ENGINEERING RESEARCH INSTITUTE

UNIVERSITY OF MICHIGAN

ANN ARBOR, MICHIGAN

FINAL

NUCLEAR RESEARCH AND DEVELOPMENT

REPORT

(For the Period April 2, 1957 to June 3, 1957)

Project 2505

Chrysler Corporation

June 3, 1957

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1.0 INTRODUCTION

This report is the fifth and final progress report issued under the contract between The Chrysler Corporation and The Engineering Research Institute on nuclear energy research. This report covers the period April 2, 1957 to June 3, 1957, with prior periods being discussed in previous reports. All experimental work was stopped as of June 3 when notification was received the contract would be terminated as of June 30. The period June 3 to June 30 was utilized to complete final reports and bring property status and inventory up to date.

2.0 SUMMARY AND CONCLUSIONS

The loop was shut down after operating 840 hours under isothermal conditions at 915°F. Power failure was the cause of shutdown. An evaluation of the sample results from the loop molten metal indicates that the period of operation was too short to provide quantitative information on corrosion at these temperatures. Loop operation during this period was generally satisfactory with the exception of operational errors in sampling. The electromagnetic pump performance was disappointing in that it pumped at only half of it's rated capacity.

Revisions in the loop sampler, pump cell and instrumentation were begun to improve general performance. The sampler riser was removed and an internal baffle installed to prevent recurrance of sampler operating errors. The gap between the pump pole pieces and cell will be substantially reduced for subsequent operation. Shutdown instrumentation was changed to permit a 3 minute delay between power outage and loop dumping. This would prevent any outage shorter than this period from shuting down the loop and eliminate shutdowns from all brief power outages. The sump tank was removed from the loop and a new filter installed.

A study was started on improving the gas purification system. As a result of this study, the titanium beds in the reactors were replaced with calcium metal. This will result in considerable improvement in nitrogen and oxygen removal from the feed gases. Evaluation of gas purity tests were under way at this time.

Methods were perfected for taking radiographs of the loop sections. On the basis of these graphs, the orifice risers in the flowmeter section were opened and cleaned after slag inclusions in the melt were shown to exist. This cleaning made possible the first accurate determination of flowrates that had been made in the loop. A poor weld in front of the orifice may be contributing to the flowrate determination difficulties. No plugs or slag inclusions were found in the loop after dumping by these techniques.

Titanium carbide coatings or graphite failed during a uranium melt test. Failure was attributed to pin holes in the coating as these holes were observed during porosity tests. It was shown by equilibration-analytical techniques that TiC is stable in the presence of uranium. Future work with this coating should be directed toward forming a thicker and more impervious coating as this material has the necessary chemical and thermal stability.

A molybdenum coating on 316 stainless steel failed upon containing molten bismuth. Failure was apparently due to bismuth penetration of the coating. Future work should concentrate on making this coating thicker and with more diffusion into the base metal for greater adherence.

Revisions on the vapor phase metal coating apparatus are virtually complete.

3.0 LOOP AND LOOP ACCESSORY STATUS

3.1 Changes in Loop Proper and Its Components

The power failure on April 26, 1957 and the resultant dumping of the bismuth from the loop gave the project an opportunity to dismantle the loop and to make a thorough inspection of the loop with its component parts.

A complete set of radiographs were taken of the loop. The results are discussed in detail in the section 7 of this report.

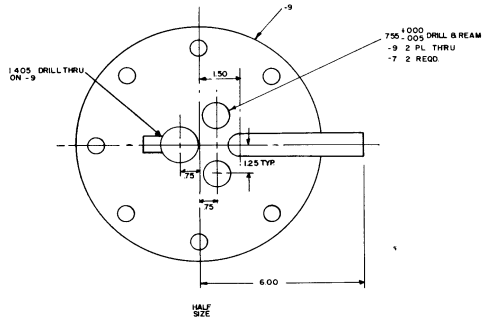
The top flange of the sampler was removed and the sampler interior inspected. Some black deposits were noted along the sampler wall. These deposits were largely removed by pickling the loop interior with a 3% HNO_3, 17% HCl solution. The black deposits were indentical in appearance to the slag removed from the orifice risers. This slag is mostly bismuth that volatilized from the melt that was mixed with small quantities of nitrides and oxides.

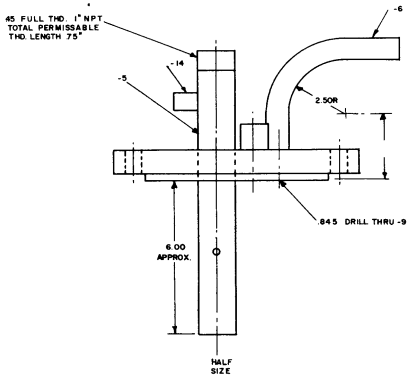
The riser pipe in the sampler was cut off above the maximum practical liquid level. This was done because the bismuth rose and solidified in the risers as results of operational errors as described in section 5 of this report.

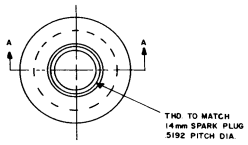
The original purpose of the riser in the sample was to insure that the samples taken were removed from the flowing bismuth stream and not from the stagnant metal inside the sampler. This was accomplished by forcing all stagnant bismuth out of the riser pipe with inert gas pressure and lowering the sample capsule into the entrance stream and permitting it to fill with the recirculating bismuth.

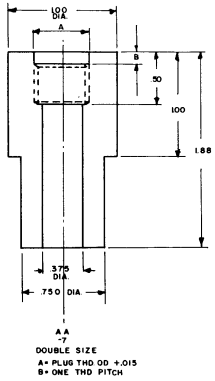
Since it was necessary to cut off the riser pipe as mentioned, some other means had to be found to obtain samples of the flowing bismuth such that the bismuth level could be kept at a reasonable distance above the inlet and outlet of the sampler. A baffle plate was installed at the inlet side of the sampler as shown in Drawing 2505-90-1004A. It was believed that this would create enough turbulence in the sampler that a representative sample could be obtained.

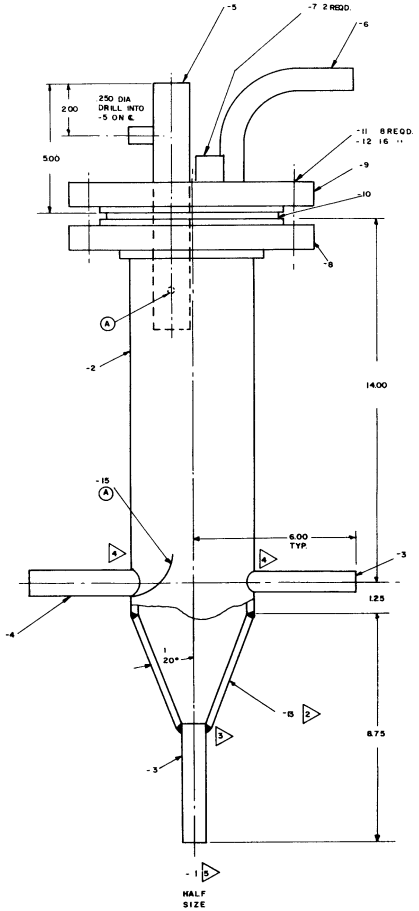
A leak was suspected in the sump tank; so this tank was removed from the system. A new dump line was installed between the dump valve on the loop and the melt tank. The line was provided with a micro-metallic filter of type 304 stainless steel with a mean pore opening of 10 microns so that the impurities in the charged bismuth would be retained on the filter. The original transfer line between the melt tank and sump tank was removed. This new system is shown schematically in Drawing 2505-92-1006A.









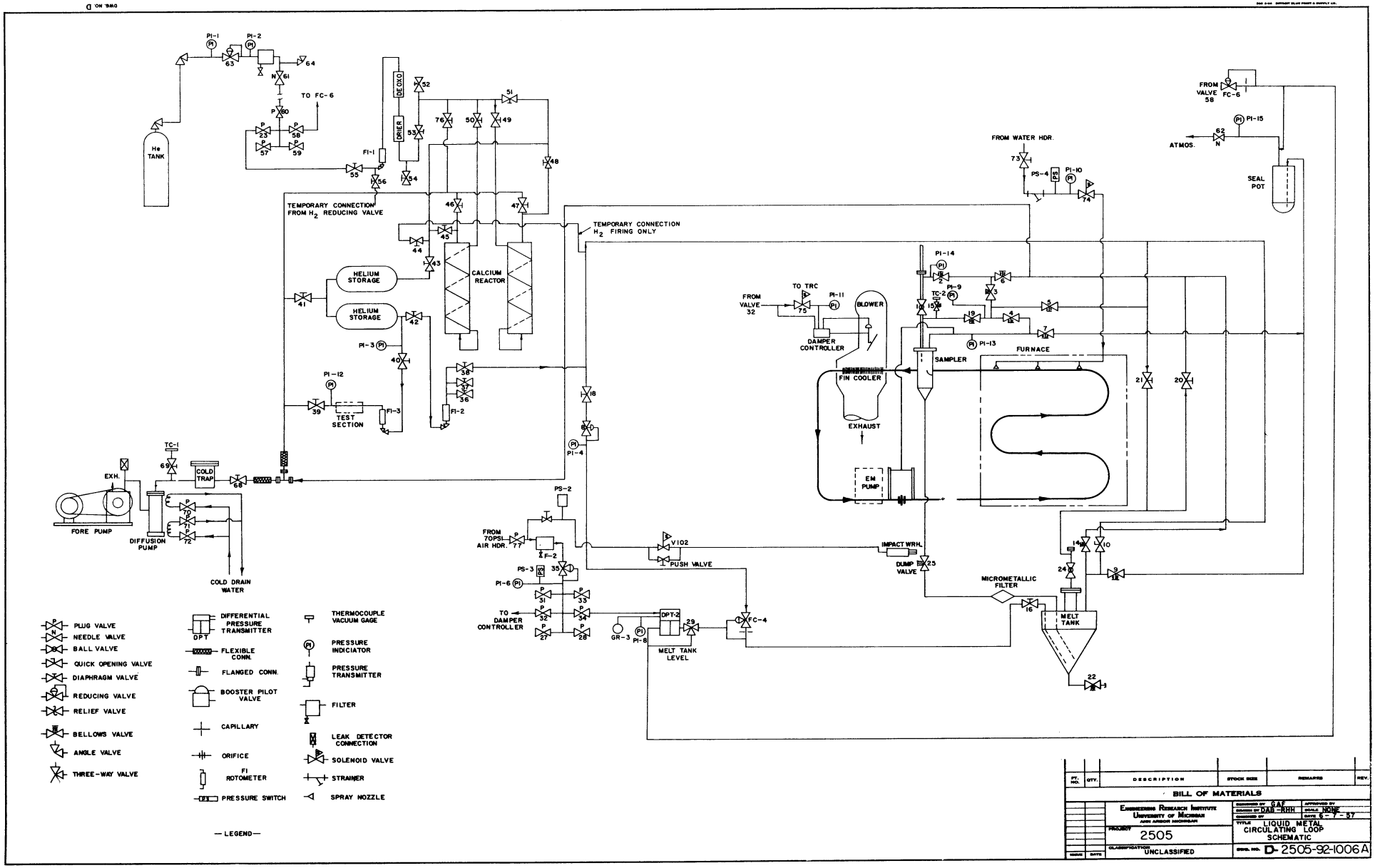


1. IT IS PERMISSABLE TO SUBSTITUTE A RAISED FACE FLANGE MACHINED TO TAKE A STD. RING JOINT
2. SWAGE OR SPIN TO SHAPE
3. NO CREVICE AT THIS JOINT
4. INLET & OUTLET PIPES TO BE GROUND FLUSH TO INTERIOR WALL
5. FABRICATION IN ACCORDANCE WITH SPEC. 2505-87-1001
6. TOLERANCE EXCEPT AS NOTED: .0X ± .05
   .00X ± .005
7. ALL WELDED JOINTS

FT. NO.	QTY.	DESCRIPTION	STOCK SIZE	REMARKS	REV.
-15	1	BAFFLE PLATE	1/4" x 4"	2 1/4 CROLOY PIPE	A
-14	1	HALF CPLG.	1/4" 3000#	304 SS	
2 -13	1	PIPE	4" x 5"	2 1/4 CROLOY SCH. 80	
-12	16	HEX NUT	1/2-13	ASTM A194-4B CLASS 0	
-11	8	STUD	1/2-13 x 4"	ASTM A193-4BT 0D. BC	
-10	1	RING	5 7/8 x 5/16	304 SS	
1 -9	1	RING JOINT BLIND-FLANGE	4"-150#	304 SS	
1 -8	1	RING JOINT SLIP-ON FLANGE	4"-150#	304 SS	
-7	2	FITTING	1" x 1"7/8	304 SS	
-6	1	PIPE	1/2" x 7"3/4	304 SS SCH. 40	
-5	1	PIPE	1" x 19"1/4	2 1/4 CROLOY SCH. 80	
-4	1	TUBE	1" x 4"	2 1/4 CROLOY 14 BWG	
-3	2	PIPE	1/2" x 4"	2 1/4 CROLOY SCH. 80	
-2	1	PIPE	4" x 5"	2 1/4 CROLOY SCH. 80	
3 -1	1	SAMPLER			

BILL OF MATERIALS

ENGINEERING RESEARCH INSTITUTE UNIVERSITY OF MICHIGAN ANN ARBOR MICHIGAN	DESIGNED BY GAF	APPROVED BY [illegible]	
	DRAWN BY RIK	SCALE NOTED	
	CHECKED BY GAF	DATE 3/16/56	
PROJECT 2505	TITLE REVISED SAMPLER		
CLASSIFICATION UNCLASSIFIED	DWG. NO. D-2505-90-1004A		
ISSUE / DATE			



FT. NO.	QTY.	DESCRIPTION	STOCK SIZE	REMARKS	REV.

BILL OF MATERIALS

Engineering Research Institute, University of Michigan, Ann Arbor, Michigan	Designed by GAF; Approved by
	Drawn by DAR-RHH; Scale NONE
	Checked by; Date 6-7-57
Project 2505	Title LIQUID METAL CIRCULATING LOOP SCHEMATIC
Classification UNCLASSIFIED	Engr. No. D-2505-92-1006A

4.0 LOOP INSTRUMENTS AND CONTROLS SECTION

4.1 Modification of Loop Instruments and Controls System

Several changes in the loop control system were required as a result of the loop modifications undertaken following the dumping of the loop after the power failure on April 26, 1957.

a) Removal of all equipment associated with the sump tank liquid level measurement.
b) Installation of a delayed automatic dump system.
c) Installation of a heater on the melt tank liquid level dip leg.
d) Revision of thermocouples associated with the dump line and melt tank dip legs.
e) Rearrangement of the electrical hookup of the heaters on the dump line, flowmeter, and melt tank dip leg.
f) Revision of cell, thermal and electrical insulation on the EM pump.

4.2 Removal of Sump Tank

The sump tank and its associated equipment was removed from the system. Removal of the sump tank from the loop system necessitated removal of the liquid level measuring equipment from the loop. The thermocouples previously assigned to this tank were re-assigned as listed elsewhere while the heater control system was shifted to the dump line as described in another section.

4.3 Installation of Delayed Automatic Dump System

A tabulation of the cause, frequency and duration of power failures caused by failure of Detroit Edison equipment was obtained from The Detroit Edison Company. This showed that only about one failure per year lasted longer than 30 seconds. Based upon this information, the decision was reached to install a 2-3 minute delay between a power failure and the initiation of the automatic dump valve.

In the original loop control system, all of the power contactor holding circuits released on a power failure and would not re-energize unless the entire startup procedure were carried out. It was, therefore, necessary to install auxilliary contacts actuated by the battery source in parallel with the original holding contacts. These auxilliary contacts allow the loop to restart itself in the event the power returns before the loop is dumped. The contacts return to their normal open position when the loop is operating normally or after the loop is dumped.

4.4 Installation of a Heater on Melt Tank Liquid Level Dip Leg

Considerable difficulty was encountered in keeping the dip leg from plugging whenever the melt tank was pressurized for transferring bismuth. The dip leg was altered to permit it to drain to the tank and a heater and thermocouple added to prevent freezing of bismuth in the line.

4.5 Revision of Thermocouples

All of the thermocouples associated with the sump tank were removed and relocated on the revised dump line and melt tank dip leg.

Thermocouples were installed on the dump line as follows:

Dump Valve	2 Couples
Filter	2 Couples
Dump Line 8" above Melt Tank	2 Couples

Two thermocouples were installed on the dip leg 10 inches above the melt tank. Drawing Number 2505-94-1021R shows the location of all thermocouples on the revised loop.

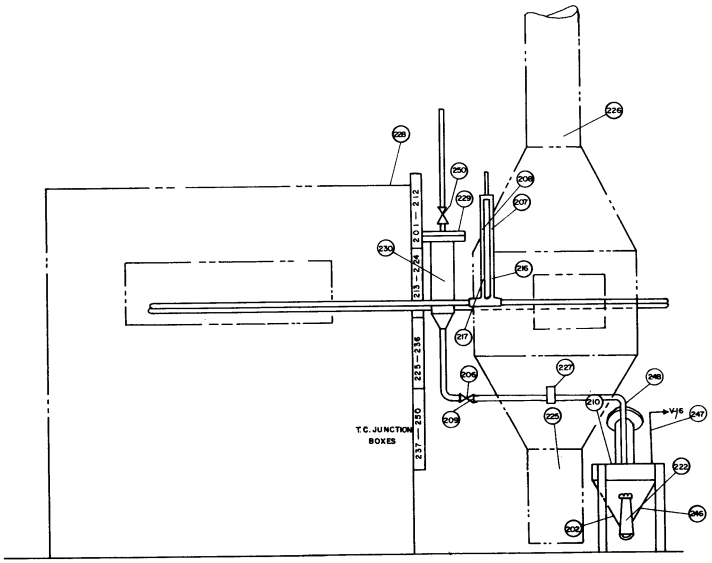
4.6 Rearrangement of Heater Circuits

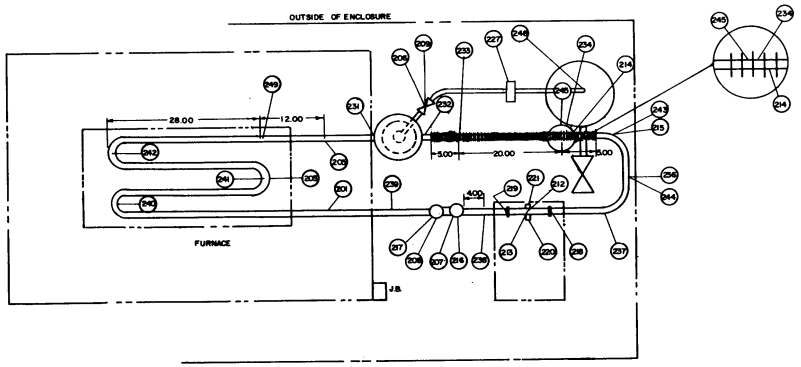
The removal of the sump tank made its heater controls available for other service. The heater on the dump line previously associated with the flowmeter risers and transfer line between sump and melt tanks was, therefore, connected to the old sump tank heater controller. The flowmeter risers and melt tank dip leg are connected in series on the transfer line circuit.

These changes should give better control of this auxilliary piping than the previous arrangement of having all lines controlled by a single controller.

4.7 Revision of EM Pump Cell Thermal and Electrical Insulation

It was decided to substitute a sheet of 0.030 mica for the wire mesh thermal insulation between the transformer pole pieces and pump cell. This will also eliminate any possibility of shunting of the cell current by the pole pieces. The thermocouples indicating pump cell temperature will still be located between the insulation and the cell. This will narrow the air gap between the pump pole pieces and cell appreciably and should result in better pump efficiencies.





PT. NO.	QTY.	DESCRIPTION	STOCK SIZE	REMARKS	REV.

BILL OF MATERIALS

ENGINEERING RESEARCH INSTITUTE, UNIVERSITY OF MICHIGAN, ANN ARBOR, MICHIGAN	DRAWN BY C.H. / CHECKED BY G.A.F. / APPROVED BY / SCALE FULL / DATE 5/24/57
PROJECT 2505	TITLE THERMOCOUPLE LOCATIONS
CLASSIFICATION UNCLASSIFIED	DWG. NO. D-2505-94-1021R

ISSUE DATE

5.0 LOOP OPERATION

5.1 Isothermal Operation

The EM pump began pumping as soon as the loop was filled with bismuth, and the loop was operated isothermally and continuously for 834.4 hours, at a temperature of 915 Fahrenheit.

The power input to the EM pump was varied during the operation. The voltage was kept at 90 volts for the first 70 hours, 105 volts for the next 266 hours, 125 volts for the following 240 hours and 105 volts for the rest of the time until shut-down. The current input was kept at 2 amps, 7.5 amps, 11 amps and 7.5 amps, respectively at the above voltages.

An attempt was made to calibrate the voltage setting of the EM pump versus flowrate of bismuth in the loop. No representative results were obtained (see section 5.2) except at a voltage setting of 200 volts. At 200 volts a pressure drop of 0.45 inches of bismuth was detected by means of radiography across the flowmeter risers as shown in Figure (2). Calculations of the theoretical flowrate at a temperature of 950 Fahrenheit through the orifice as a function of pressure drop of the bismuth in the flowmeter as shown in Figure (1) gives a corresponding flowrate of 1.4 fps., based upon the $\frac{1}{2}$ in. sch. 80 pipe which is the size of the loop cold leg.

Shut-down of the loop was caused by a power failure on April 26, 1957 at 7:54 p.m., which activated the automatic dump system and emptied the loop. It was then decided to discontinue operations until some corrections on the experimental system and evaluation of loop operation could be made. The corrections in the system are discussed in detail in this report under the section entitled "Loop and Loop Accessory Status".

5.2 The Difficulties encountered during Operation

During the early operation of the loop, some difficulties were encountered with helium being trapped in the loop proper, resulting in irregular flow. This gas was removed by evacuating the loop under operating conditions to a less than 30 microns of Hg. and repressurizing to operating pressures with helium.

The two liquid level measuring probes in the sampler shorted out due to splashing bismuth that froze between the probes and the top flange of the sampler. The probes were removed and cleaned. After reinstallation, one probe was placed right at the present liquid level while the other was kept withdrawn as much as possible and only used when accurate liquid level measurements were required. This procedure always holds one probe in reserve.

FIGURE - 1
COMPUTED ORIFICE CALIBRATION
BISMUTH CIRCULATING LOOP
D=0.834, d=0.625, FLANGE TAPS, $Q = 1.577\sqrt{H}$

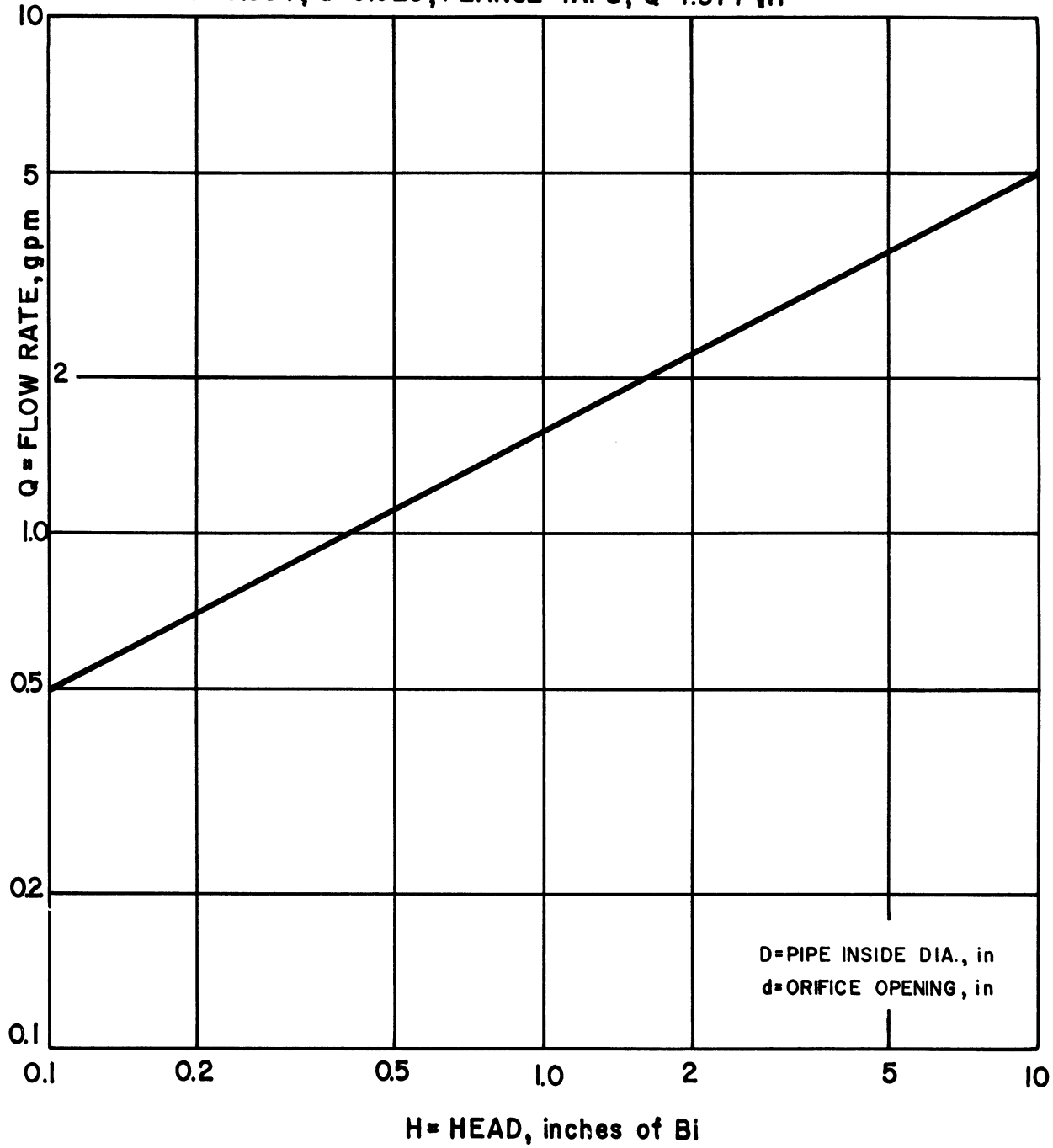


FIGURE 2.

RADIOGRAPH OF ORIFICE RISERS AFTER CLEANING

A low rate of helium purge will be used during the future operations to minimize the amount of splashed bismuth during transfer from melt tank to loop.

It was noted after some time of operation that the ball valve between the sampler and the stand pipe was leaking gas. The reason for this being the effect of the heat on the valve seat and the gas pressure below the valve. Since it is a spring loaded valve, any pressure internally acts against the spring so outward leakage is not surprising. The valves seat, ball and spring were replaced once during the operation of the loop. The leak in the valve is not harmful since the gas escaped into the sampler tube which was sealed off from the atmosphere by an O-ring seal. This O-ring seal was found to leak after it had been in use for a short time. The combination of heat and pressure exerted upon the seal created flatspots on the O-ring and thereby caused leakage. It was, therefore, made standard procedure to replace the old O-ring with a new one each time a sample was taken. To avoid loss of the purified helium blanket above the bismuth in the loop, the blanket pressure was raised from normal running conditions of 3 in. of Hg. (gage) to 20 psig. each time the loop was left unattended for a long period of time such as the weekends.

As mentioned earlier, an attempt was made to calibrate the voltages setting of the EM pump versus flowrate of the bismuth in the loop. Radiographs which were taken of the flowmeter risers, showed that some slag had collected in the riser pipes, and the liquid level could not be determined with any accuracy. The tops off the riser pipes were, therefore, cut off and the slag was removed with an auger, while under a continuous purge of an inert gas. When the slag was removed, the calibrations were repeated using a conductivity probe for measuring the liquid level in the risers. This was also done under a purge with an inert gas.

The risers were then welded close with extensions on each leg so that the ends would be above the insulation and thereby permit easier removal of slag in the future.

Several times during the sampling operations, the bismuth rose in the riser pipe in the sampler and froze there. This rise of bismuth was due to operational mistakes in manipulating the controls. Nothing to prevent this could be done except to instruct the personnel in proper sampling procedure. After the shutdown of the loop, the riser was cut off as described in the section of this report entitled "Loop and Loop Accessory Status".

5.3 Results

Samples of the bismuth were taken at frequent intervals during loop operations. The major part of these samples and samples of the slag removed from the flowmeter risers were submitted for analysis to Dr. P. J. Elving of the Department of Chemistry at The University of Michigan. Some samples were also submitted to The Chrysler Corporation. Analytical results of the loop samples are given in Table I of this report.

Table I

Analytical Results of Loop and Slag Samples

Sample No.	Elapsed Operating Time Hours	Results Zr (p.p.m.)	Mg (p.p.m.)	Fe (p.p.m.)	Cr (p.p.m.)	Analyst
1-LUA	138.7	158 ± 6	279 ± 22	46 ± 2		Elving
1-LUB	138.7	164 ± 6	284 ± 2	17 ± 1		Elving
2-LU	264.2	261 ± 3	322 ± 12	31 ± 1		Elving
3-LU	283.7	325	325	60	10	Chrysler
4-LU	287.1	261 ± 7	340 ± 39	33 ± 1		Elving
5-LU	449.5	248 ± 7	308 ± 10	16 ± 1		Elving
6-LU	571.6	258 ± 9	321 ± 14	18 ± 1		Elving
7-LU	812.5	237 ± 20	276 ± 14	26 ± 1		Elving
*Special 1	262.5	1,200	3,000	1,000	150	Elving
Special 2	262.5	20,000	600	600	80	Elving
Special 3	744.2	1,200	600	600	1,000	Elving

*Special samples #1 and #2 are samples of slag which adhered to the zirconium basket during Zr loading operation. Special sample #3 is a sample of slag removed from the flowmeter risers. Sample #3 was also submitted to Chrysler. No results available as yet.

An examination of Table I shows that the concentration of the additives (zirconium and magnesium) in the two first samples is fairly consistent. The concentrations were lower than the design values, originally obtained from the Brookhaven National Laboratory, which were: 250 ppm. for Zr and 350 ppm. for Mg. An additional amount of both Zr and Mg was, therefore, added through the sampler. According to Table I, the concentration of the additives, after the second addition, were close to the design value and stayed fairly constant, both for Zr and Mg until the last sample. The Chrysler analysis due to different analytical procedures are not consistent with the University determined values and were not included in the following discussion. In lieu of the spectral analysis and the small amounts of Zr and Mg involved, there is an indication that the concentration of the additives stabilized rapidly. The running time between samples 1-LUB and 3-LU, approximately 145 hours, is indicative of the operating time required to reach the additive stabilization point.

The analytical results show the concentration of Fe in the Bi to be 16-46 ppm. The calculated solubility at 915°F of Fe in Bi from Brookhaven National Laboratory data is 15 ppm. The highest Fe analysis was from the first loop sample. Subsequent samples, omitting the Chrysler analysis, showed much lower Fe content than the saturation value. This is presumably due to occluded Fe in the molten bismuth.

The slag samples showed a much higher Fe concentration than the regular samples. Since these had to be chiselled out, and therefore, may be contaminated by chips from the tool used or from the pipe wall, no importance is attached to the analytical results from these samples.

On future runs, it was planned to take filtered and non-filtered loop samples. Comparison of analytical results from these two samples would indicate the respective quantities of occluded and dissolved Fe in the melt. Varition of occluded Fe concentration would indicate the quantity of Fe "floating out" in parts of the loop such as the orifice risers and the sampler. This would serve to give quantitive information on corrosion rates.

From the data available to date, no firm conclusion can be reached regarding mass transport. It is very probable that occluded Fe was being collected in the orifice risers. However, the rate of corrosion or erosion could not be determined from the run data on so short a run. A long term run correlating Fe buildup versus running time would be required to set any reliable corrosion or mass transport rates.

6.0 PURIFICATION OF THE INERT GASES AND HYDROGEN

6.1 Objectives

6.1.1 For the purpose of supplying the loop with an inert gas blanket of very high purity, special purification equipment based upon published data (10,11)* for high temperature gas-metal reactions was designed and built (2)*. Originally intended for the ultimate purification of welding grade helium and/or argon, its use was extended to the purification of hydrogen for the materials program and for the removal of oxides from the interior surfaces of the loop during hydrogen firing.

6.1.2 Since the welding grade of helium used as the raw gas in this work is supplied in a fairly high order of purity (approximately 20-50 ppm), the means for detecting the amount of residual impurities, which is not to exceed 3 ppm, must be quite sensitive. Following the experiences gained in the performance of similar work at Brookhaven National Laboratories (12)*, a highly polished surface on a small block of reactor-grade uranium metal (Mallinckrodt) was chosen as the purity indicator. Conducted at temperatures in the vicinity of 1000°F, this test, though empirical, was expected to provide the necessary sensitivity to detect the ten to one reduction in impurity concentration regarded as the minimum ratio. The purity test would thus serve not only as a criterion of gas usability but as an indirect indication of the leak-tight condition of the equipment as well. No corresponding purity test was considered necessary for the hydrogen.

6.2 Loop Supply Purification Train

6.2.1 This equipment was originally designed to provide duplicate high-temperature reaction furnaces, each with a rated capacity of 20 SCFH of helium based upon a gross impurity content of 200 ppm and a bed life of 3600 hours. Each furnace carried a charge of 16 pounds of titanium sponge metal with a measured porosity of 73% (voids/total). The design temperature of the bed was 1450°F but the instrumentation was insufficient to establish if the entire bed was maintained at this temperature. Temperatures were falling or approximately 200°F at each end of the bed. However, at the gas flowrates used and in consideration of the liberal safety factor designed into the reactor, it is reasonable to assume that a rate constant for the gas-titanium reaction somewhat lower than that taken as the design value (2)* would not pose any problem in the overall operation. Several batches of helium gas were purified but the purity tests performed on each batch were inconclusive, as will be described in Section 1.3.

*All references will be found in the appendix.

6.2.2 A need for hydrogen gas of purity higher than that commercially obtainable arose from the initial hydrogen firing tests (6) preparatory to the hydrogen reduction of the oxide films associated with the interior loop surfaces. A further need for a high purity hydrogen had previously developed in the plans for the reduction of the metal chlorides used in the vapor plating processes simultaneously being investigated in the materials program (5). These two needs necessitated the commitment of one of the titanium furnaces to the exclusive purification of hydrogen. The hydrogen firing tests indicated that Croloy-2-1/4 and bismuth tests specimens became badly tarnished when allowed to cool in an atmosphere of raw hydrogen, but remained clean in an atmosphere of purified hydrogen. This effect is probably due to the effective removal of trace quantities of water vapor in the titanium bed which passed through the silica gel drier. However, the passage of hydrogen into the heated titanium sponge presented a new problem. At the temperature of the reaction bed (1450°F), the equilibrium content of hydrogen in the form of titanium hydride is approximately 38 SCF for the 16 pound titanium charge. This equilibrium shifts to a hydride concentration corresponding to approximately 100 SCF of hydrogen as the bed approaches ambient temperature. Because of the extreme pyrophoric nature of the hydride, extra care must be observed in the handling of the bed and, in fact, a prolonged nitrogen purge at high temperature was utilized to remove as much hydrogen as possible prior to opening the reactor to atmosphere. In bringing this reactor up to operating temperature after previous runs, it is essential that the initial absolute pressure be low. This is to avoid overpressurizing the system as a result of the high dissociation pressures which are attained at the operating temperatures.

6.3 Evaluation of Gas Purity Analysis Technique

6.3.1 Various techniques have been proposed for the measurement of very low concentrations of contaminating gases in the inert gases (8,10,12). Following the experience of Brookhaven National Laboratory (12), a highly polished surface on a uranium specimen was chosen for this work. A test section of approximately 10 in.^{3} total volume consisted of a Corning No. 8560 glass tube with a 2.5 mm wall thickness and 15 mm inside diameter, suitably valved to the purified gas supply. Provisions were made for the evacuation of the test volume and for heating the uranium specimen to 1000°F. As the initial heat-up of the polished test specimen progressed, under a pressure of less than 5 microns measured with a thermocouple type gage, a severe surface tarnish appeared at 850°F. As the temperature continued to rise, this color ranged from bronze through orange and blue-

black to a light blue. Upon prolonged heating under vacuum, the light blue tint seemed to disappear but upon viewing it directly, after cooling and removal from the test section, a residual tarnish was found to be present. Repeated tests of similar nature produced identical results. Variations in the procedure such as purging with helium during evacuation and heating under helium did not alter the tarnish effect. In view of the possibility of air leakage in the vicinity of the test specimen, a more thorough investigation of the precise vacua being obtained was made. An ionization type gage was mounted in the line between the vacuum diffusion pump and the test section. A reading of 0.1 micron was obtained before the heater power was turned on. As the heat-up progressed, the pressure decreased to 0.09 micron as the tarnish began to appear at 900°F. The usual range of colors appeared as the temperature increased to 1000°F. A rough leak check was made at this point by closing the vacuum valve for two minutes. During this time, the pressure reading (on the pump side of the valve) dropped to 0.08 micron. Upon opening the valve, the pressure exceeded 0.1 micron for five seconds then returned to 0.09 micron. No calculations were made to prove or disprove that this might be indicative of a significant leak, in consideration of pumping speed of the vacuum equipment, or that it might be a matter of continued outgassing of the unheated parts of the system. Continued heating of the specimen resulted in the interesting effect that the color cycle repeated itself about every 15 minutes. One test was performed after evacuating the test section for 45 minutes with the result that no tarnish appeared at any temperature.

6.3.2 In order to investigate the nature of the tarnish phenomenon under rigidly known vacuum conditions, a second heated test section was assembled. This apparatus consisted of a heated and insulated pyrex glass tube which was evacuated through connecting tubing of 1/32 unit wall tygon tubing. After the gross leaks were located and eliminated, it was found that the required vacua still could not be obtained at a measuring point in the vicinity of the located section. Using an ionization gage, pressures of less than 0.1 micron were obtainable at the vacuum diffusion pump, but were as high as 25 microns at a distance of approximately 30 inches away. Thorough testing with a helium leak detector led to conclusions that the tygon laboratory tubing was at fault in this system, due to one or both of the following causes:

(1) outgassing of the tygon material,
(2) diffusion of air through the tubing walls. The large differences in vacuum between different parts of the system cast doubt on the validity of the vacuum readings taken during the helium purity testing.

6.3.3 A new system is currently being designed to permit quantitative evaluation of this general method for detecting

trace quantities of the contaminating gases. This system should

(1) minimize outgassing difficulties,
(2) minimize potential leakage sources,
(3) provide for more accurate measurement of specimen temperature,
(4) provide improved optical system for more sensitive visual detection of surface changes,
(5) provide for admitting accurately known gas samples of very low pressure,
(6) facilitate leak detection,
(7) provide for movement of specimen within evacuated test chamber through sliding and rotating vacuum seals.

6.3.4 The surface color phenomenon seems explainable in an obvious way from the outgassing and/or leakage standpoint. Yet it is not definite that such concentrations as indicated by measurement would be great enough to account for the observed effects. For example, a partial pressure of one micron of air (10^{-6} meter) in helium at a total pressure of one meter (1.31 atmosphere) would be a concentration of one part per million by volume. If the system pressure were maintained at less than 0.1 micron at test temperature, it seems unlikely that this very low concentration would be significant. This assumes that the vacuum measurements are valid. The color cycling phenomenon reported above might suggest that something else is in evidence, such as the continued release of occluded gas. The latter theory may find substantiation in the report (Ref.) that various uranium melts in thoroughly outgassed inert crucibles under high vacua showed consistent tendency to form a layer of slag. This suggests the possibility of specially purified uranium test specimens being necessary to the successful performance of this technique. Smithells (7) lists three distinct gas-metal reactions:

(1) chemisorption, which leads to the formation of a strongly held monolayer,
(2) physically absorbed layer of gas as a result of Vander Waal forces,
(3) gas-metal compound formation accomplished by a decrease in free-energy.

Further work should identify the conditions under which any of these mechanisms are possible for the system under discussion. In addition, the solubilities of oxygen and nitrogen in uranium are well-known. The observed phenomena may well be manifestations of transitions involving any or all of the above processes.

7.0 RADIOGRAPHY

The 1.2 curie cobalt 60 source has been used to take approximately 100 radiographs thus far. The first were test exposures of a mockup of the flowmeter risers to determine proper exposure times. A large number of exposures were made of the actual flowmeter and the entire loop was radiographed after the bismuth was dumped. Several radiographs were also made of some welded vessels fabricated in our shop.

Several important facts were discovered by means of radiography. The first pictures of the flowmeter showed that the risers were at least partially plugged with slag both above and below the bismuth surface (Fig. 3). The tops were, therefore, cut off the risers and the slag removed mechanically. A radiograph of the cleaned risers with a pump setting of 200V showed a pressure drop of 0.45 inches of bismuth across the orifice (Fig. 2), and this corresponds to a flowrate of 1.4 ft./sec. The pump, according to the General Electric Company data sheets, should give a flowrate of 4 ft./sec. at 200V. Fig. 4 shows a possible reason for this apparent discrepancy. The radiograph of the orifice assembly shows that the weld upstream of the orifice which is only 2 inches from the upstream riser has severe penetration. This weld forms a second orifice and lowers the pressure at the upstream riser which causes a low reading across the flowmeter. Another possibility is that the pump is not performing at its specified rate.

The radiographs of the emptied loop show that two of the welds on the loop have slag inclusions. Nearly all of the welds show excessive penetration.

Two lead casks were used in this work. The original cask weighing 670 pounds proved to be too bulky for radiographing certain sections of the loop during operation. A light, 25 pound cask was built to replace the bulky cask. This cask fitted into a heavier storage cask when not in use. An exposure factor of 50 without insulation and 60 with insulation proved quite satisfactory for radiographing the sections of the empty loop with Kodak Type AA film and two .005 in. lead screens in front and behind the film. The exposure factor (F) is defined by the following equation:

$$F = \frac{(\text{Exposure time, min.})\ (\text{Milli-curies of source})}{(\text{Source-to-film distance, in.})^2}$$

FIGURE 3.
RADIOGRAPH OF ORIFICE
RISERS BEFORE CLEANING

FIGURE 4

RADIOGRAPH OF ORIFICE SECTION

8.0 MATERIALS PROGRAM

The approach to the materials problem for molten fuel reactor application has been given the latter three progress reports. This final quarterly report contains a continuation to the previous periodic report (October 2, 1956 - April 2, 1957).

In this report there are two phases of investigation carried on; namely:

1) Titanium carbide coated graphite.
2) Molybdenum coated stainless steel.

Evaluation of both coatings have been completed. The following sections will describe the experimental work and results.

8.1 Titanium Carbide Coating

The last report described the testing of this coating with molten uranium at 2300°F. Since the coating proved to be permeable, the work since then has been directed toward determining the reason for this defect.

An investigation of the solubility of titanium carbide in molten uranium was conducted. Before this could be done, however, a sample of titanium carbide had to be prepared by direct combination of titanium and carbon. After purification, this sample was analyzed and found to be 99.7% titanium carbide.

Solubility determinations were made by floating a 3 gram sample of titanium carbide on top of molten uranium at 2400°F for 4 hours. After cooling the melt, a small fraction of the parent uranium was removed for analysis. The analysis showed a maximum titanium content of 0.02%.

Assuming that equilibrium had been attained in 4 hours, one can say that titanium carbide is practically insoluble in uranium at these temperatures. Thermodynamic calculations serve as a rough check of this insolubility. However, data on the free energies of formation on TiC from two sources (7), (13) are inconsistent, so the data of Quill (13) must be used while that from Smithells (7) would appear to be incorrect if the analytical data are to be believed.

In conjunction with these solubility experiments, a method for making calcium oxide crucibles was developed. This consisted of heating chemical pure calcium oxide at 1200°F for one hour to drive off any water of hydration. The dried powder was packed into a graphite crucible with a tapered mandrel. After removing the mandrel, the crucible was fired for two hours at 3000°F. This

resulted in a rather dense and non-porous crucible; which will contain uranium metal at 2400°F and not volatilize at pressures below one micron.

Three titanium carbide coated crucibles were measured for porosity by gas diffusion. Two of these appeared to be no tighter than uncoated graphite due to small flaws in the coating which resulted in localized leaks. The third crucible, however, proved to be 18 times tighter than the other two.

From this work, one can say that titanium carbide is a satisfactory coating material provided that it be made uniform and of sufficient thickness.

Two runs were made in the large resistance furnace using a feed of $TiCl_4$ and C_3H_8. These runs were made at crucible temperatures of 3200 - 3400°F. Large crystals of TiC formed in the feed tube and on the sides of the crucible. Leaks in the system permitted some of the feed to escape outside the crucible. Sighting through these vapors gave erroneous temperature readings and consequently, the temperatures were too high for effective coating resulting in the growth of large crystals in the feed tube and crucible. Temperatures of 2900°F would avoid this difficulty.

8.2 Molybdenum Coating

The molybdenum coating on a 316 base stainless steel was thermal cycled fifteen times from 400°F to 1700°F. A light blue discoloration on a small part of the surface resulted, but the coating did not dust or flake off. Next, the coating was subjected to molten bismuth at 1600°F for 26.5 hours. The coating was removed after approximately one hour. The removed molybdenum floated on top of the bismuth in small flakes. The parent stainless had been wetted by the melt.

The conclusions are that molybdenum is satisfactory as a coating provided it can be built up to a thickness of approximately 0.020 in. and that life might be improved by aging the coating at 2400°F for several hours to allow the molybdenum to diffuse into the parent material. Future runs had been outlined earlier to test this hypothesis.

8.3 Metallic Materials - Induction Furnace

The rebuilt vapor plating furnace described in the quarterly report dated April 2, 1957, has been completed and operational checks of the various parts of the furnace begun.

The furnace and associated tubing is sufficiently vacuum tight for plating operations, however, when the high frequency power was turned on, considerable arcing between the radiation shield and top flange resulted. Shortening of the ceramic tubing supports of the radiation shield by 2 inches should remedy this difficulty.

Calibration of the hydrogen and chlorine rotameters was begun.

9.0 MANPOWER EXPENDITURES

Time spent on the project for the two areas of activity is tabulated below. An eight hour working day is assumed for each working day.

	Work Days
Loop	128.6
Materials Program	25.0
Total	153.6

10.0 EQUIPMENT AND MATERIAL STATUS

Status of the equipment and outstanding purchase requisitions are given in Table II. This table covers the period of April 2, 1957 to June 3, 1957. Previous periods were covered in other reports.

TABLE II

MATERIAL STATUS REPORT

ORDER NO.	QUANTITY	DESCRIPTION	MANUFACTURER		REQUISIT. TO CHRYSLER	CHRYSLER APPROVAL RECEIVED	REQUISIT. TO PURCH'G	ORDER TO VENDOR	PROMISED DELIVERY	REMARKS		COSTS
1	18	Film Holders, Lead Alloy Screens	Picker X-ray Corporation	(C.H.)	**		4/4	4/12	Stock	*4/22	224887	$24.00
2	4	Hoke Valves	H. E. Lennon and Company	(J.P.)	**		4/4	4/4	Stock	*4/29	223714	$35.60
3	1	Micro Switch	Wedemeyer Electronic Supply Company	(C.H.)	**		4/4	4/4	Stock	*4/4	L-5253	$7.65
4	1	Repair of IBM Typewriter	IBM	(R.D.)	**		4/5	4/5	Stock	*4/5	223723	$6.14
5	6	Silicone O-Rings	Detroit Silicone Rubber Company	(R.K.)	**		4/9	4/9	Stock	*4/18	223739	$3.30
6	1 10'	All Iron Gate Valve Hard Copper Water Tube	Taylor Supply Company	(D.T.)	**		4/9	4/16	Stock	*5/1	225663	$9.64
7	25	Couplings	Taylor Supply Company	(C.D.)	**		4/12	4/17	Stock	*4/29	225946	$5.50
8	4	Adapter for Air Line Coupler	Chas. A. Strelinger Company	(C.D.)	**		4/12	4/17	Stock	*4/25	225945	$1.00
9	25	O-Rings	Crane Packing Company	(C.D.)	**		4/12	4/17	Stock	*4/29	225944	$5.00
10	3	High Speed Steel and Socket Wrench	Royall Inc.	(C.D.)	**		4/12	4/17	Stock	*4/22	225943	$2.35
11	34	Radio Activity Tags and Signs	Atomic Instrument Company	(C.H.)	**		4/15	4/18	Stock	*5/6	226265	$2.00
12	6	Tubes	Wedemeyer Electronic Supply Company	(C.H.)	**		4/16	4/22	Stock	*4/24	226467	$6.90
13	1 qt.	Paint	A. R. Congdon and Sons, Inc.	(C.H.)	**		4/17	4/17	Stock	*4/17	226001	$2.15
14	150	1¢, 3¢, 10¢, 20¢ Postage Stamps	University of Michigan	(R.D.)	**		4/17	4/20	Stock	*4/22		$7.70
15	3	Casters	Chas. A. Strelinger Company	(C.H.)	**		4/18	4/17	Stock	*4/22	226009	$3.00
16	150 6	Sheets of X-ray Film Film Holders and Lead Alloy Screens	Picker X-ray Corporation	(C.H.)	**		4/18	4/22	Stock	*4/29	226456	$40.00
17	1	Wattmeter	Purchase Radio Supply	(C.H.)	**		4/18	4/22	Cancelled		226457	$30.00
18	1	Current Transformer	Wedemeyer Electronic Supply Company	(C.H.)	**		4/18	4/22	Cancelled		226458	$9.00
19	20	Drills	General Stores	(R.K.)	**		4/19	4/27	Stock	*5/13	253371	$3.02
20	1	5 lb. - Alundum	Eberbach and Son Company	(L.Y.)	**		4/19	4/19	4/19	*4/19	L-5324	$3.25
21	1	Repair of IBM Typewriter	IBM Company	(R.D.)	**		4/22	4/18	4/22	*4/22	226036	$6.00
22	24	Hacksaw Blades	Royall Inc.	(C.D.)	**		4/24	5/1	Stock	*5/13	228118	$14.64
23	25	Swagelok Fittings	H. E. Lennon and Company	(C.D.)	**		4/24	5/1	Stock	*5/7	228119	$10.00

*Item received.
**Expendable item, Chrysler approval not required.

TABLE II (CONT.)

MATERIAL STATUS REPORT

ORDER NO.	QUANTITY	DESCRIPTION	MANUFACTURER	REQUISIT. TO CHRYSLER	CHRYSLER APPROVAL RECEIVED	REQUISIT. TO PURCH'G	ORDER TO VENDOR	PROMISED DELIVERY	REMARKS	COSTS
24	54 --	Terminals Rotary Switch and Brass Inserts	Wedemeyer Electronic Supply Company (C.H.)	**		4/25	5/2	Stock	*6/5 228277	$4.90
25	4	Notebooks	Mayer-Schairer Company (C.H.)	**		4/25	5/2	Stock	*5/27 228278	$13.00
26	12'	Copper Tubing	Steel Sales (H.M.)	**		4/26	4/26	Stock	*5/3 226092	$2.50
27	6	Rolls of Drip Tape	Buhl Sons Inc. (C.H.)	**		4/26	4/30	Cancelled	227807	$10.00
28	40'	Vacuum Tubing	F. B. Wright Company (W.S.)	**		4/29	4/29	5/13	*5/13 226096	$93.20
29	1	Galvanized Steel Stove Pipe	Plant Stores (Univ. of Michigan) (H.M.)	**		4/29	4/29	Cancelled		$1.50
30	2	Sealed Pyrex Brand "Double Tough" Pipes	Fisher Scientific Company (C.H.)	**		4/30	4/30	6/15	Cancelled 227804	$70.00
31	2	Pipe and Pipe Cap	Fisher Scientific Company (C.H.)	**		4/30	5/6	Stock	*5/10 227814	$35.60
32	40#	6-Mesh Nodules of Commercial Grade Calcium	Nelco Metals Inc. (L.C.)	**		4/30	4/30	Stock	*5/8 227808	$100.00

*Item received.
**Expendable item, Chrysler approval not required.

TABLE II (CONT.)

MATERIAL STATUS REPORT

ORDER NO.	QUANTITY	DESCRIPTION	MANUFACTURER	REQUISIT. TO CHRYSLER	CHRYSLER APPROVAL RECEIVED	REQUISIT. TO PURCH'G	ORDER TO VENDOR	PROMISED DELIVERY	REMARKS		COSTS
1	12	Mica Sheets	Tarheel Mica Company (C.H.)	**		5/6	5/13	Stock	*5/20	229777	$5.00
2	4	Dozen - Saber Saw Blades	Sears, Roebuck and Company (C.D.)	**		5/6	5/10	Stock	*5/27	229444	$3.68
3	30 5#	Troy Ounces of Silver Solder Silver Solder Flux	Welding Equipment and Supply Company (C.D.)	**		5/6	5/10	Stock	*5/14	229443	$40.50
4	5	Contact Assy, Parts, and Octal Tube Sockets	Wedemeyer Electronic Supply Company (C.H.)	**		5/6	5/10	Stock	*5/21	229442	$3.74
5	2	Time Delay Relay	Amperite Company (C.H.)	**		5/6	5/3	Stock	*5/10	227850	$4.80
6	1	Repair of Drive Drill	Sears, Roebuck and Company (C.D.)	**		5/6	5/6	Cancelled		227849	$5.00
7	2	Cylinders of Argon	Plant Stores (Univ. of Michigan) (R.K.)	**		5/9	5/10	Stock	*5/14	253393	$60.00
8	1	Cable and Hose Assembly	Liquid Carbonic Corporation (G.E.)	**		5/10	5/7	Stock	*5/10	227870	$15.00
9	1	Mica Window Radiation Counter	Nuclear-Chicago Corporation (C.H.)	**		5/14	5/20	6/10	*6/5	230853	$50.00
10	1	Fabricate Induction Furnace Pipe	F. G. Shaefer Company (C.H.)	**		5/14	5/14	Canceled		229821	
11	25	Brady Wire Markers	Graybar Electric Company (C.H.)	**		5/16	5/22	Stock	*5/31	231271	$3.75
12	5	Tube Fittings	Taylor Supply Company (H.M.)	**		5/16	5/24	Stock	*6/5	231677	$2.10
13	4	Crucibles	Norton Company (J.P.)	**		5/17	5/17	6/28	*6/5	229853	$60.00
14	2	Hoke Bar Stock Angle Valve	H. E. Lennon and Son (C.H.)	**		5/20	5/20	Stock	*5/24	229864	$12.00
15	2	Chromel Alumel Thermocouple Wire	Thermo Electric Company, Inc. (C.H.)	**		5/20	5/22	Stock	*5/27	229887	$8.00
16	3	Ring Joint Flange	Crane Company (R.K.)	**		5/24	6/3	Stock	Cancelled	232835	$12.00
17	--	Screws, Nuts, Washers	Royall Inc. (C.D.)	**		5/24	6/3	Stock	Cancelled	232836	$13.70
18	125	Cinch Y-142, Belden Lugs	Wedemeyer Electronic Supply Company (C.H.)	**		5/22	5/23	5/22	*5/22	L-5516	$3.12
19	2	Rotary Switch and Dial Plate	Allied Radio Corporation (C.H.)	**		5/22	5/22	Stock	*5/31	231268	$2.45
20	4	Rolls of Electric Tape	Puritan Electric Company (C.D.)	**		5/29	5/29	Stock	*6/7	231840	$8.00
21	1	Dayton Shaded Pole Motor	W. W. Grainger Company (C.H.)	**		5/29			Cancelled		$4.30

*Item received.
**Expendable item, Chrysler approval not required.

Appendix

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