Steel Type	Atmos-phere	Pressure	Surface Pretreatment Physical	Surface Pretreatment Chemical	Temp. Bi	Temp. Steel	Experimental Handling Techniques Mech. Imm.	Experimental Handling Techniques In Contact With Melt. Bi	Imm. by Rising Level	Wetting
1	1	air	1 atm	yes	yes	600	1500	yes	no	no	no
2	2	air	1 atm	yes	yes	600	——	no	yes	no	no
3	2	air	4 mm Hg	yes	yes	600	——	no	yes	no	no
4	3	air	1 atm	yes	yes	600	1000	yes	no	no	no
5	1	N_2	1 atm	yes	yes	600	1485m	yes	no	no	no
6	2	N_2	1 atm	yes	yes	600	1485m	yes	no	no	no
7	3	N_2	1 atm	yes	yes	600	1000	yes	no	no	no
8	1	N_2	1 atm	yes	yes	600	1400	no	yes	no	no
9	2	N_2	1 atm	yes	yes	600	1400	no	yes	no	no
10	3	N_2	1 atm	yes	yes	600	1000	no	yes	no	no
11	1	He	1 atm	yes	yes	600	1300	no	yes	no	no
12	2	He	1 atm	yes	yes	1000	1300	no	yes	no	no
13	3	He	1 atm	yes	yes	600	900	no	yes	no	no
14	1	N_2	1 atm	yes	no	600	1500	no	no	yes	no
15	2	N_2	1 atm	yes	no	600	1500	no	no	yes	no
16	3	N_2	1 atm	yes	no	600	1000	no	no	yes	no
17	1	He	1 atm	yes	no	1200	1465m	no	no	yes	yes

Summary of Experimental Data (cont.)

Run	Steel Type	Atmosphere	Pressure	Surface Pretreatment Physical	Surface Pretreatment Chemical	Temp. Bi	Temp. Steel	Experimental Handling Techniques Mech. Imm.	Experimental Handling Techniques In Contact With Melt. Bi	Imm. by Rising Level	Wetting
18	2	He	1 atm	yes	no	600	1465	no	no	yes	no
19	3	He	1 atm	yes	no	600	1000	no	no	yes	no
20	2	He	1 atm	yes	no	1000	1488m	no	no	yes	yes
21	3	He	1 atm	yes	no	1000	1000	no	no	yes	no
22	1	A	1 atm	yes	no	600	1450	no	no	yes	rev.
23	2	A	1 atm	yes	no	600	1450m	no	no	yes	rev.
24	3	A	1 atm	yes	no	600	1000	no	no	yes	no
25	1	A	1 atm	yes	no	1200	1465	no	no	yes	yes
26	2	A	1 atm	yes	no	1200	1465m	no	no	yes	rev.
27	3	A	1 atm	yes	no	1000	1000	no	no	yes	no
28	2	A	1 atm	yes	no	1200	1481m	no	no	yes	rev.
29	1	A + H_2	1 atm	yes	no	600	1485	no	no	yes	no
30	2	A + H_2	1 atm	yes	no	600	1485m	no	no	yes	no
31	3	A + H_2	1 atm	yes	no	600	1000	no	no	yes	no
32	1	A + H_2	1 atm	yes	no	900	1488	no	no	yes	no
33	2	A + H_2	1 atm	yes	no	1300	1488m	no	no	yes	no
34	3	A + H_2	1 atm	yes	no	900	1000	no	no	yes	no

Bibliography

1. Stanford Research Institute - Final Report VII, June 1, 1949 - May 31, 1950, Navy Contract N6ONR25120 Project NR031361, Investigation of materials for use in a heat transfer system containing molten lead alloys.

2. Metals Handbook, ASM (1939).

3. Kehl, G. L., The Principles of Metallographic Laboratory Practice, McGraw-Hill, (1943).