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OAK RIDGE NATIONAL LABORATORY

Oak Ridge, Tennessee

TECHNICAL DIVISION

Report for Quarter Ending March 1, 1948

M. C. Leverett

March 5, 1948

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Technical Division Report for Quarter Ending March 1, 1948

0. Abstract

The Technical Division continued to work mainly on the high flux pile and its associated chemical and metallurgical problems during the quarter ended March 1. The principal developments during this period were as follows:

1) Extensive testing of the active assemblies for the pile shows that they are entirely satisfactory from the points of view of mechanical stability, heat transfer, and corrosion.

2) A full-sized beryllium assembly made of beryllium rods bolted together was completed with its dimensions well within tolerance. This is an important milestone, since it is an actual solution of one of the most difficult problems of the pile, namely the fabrication of the beryllium. It remains to subject this assembly to functional tests.

3) Testing of the components of the pile has been essentially completed, except for tests on the regulating rods, the shim-safety rods and the magnets. Most of the remaining work will be done on the full-scale pile mock up which is now starting into construction.

4) It is now proposed to replace the graphite immediately adjacent to the pile tank with BeO pellets in order to save on metallic beryllium formerly required for sleeves around experimental holes. A saving in air flow will also result.

5) Several simplifications of the design of the pile are being made by the elimination of some of the experimental facilities, which it is now felt are not needed.

6) Work on corrosion and heat transfer in the pile has resulted in a change of the design basis for the circulating water system from the requirement that the purge and makeup be at the rate of 5% of the circulation rate to a new requirement that the purge and makeup be 1% of the circulation rate. This will save considerably on demineralizer capacity and on retention basin capacity.

7) Operation of the pilot plant for the separation of U^{235} from fission products and aluminum was carried on throughout the period. The maximum level of radioactivity reached so far is that corresponding to the Clinton pile. Although it is not possible to determine exactly the decontamination factors achieved under these circumstances, it is clear that they are in excess of 10^4 and it is felt strongly that they are well above the required 10^5 for two extraction cycles. The uranium losses have been well below the limit of .1% and have averaged about 1/5 of this. No serious difficulties in operation of the equipment have been encountered, although considerable work remains on the filtration step.

[redacted]

Technical Division Report for Quarter Ending March 1, 1948

1. General

The work of the Technical Division continued during the past quarter to be primarily on the problems associated with the high flux pile. During the quarter it was announced that this work will eventually be transferred to the Argonne National Laboratory, and as a result certain parts of the design work have been dropped. These were primarily the work on the site plan, blasting tests, work on the pile building and the water system. The principal design effort has been on the details of the pile mock-up. Arrangements for continuing the design of the pile have not yet been completed.

Although the change in plan interferred seriously with design work, experimental work continued with only minor interruption.

In addition to its work on the high flux pile, the Technical Division continued work in heat transfer to molten metals, in heavy aggregate shielding, the preparation of Ra-Be sources and in the testing of small pumps. It took on two new assignments; one for devising a method of monitoring the air flowing through the Clinton pile and the other for developing a standard design of hoods for laboratory work.

2. Pile Design and Development

2.1 Design Progress (J. R. Huffman)

Considerable time was lost in design due to the withdrawal of Kellex, the contemplated appointment of Jackson and Moreland, and the Atomic Energy Commission's decision to move the reactor program to Chicago. On January 1, 1948 it was decided to concentrate effort on the remaining problems associated with the pile structure and the detailed design of the mock-up. Work on these has progressed at a satisfactory rate and is described in the following paragraphs. On February 8 and 9 meetings were held with the Atomic Energy Commission's Reactor Safeguard Committee, whose report is not available at this time. Several ideas of improved design resulted from these meetings and are being studied. In view of the change in reactor program, a considerable number of decisions are required before design can proceed at full speed again. Until these are forthcoming, work will continue to be directed toward completing the pile structure design as broadly conceived before January 1, 1948.

Beryllium Reflector Design

During the last period several problems developed in the design of the beryllium reflector. These problems have been solved fairly satisfactorily. First, to insure that thermal stresses do not exceed certain limits, it was necessary to design two types of beryllium pieces in the reflector: those next to the active lattice to

have 0.080" x 1" slots constituting 9% water by volume and developing 6000#/sq.in. stress and those elsewhere in the reflector to have 1/8" holes on 1" centers, having 2% water by volume and 3000#/sq.in. maximum stress. Second, the aluminum indentations in the tank develop more heat than previously thought and have been designed with an annulus fed by water through enlarged holes in the beryllium above and discharging into similar holes in the beryllium below them.

The general fabrication scheme is shown on Drawings TD-390 and TD-391. (These drawings are not included in this report.) Beryllium bars, assembled by brazing several extrusions together, are placed in the tank with their long axes vertical. Each bar is made up of one, two, three or four individual extrusions, brazed together as required in lengths of approximately 40 inches, the height of the reflector. Each extrusion is of approximately 1" x 3" cross section as brazed, and an outside limit of 1-3/8" x 3-5/8" has been assumed as the maximum cross section available. Only in one or two cases are these limits exceeded. The entire assembly of metal is supported at the lower grid support structure, including that metal over beam hole indentations wherein the load is transmitted through the indentation itself to the beryllium metal below, thence to the support structure.

Orientation of the pieces other than the directly removable sections is effected by the use of two dowels at the lower ends of the sections into the lower grid support structure. Top positioning is effected through a single top dowel in each piece extending through the upper grid support, except for the peripheral pieces at the tank wall which are doweled at their upper ends to a tank wall flange. Each directly removable section is oriented and held vertical by an end box similar to that used on the standard fuel assembly. It is anticipated that these sections may be replaced by plate type assemblies of a substitute metal which might require large volumes of cooling water, consequently a large lower opening through the supporting structure is provided below the square type end box. A nominal clearance of 0.017 inch has been provided between the various sections of the reflector matrix. Cumulative tolerances are assigned to provide a minimum of 0.005 inch and a maximum of 0.029 inch clearance. As now designed, all the beryllium can be removed and replaced with difficulty except that beneath the beam hole indentations. While this metal might conceivably be removed without harm to the tank, it is doubtful that it could be replaced remotely.

In accordance with the plans for detailing the pile mock-up at this Laboratory, the job of detailing the reflector has been under way continuously since December 1. Major changes were effected as late as January 15; however, as of February 1 approximately 90% of the information required for detailing was made available. The present status of the detailing job is summarized as follows:

Approximate number of pieces in the reflector		220
Approximate number of drawings required		46
Detail drawings in process		8
Detail drawings completed and checked		13
Number of pieces represented by details completed		76
Active Section Beryllium		complete, including end boxes
Reflector Assembly Drawings		
Number	4	
Complete	1	
30% Complete	1	
Estimated status of entire job		50% complete

Information for the design of isolated pieces around the high temperature experimental opening and in the discharge hole is still required.

The design, while solving the cooling and stress problems, required a large amount of cooling water. The pressure imposed upon the system in order to cool the shim-safety rods is more than adequate for proper flow in and around the other components in the tank. Flows are balanced by restrictions at the lower assembly grid and in the plate under the beryllium. The total flow of cooling water amounts to 19,300 gpm, which is considerably above the 15,000 gpm previously chosen for the design. This value is based on a pressure drop of 35 pounds per square inch across the pile, and 30 feet per second water velocity through the lattice and the beryllium.

By cutting flow outside the assemblies in the active lattice to 25 ft/sec, balanced pressure drop through the pile can be obtained with 18 ft/sec velocity in the permanent beryllium. There will be only a small amount of cross flow from the reflector to the lattice which occurs near the top in order to adjust entrance losses. This reduces the flow to 16,500 gpm. To reduce the flow outside the assemblies to this velocity requires that the clearance between the shim-safety rods and the lower assembly grid be reduced and that the maximum metal temperature increase by about 7°F.

By interlocking the beryllium pieces in the row adjacent to the active section, proper flow patterns in the beryllium are obtained for a velocity of 12 ft/sec in the beryllium. The resulting cross flow is small and the total flow is reduced to 15,400 gpm. Decision between these alternates has not been made.

Mock-up Design

All design work on the mock-up except the reactor tank and its contents have been completed by Kellex, approved by the Technical Division and turned over to J. A. Jones for construction. The Design Section has set up a group under W. R. Gall to complete the design as rapidly as possible. This group started to work on January 15, 1948 and is scheduled to provide final drawings on the tank and its important contents at various times between March 1 and April 1, 1948. Other components to be tested in the mock-up will be scheduled later.

The attached sketch pictures the tank with its principal dimensions but does not show any interior mechanical details. The following paragraphs outline important details and problems.

Sections A and B: Drawings for these sections of the tank are essentially completed and ready for checking. Inside diameters of the tank sections, inside, outside and bolt circle diameters of flanges and gasket dimensions are those for the actual pile in order to test accessibility and other characteristics of the joints. These sections will be constructed of steel plate ASTM-H70 fire-box quality, having ½" walls. The two sections will be joined by serrated face steel flanges against an aluminum gasket 1/2" wide by 1/16" thick. The top plug will be joined by grooved flanges with neoprene rubber gasket. Water connections are 36" O.D. pipe welded to the tank wall and provided with 36" flanges. In the mock-up the pipe size is reduced to 24" O.D. by a reducing flange.

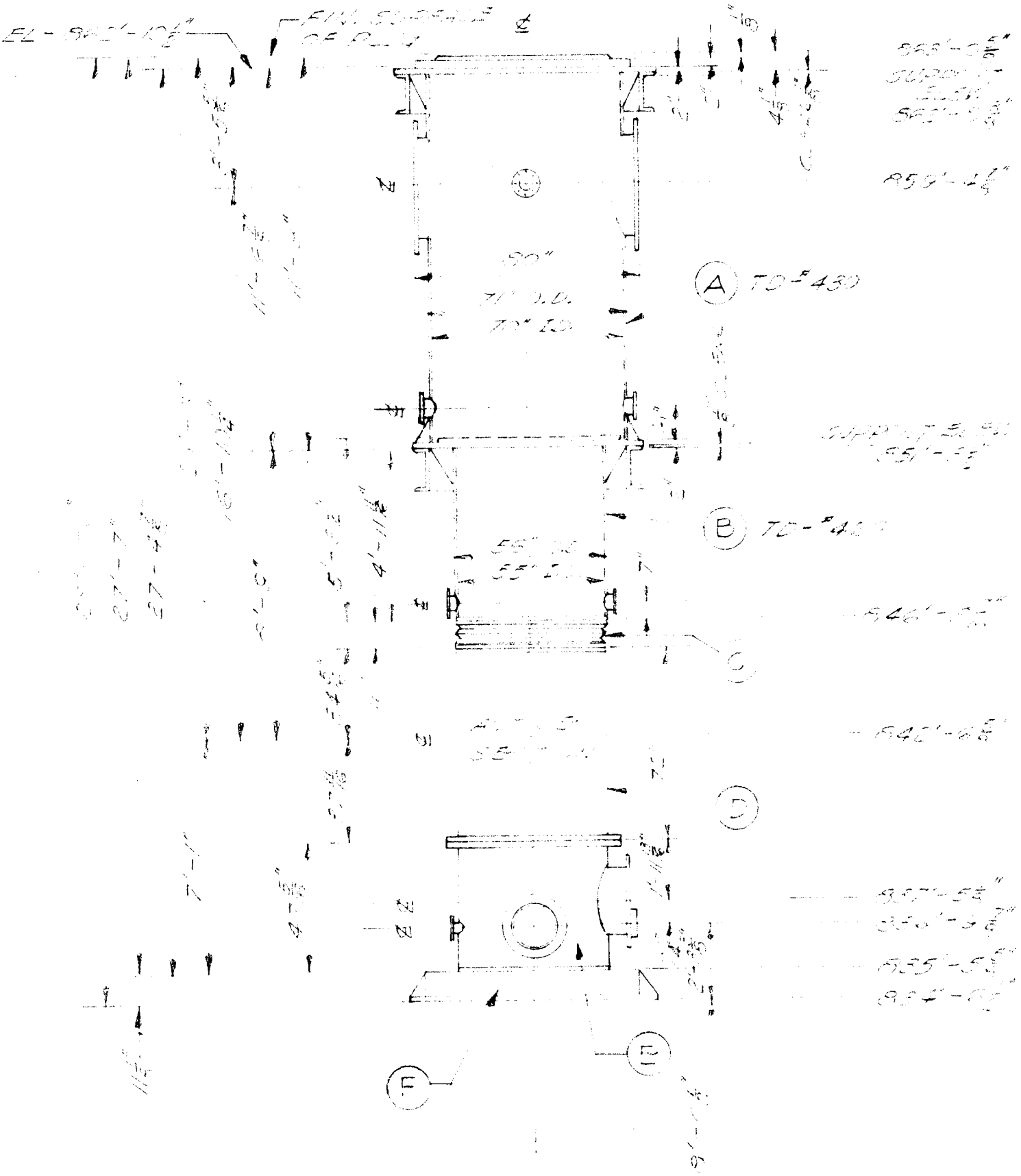
Section C: The expansion joint section is to be supplied by the Cook Electric Company of Chicago to fit the following specifications.

Bellows: Material, stainless steel type 347
Inside diameter, 50"
Outside diameter, 56"
Number of convolutions, 4
Design pressure, 75 psi
Longitudinal flexibility, 3/16"
Transverse flexibility, 1/32"

End Flanges: Material, carbon steel
Inside diameter, 49-3/4"
Outside diameter, 55-3/4"
Bolt Circle diameter, 52"
Minimum thickness, 1-1/4"
One flange to be drilled and tapped
1/8"-9 N.C., 44 holes

Drawing # 5414

DIMENSIONAL LAYOUT OF MOVE TANK



The other flange drilled 7/8" dia. 44 holes
Each flange to have a serrated face between the bolt circle and the outside diameter
Bellows to be joined to outside edge of flanges so bolts will be on the inside.

Section D: This section, made of aluminum 3/4" thick, is in the final stages of detailing and will be ready for checking in March. Methods have been developed for attaching the upper and lower grid support to this section; the former by lugs and dowels and the latter by an internal groove and segmented slip rings. Section C is joined by means of internal flanges and an aluminum gasket using forty-four 3/4 inch bolts. Section E is joined by external flanges and an aluminum gasket using forty-eight 1 inch bolts:

Six 6" I.D. indentations corresponding to the horizontal experimental holes in the pile are to be welded into the tank wall and will be determined by the design now in progress on the beryllium reflector. For the mock-up, sight glasses will be placed at the faces of these indentations. A feasible design for the 1" horizontal pneumatic tubes is ready for checking. For the 1/2" tubes design is complete but drawings have just been started.

The advisability of including these pneumatic tubes in the pile is not yet established beyond doubt. The final decision may depend on the results of the mock-up tests.

Design of the grid supports is in progress. Methods of locking the grids in place are being developed.

Sections E & F: Section F is in the drafting room and will be ready for checking March 1. Section E is being laid out and should be ready March 5. Section F is essentially the same as designed by Kellex. It simulates the bottom shielding plug of the pile except in depth. Stress calculations on the load and impact caused by the shim-safety rod shock absorbers are in progress. These sections are of steel and are joined by serrated flanges and an aluminum gasket.

Graphite Reflector Design

Due to the serious deterioration of the graphite under fast neutron bombardment, which necessitates protective beryllium sleeves around each of the beam holes, consideration has been given to the possibility of substituting a one-foot annulus of beryllium oxide pebbles for the graphite adjacent to the tank. The advantages of such a substitution are listed as follows:

a) Elimination of the "replaceable" graphite ring

b) Elimination of the beryllium protective sleeves

c) Absence of the Wigner effect on beryllium oxide

d) Marked decrease in the air cooling requirements

e) Ease of installing and removing pebbles compared to graphite slabs

The comparative high cost of the beryllium oxide pebbles will be compensated by the elimination of beryllium sleeves and the reduced power cost for the air cooling. The results of preliminary calculations of the cooling requirements for pebbles are given below:

Pebble Diameter	Pebble Temperature	Air flow @ S.T.P.	Pressure Drop
1/2 inch	500° F	3100 CFM	39" H_2O
1/2 inch	600° F	2500 CFM	26" H_2O
1 inch	500° F	3200 CFM	17.5" H_2O
1 inch	600° F	2600 CFM	12.0" H_2O

Since the heat capacity of the air is controlling, the air flow requirement is essentially independent of pebble diameter. The pressure drop, however, decreases with increasing size of pebbles, which points toward the use of the largest size of pebbles consistent with the ease of removal. The pressure drop across the whole system should be kept at 30 - 40 inches to minimize the number and size of entrance ducts.

Consideration has been given to the problem of stacking the graphite in the section beyond the beryllium oxide annulus. An isometric drawing has been prepared to show one possible arrangement and to aid in the design of the graphite around beam holes, etc. The graphite in this section so far remains unchanged from the original conception.

The question of heat generation in the reflector has been reviewed to provide more accurate design data than given in MON P-272. Values in MON P-272 are high due to the assumption of an infinite slab source.

Design is in progress on experimental hole liners and plugs. This involves evaluation of types of experiments to be done, handling of plugs by coffins, shielding and heat removal. Several conceptions are developing, but it is too early to present concrete proposals.

Kellex Contract

The Kellex Corporation has been preparing Status of Work Reports and assembling the Design Notes Files. All drawings have been completed and will be incorporated in these reports. In addition Kellex is preparing a general report giving new ideas, opinions and recommendations. Except for the last mentioned report, these documents will be delivered to the Technical Division prior to March 1, 1948.

Contact men have followed, checked and assisted in the work of preparing these reports. Individual ideas, concepts and recommendations discussed with Kellex personnel prior to their withdrawal from the project have been collected and noted for future consideration.

2.2 Engineering Development (R. B. Briggs)

Scale Formation and Water Treatment

Enough experiments have been completed and the mechanics of scale formation are understood well enough to make possible the conclusion of the original program of scale formation studies within the next few weeks. Scales are formed by two mechanisms on aluminum heat transfer surfaces cooled with demineralized water, one being the deposition of aluminum and iron from the water stream and the other the adherence of corrosion products. Experiments have been run in which the scale formation rates were maintained well below the permissible maximum.

Two sets of apparatus are used in the tests. Both consist of a stainless steel tank to contain the water, a pump for circulating the water, aluminum and stainless steel tubular specimens heated by steam and operated in parallel and a falling film cooler for cooling the liquid before it is recirculated. Rates of total scale formation are determined by measuring the rate of change of the overall heat transfer coefficient as calculated from the increase in metal wall temperature and the increase in steam-side coefficient necessary to maintain a constant heat flux of 500,000 BTU/ft^2 hr. The distribution of scale at the end of each run is determined by the change in overall heat transfer coefficient that occurs as the scale is removed from the coolant side of the specimens. Extensometers are used to measure the metal wall temperatures as indicated by the length of the specimen and empirically determined correlations of steam pressure drop and steam-side coefficient permit calculation of changes in the steam-side coefficient.

Scale formation rates are expressed in terms of resistance units per day where a resistance unit is $10^6 \times \Delta \frac{1}{U}$ and U is the heat transfer coefficient in units of BTU/(hr) (ft^2) (^oF).

Scale Formation and Water Treatment

Rates of scale formation of less than 2 units/day are considered to be satisfactory. The relative importance of the two methods of scale formation for a particular set of conditions is determined by a comparison of data for the stainless steel and aluminum specimens operated in the same system. The only scale that forms on stainless steel at a measurable rate is deposited from the water.

In the aluminum and stainless steel systems under consideration the corrosion rate of stainless steel in water is so small that the corrosion rate of the aluminum and the resulting concentration of aluminum in the water determine the rate of deposition of scale. Data for the pH range 4.0 to 7.0 show that the rate of deposition of scale is less than 0.5 units/day if the aluminum concentration is kept below 0.1 ppm. With 0.18 to 0.22 ppm of aluminum present in the water scales formed at rates of 5 to 35 units/day. Such scales are gelatinous and transparent and thick enough in some instances to increase the pressure drop in the system.

With less than 0.1 ppm of aluminum in the water the scales that retard the transfer of heat are primarily the normal aluminum oxide films which protect the surfaces from corrosion. They form at a rate of the order of 5 units/day for the first few days and then continue to form at a rate of 0.1 to 0.5 units/day for an indefinite length of time. These scales are very dense and less than 0.001 in. thick so they have no effect on the pressure drop in the system.

Some work has been done in cooperation with Dr. Newman of the Chemistry Division in an effort to reduce the rate of formation of the normal aluminum oxide oxide films by pre-forming a thin, highly resistant film. It has been found that heating aluminum in a 1/2% sodium dichromate solution at 125° C. for periods as long as 30 days results in a film 0.1 micron in thickness and with chromium as the major metallic constituent. The film resists attack by dilute hydrofluoric acid and its resistance to flow of heat is negligible. Scale formation rates obtained for a tube treated in that manner were 0.2 to 0.3 units/day for a period of 30 days.

In planning for the cooling of the high flux pile it was initially assumed that the purge rate for the system might be as much as 5% of the recycle rate. However, results of the present experiments have demonstrated that operation with less purge should be possible. A test has been completed under conditions approximating those considered reasonable for the pile coolant system. Operation for 26 days at a heat flux of 500,000 BTU/ft^2 hr, an average water temperature of 50° C, with water at pH 5.8 and containing steady state concentrations of 0.005 M H_2O_2, 0.05 ppm of iron and 0.08 ppm of aluminum resulted in scale formation rates of

less than 1 unit/day. The purge was 1% of the recycle rate and the make-up water contained 0.03 ppm of iron and 0.06 ppm of aluminum. It is, therefore, now recommended that the cooling system be purged at 1% of the circulation rate.

Laboratory testing of flocculation procedures aimed at removing a maximum amount of iron and aluminum from Clinch River water have been completed. It was found that the lowest residuals were obtained by flocculation at pH 6.5 to 7.0. With alum as the flocculent the aluminum residual was 0.1 ppm and the iron residual was 0.02 ppm. When "Ferrisul" was used as a flocculent, the amount of iron in the water remained about the same but the aluminum content was reduced to about 0.08 ppm.

Tests are being run with "Permutit" resins from the Hanford demineralizers to determine whether they will be usable in the demineralizers associated with the high flux pile. Indications are that they will be satisfactory. The iron and aluminum content of the water are reduced to 0.02 to 0.04 ppm., the sodium content is generally less than 1.5 ppm and the average specific resistivity of the water is about 150,000 ohms/cm^3. The capacity of the resins appears to be normal.

Vibration of Sub-Assembly

Investigation of the vibrations induced in the fuel assemblies by the high velocity water stream has been completed with the testing of an 18 plate assembly made with "combs", or spacers, brazed to the centers of the leading and trailing edges of the plates. Installation of the combs made a marked reduction in the amplitude of vibration not only at the ends of the plates but also at the center. The maximum amplitude was reduced from 5.0 mils to 0.8 mil at the leading edge, from 2.6 mils to 1.2 mils at the center and from 2.0 mils to 1.0 mil at the trailing edge. Amplitudes of vibration of the side plates remained essentially unchanged. Vibration frequencies were similar to those observed in the absence of combs.

Following the vibration measurements, the assembly was run for slightly more than 900 hours with the water velocity maintained at 30 ft/sec. There was no evidence of faults in the design or construction of the assembly.

Thermal Warping

Laboratory investigation of the thermal warping of the fuel assemblies has been discontinued with the testing of an 18 plate assembly fabricated of 2S aluminum. The curved plates of the assembly were heated by chromel strip heaters installed along the edges of the plates and heat was removed from the assembly by spraying water on the side plates. Temperatures throughout the assembly were measured with thermocouples attached to the plates and with a probe made from a hypodermic needle. Warping of the assembly and of individual plates was determined from the change in position of the points on the assembly as measured from a reference stand.

With a temperature difference of 35° C between the outer surface of the side plates and a position on the curved plates 1.5 cm. from that surface, the curved plates bowed 0.005 in. in the direction of curvature. The conditions correspond reasonably well to those expected in the pile. The observed longitudinal expansion was 0.025 in. and agreed well with values calculated from the change in the average temperature of the assembly. No permanent change in dimensions was observed during tests which included an 8-day thermal cycling test, involving about 30 cycles.

Plate Buckling in Water Stream

Buckling of the outermost concave plate occurred in the old 12-plate assemblies on being tested in high velocity water streams because the gap on the concave side of the plate was greater than that on the convex side. This buckling was caused by Bernoulli forces resulting from the greater-than-normal velocity and less-than-normal static pressure in the large gap. Although buckling has not been observed when testing the latest design 18 plate assemblies, the magnitude of the possible pressure differences has been calculated and static load tests have been performed to determine the pressure differences required to buckle the curved plates.

Calculations have shown that, neglecting the possibility of a channel with a sealed outlet, the maximum possible pressure difference across a single plate for a pressure drop across the pile of 30 lb/in^2 is 16 lb/in^2 occurring when the concave surface of a plate faces a channel where an entire assembly has been omitted. For a more reasonable but still unlikely case, the expected maximum pressure difference is about 8 lb/in^2.

Measurements of the static pressure differences required to buckle the plates have been made using water pressure in assemblies with flanged end boxes. With assemblies of 2S aluminum plates it was found that pressures of 10, 15 and 20 lb/in^2 produced permanent reductions in the sagitta of 0.000, 0.011 and 0.024 in. respectively and that the first concave plate reversed curvature when the pressure reached 23 to 25 lb/in^2. Strain gage measurements showed that a stress of 5000 lb/in^2 is reached at the surface of the center of the plate when the pressure is ~12 lb/in^2 and that the stresses near the edges of the plate are much less than those at the center. Similar tests with an assembly containing curved plates with uranium-aluminum alloy cores and 2S aluminum cladding gave the same results.

In some of the tests care was taken to restrict the width of the assembly by clamping steel bars along the sides so as to make possible a comparison of the observed plate deformation with the deformation calculated for this special case. In one such experiment the maximum increase in width of the assembly was 0.005 in. and the critical pressure was 34 lb/in^2 compared with 40 lb/in^2 calculated for no change in width.

The maximum increase in width of an assembly with no side support was 0.012 in. and the critical pressure was 24 lb/in^2.

Mechanical Stability of Plates - Conclusions

The foregoing tests permit the conclusion that the active assemblies as now designed and fabricated are mechanically stable to a satisfactory degree.

Testing of Shim-Safety Rods

The shim-safety rod for the high flux pile has four major components; the drive mechanism, the magnetic coupling, the control section with guides and the damper. Testing of the drive mechanism, guides and damper designed for the original 12-plate control assembly are in progress now, as is testing of the latest design of magnetic coupling.

The drive mechanism consists of a 10 ft. long, vertical tube through which passes a 13 3/4 ft. long shaft. The shaft is guided by a spline and driven by a motor through a worm drive to an acme thread on the top 3 ft. of the shaft. Measurement of some of the important characteristics of the drive mechanism has been completed. Data show the following:

Maximum travel	32 3/4 in.
Backlash	
Worm and worm gear	
Angular movement of motor shaft	4 1/2°
Equivalent travel of drive shaft	0.0002 in.
Spline	
Angular movement of drive shaft	1 1/4°
Equivalent travel of drive shaft	0.0006 in.
Total	
Angular movement of motor shaft	20° to 22°
Equivalent travel of drive shaft	0.0008 in.
Overtravel of drive shaft after current cut-off	
Upward overtravel	0.11 in.
Downward overtravel	0.17 in.
Number of revolutions of motor after current cut-off	
Upward overtravel	9
Downward overtravel	14
Speed of travel	
Upward	0.357 in/sec
Downward	0.368 in/sec

Time required for the mechanism to change direction in response to a signal will be measured when instruments are released from other experiments.

The overtravel is greater than the 0.02 inch upward and 0.07 inch downward specified in the design. However, the speed of the test rod is approximately four times that now planned for the pile. A new drive of one-quarter speed is being made up and it is believed that the overtravel will then be reduced to a satisfactorily low value. In the most recent design a worm drive has been specified which should reduce the speed and overtravel to 25% of the values reported here.

A horseshoe magnet with laminated pole cores and a solid armature is the latest coupling design. Coils formed around each core contain a total of 7400 turns of #24 ga copper wire with a total resistance of 200 ohms. When made with SAE 1010, annealed steel, pole cores the magnet is designed to support a load of 1100 lbs, with an effective air gap of 0.015 in. and 200 milliamps of current flowing through the coil. The normal load will be 570 lb. and it is required that the load be released within 30 milliseconds after the release is signaled.

The magnet fabricated for testing was not made entirely according to specifications. Laminae for the pole cores were made of SAE 1020 steel; difficulties experienced in winding the coils limited the total number of turns to 7010; assembly techniques were not well developed enough to reduce the internal air gaps below an estimated 0.010 in. However, data show that the supporting power of the test magnet is adequate.

Tests are made with the magnet bolted to a structural steel support. Fastened to the armature is a bucket which can be loaded with as much as 400 lb. of lead. Additional load is provided as live load by a lever system attached to the bucket with springs and loaded with lead weights. Shims of mica separate the armature from the magnet simulating the non-magnetic material that will be used as a corrosion resistant coating for the steel. The thickness of non-magnetic material will be adjusted to provide adequate protection and minimum release time so some data have been obtained for shim thickness from 0.001 to 0.010 inch. A shim thickness of 0.003 in. is thought to be the minimum reasonable thickness so most of the data are obtained with 0.003 in. shims.

Pertinent results are reported below:

Supporting Power vs. Current for 0.003 in. Shim

Current - milliamps	0	40	80	120	160	200
Max. load - lb.	0	170	560	850	1000	1020

Supporting Power vs. Shim Thickness for Current of 200 Milliamps

Shim Thickness - Inches	0.002	0.003	0.005	0.008	0.010
Maximum Load - Lb.	1050	1020	960	870	810

Release Current vs. Load for 0.003 In. Shim and Initial Current of 200 Milliamps

Load - Lbs.	850	600	450	300	150
Release Current - Milliamps	75	38	23	15	5

Measurement of release time and realization of a satisfactory release time have both proved to be troublesome. The release time is determined by measuring the interval between the signal to release and the making of a contact after the magnet falls a measured distance, usually 0.002 to 0.010 in. Originally this time was measured with an RCA microsecond timer but the results proved to be so erratic that a G.E., mechanical oscillograph has been substituted for the timer. Traces made by light reflected from two galvanometers are recorded by a camera to show the decay of voltage or current in the circuit and the release time superimposed on a time trace from an oscillator.

Power is supplied to the magnet from an electronic circuit like that proposed for the pile. A steady voltage of 10 volts is fed to the circuit to simulate the signal from the ionization chambers. As the voltage increases to 24 volts, indicating an increase in power level, the current to the magnet decreases from 200 milliamps to zero. In the first tests the increase in voltage has been instantaneous and the following results have been obtained:

Effect of Parallel Capacitance on Release Time

200 Milliamps - 0.003 in. Shim - 610 Lb. Total Load

Capacitance, farad	0	0.25	1	2	10
Release time, millisec.	43 to 51	37 to 42	34 to 39	31 to 38	53to55
Current decay time, millisec.	1.5	10	---	---	---

With no parallel capacitance, a 0.003 in. shim, 610 lb. load and 80 milliamperes current, the release time is 29 milliseconds. With no parallel capacitance, a 0.010 in. shim, 610 lb. load and 140 milliamperes current the release time is 8 milliseconds.

Although this last result is very encouraging, data have been obtained for only one such test so the release time may be in error.

Preparations are now being made to repeat some of the tests using a laminated armature and to feed the simulated signal from the ionization chamber with periods varying between 10 and 50 milliseconds thus providing a closer approach to pile conditions. It is expected that these changes and possibly some increase in shim thickness over the present 0.003 in. will result in a release time of less than 30 milliseconds.

Testing of the shim rod guides and damper is being accomplished using a 10 in. pipe column containing a stainless steel mock-up of part of a control element. Fall of the element is guided by standard guides and is stopped by a standard damper. Water flows through the assembly with a pressure drop that can be regulated between 0 and 50 lb/in^2. The position of the rod as a function of time of fall is recorded using the G.E. oscillograph. Contact is made between a point on the rod and stationary points at measured distances along the path and a record of those contacts is pictured on the camera film. The pressure in the dash pot is measured and recorded using a strain gage pressure transmitter and a Brush recorder.

A comparison of design and measured characteristics for a pressure drop of 40 lb/in^2 across the rod shows the following:

	Design	Measured
Fall, in.	30 7/8	30 7/8
Initial Acceleration, g	4	4
Maximum velocity, ft/sec	22 1/2	16 to 20
Velocity when Plunger Contacts Bottom of Dashpot, ft/sec	2 1/2	3 1/2
Maximum Pressure in Dashpot, lb/in^2	475	238

Since the acceleration required for control purposes is 1 g, the difference between the expected and measured maximum velocities is unimportant. The difference results from friction in the guides, forces opposing the passage of the high velocity rod through the water flowing at low velocity below the shim rod and, in the experimental apparatus, to a reduction in pressure above the rod as it falls and air expands to fill the space vacated by the rod.

The maximum pressure in the dash-pot was found to be 50% less and the kinetic energy transmitted to the water 20% less than was calculated from deceleration data. This suggested that the plunger was scraping against the dash-pot and when the apparatus was dismantled, scratches were found on the side of the plunger to support that explanation. The scratches may have resulted from a misalignment of the mock-up parts or from movement of the rod during its fall. The damage was not severe and did not appear to hinder the operation of the rod.

Examination of the guides revealed no faults or wear. Their operation appears to be satisfactory.

A complete shim rod is being assembled for testing.

Pile Mock-Up

Work on the Mock-up for testing the hydraulics and the mechanical operability of the high flux pile has progressed to the extent that the structural design and the design of the water system has been completed. Materials are now being procured for that part of the project.

Design of the pile tanks and internal elements for the Mock-up is about one-third completed. Procurement of some items has begun.

Further Work

With the issuing of this report, many of the heretofore doubtful features of the pile design have been justified experimentally. A few additional items remain to be investigated. Of these the pile mock-up, mentioned in the previous section, and the regulating rods are the most important. Additional work on the magnets also will be required.

2.3 Corrosion

The problems of corrosion connected with the high flux pile are solved in all their critical aspects and the work reported this quarter bears mainly on questions of improving an already satisfactory situation. The only condition in the pile which is expected to lead to serious corrosion if not carefully controlled is the junction between the beryllium and the stainless steel. Serious effects here can be avoided by making the beryllium massive and by limiting the number of such contacts. The experimental work during this quarter was carried out under simulated pile conditions (85° C, pH 6.0 - 7.5, and hydrogen peroxide .0005 or .005 M). The principal item of investigation was the use of sodium nitrate as an inhibitor.

Beryllium

The corrosion resistance of extruded beryllium is improved by the use of 3 - 4 ppm of sodium nitrate as an inhibitor. The penetration rate and pitting attack are both most effectively reduced by this inhibitor present in this concentration.

Tests of Q Process beryllium are erratic and sodium nitrate is not an inhibitor for this metal but sodium dichromate appears to have some inhibiting effect.

A beryllium cylinder coated with aluminum by dipping in molten metal showed an overall corrosion rate of 0.06 mil/month with pit frequency of 10 per sq. in. Maximum pit depth was 9.3 mils. One pit at the interface between aluminum and beryllium on a machined end had penetrated along the interface for a depth of 50 mils. When the aluminum cladding was penetrated, the attack of the beryllium was accelerated.

2S Aluminum

Tests of this material show 3 - 4 ppm of sodium nitrate to be optimum inhibitor concentration, but the reduction in penetration rate is only slight.

Specimens, as anodized by Reynolds Metal Company, show only negligible attack. The use of such protection depends upon the feasibility of anodizing a fuel rod assembly and the effect of radiation on the anodized film.

Tests conducted on full size 2S aluminum plates, in three and seven plate assemblies showed negligible attack of plates or brazed joints.

Aluminum Alloys

Tests of 43 casting alloy show sodium nitrate, sodium pyrophosphate and sodium dichromate to be ineffective in inhibiting the corrosion of this alloy. Tests of 52S alloy show marked increase in resistance when the medium is circulated at a velocity of 15 ft/sec. Under such conditions the penetration rate is of the order of 0.07 mil/month. At higher velocities (30 ft/sec.) it is 0.15 mil/month and under static conditions 0.80 mil/mo.

2S Aluminum - 347 Stainless Couple

In such couples the aluminum is attacked preferentially and the stainless is attacked only negligibly. The maximum inhibiting effect of sodium nitrate upon the attack of the aluminum is obtained at 5 - 10 ppm. The pitting attack is severe in all cases and occurred predominantly at the exposed surface adjacent to the outer edge of the aluminum-stainless interface. The application of a thin layer of zinc (4.0×10^{-5}) to aluminum by treatment in a sodium zincate solution has shown results that are quite favorable. The 2S aluminum was still anodically attacked but not nearly so severely as when no zinc layer was applied. Attack of the stainless was still negligible. Pit depth was not reduced but count was reduced by a factor of about 10 as was penetration rate.. This is so far the most successful method of protecting 2S aluminum against corrosion when in contact with stainless steel.

3. 25 Separations Process Design and Development

3.1 Design Progress (J. R. Huffman)

Design work in the 1200 Area reached the preliminary process design report stage during the past period. The first rough draft of the report has been prepared and reviewed. A total of twenty-nine memoranda to the 12-1300 Area Design File and a total of forty-three drawings have been prepared for the 1200 Area design. These drawings and reports present the first overall concept of the functions and scope of the 1200 Area. In view of a change of design basis, and in order to incorporate pertinent data relating to the chemistry and equipment performance for the process, revisions to many phases of the first design must be made before detailed design by an outside contractor can be undertaken.

The major changes to be made in the design of the 1200 Area before the completion of the process design report are:

1) The maximum capacity of the main processing line will be less than 600 grams of 25 per day. The original specification of a capacity of 600 grams per day was based on the assumption that the plant would service 3 piles: the high flux pile, the Daniels' power pile and one other which, it was assumed, would probably be built somewhere in the United States within the next 10 years. It is now believed that the depletion of the U^{235} content of the high flux pile can be carried to 15% instead of 10% as previously assumed and that the building of two other piles in the next 10 years is very doubtful. The question of proper design capacity is thus again thrown open for redecision by the AEC.

2) For the present the chemical flowsheets will be changed to a basis of 240 grams per day (two eighth-pass fuel assemblies) with chemical processing steps and conditions in accordance with the best known chemistry to date.

3) The process equipment flowsheets will be revised to show all pertinent equipment, service, lines, and facilities.

The design status of various phases of the 1200 Area as presented in the first writing of the design report is outlined in the following paragraphs.

The spent fuel assemblies for the pile are to be cooled for approximately 130 days before processing in the 1200 Area, where three weeks will be required to decontaminate and recover the 25 for processing in the 1400 Area. An overall fission product decontamination of 10^8 after cooling, and a Pu decontamination of 10^4 must be accomplished, along with a 99.9% recovery of 25. Spent shim-safety rods containing 25 will be processed in the same equipment under identical chemistry conditions.

The sizes and general function of all pieces of equipment for three extraction cycles for irradiated 25, for solvent treatment and recovery, and for processing cold reject assemblies from the 1400 Area have been discussed in numerous memoranda to file. The chemistry of the process is outlined in chemical flowsheets TD-366 through TD-368; process equipment and main flow diagrams are shown on TD-362, TD-370, TD-372, TD-381, and TD-364. Drawing TD-421, a master flowsheet showing the entire 25 process, will soon be issued.

The estimated activity contained in one 25 fuel assembly after 20-day irradiation in the pile operated at 10 kw/gram 25, and after 150 days cooling is:

Total curies	17,000
Total gamma watts	57

The estimated activity levels in various process streams based on a batch containing 2-1/2 fuel assemblies (i.e., 300 grams of 25) are tabulated below:

1st cycle dissolver product	500	curies/gal.
1st cycle aqueous raffinate	320	" "
2nd cycle column feed	1.4	" "
2nd cycle aqueous raffinate	1.1	" "
3rd cycle column feed	0.14	" "
3rd cycle aqueous raffinate	0.01	" "
3rd cycle stripping product	3×10^{-3}	"

Preliminary study of the problem of equipment and cell decontamination has been made and a drawing, TD-416, issued showing schematically the system for tank and equipment decontamination. Three cycles of decontamination were considered, graded in accordance with activity level. Decontamination acid will be used countercurrently through the equipment, and then fed to the assembly dissolver unless crud removal is complicated thereby. Such a procedure would make uranium recovery from decontamination acid unnecessary.

Salvage facilities for the recovery of 25 from waste materials have been outlined on the basis of rather uncertain estimates of the type and quantity of uranium-bearing wastes. Three cells in Bldg. 1205 have been assigned to salvage operations. Drawing TD-386 shows equipment for salvage schematically.

At the present time, the behavior of Np^{237} in the extraction process is very uncertain. For this reason, no provision has been made for Np recovery in the present design concept.

The waste collection and storage system for the 1200 Area has been combined with those of the 1100 and 1300 Areas. The combined waste system has been designed to prevent loss of fissionable materials from any of the process areas, to provide adequate storage capacity for radioactive

waste solutions, and to provide facilities for the controlled disposal of radioactive wastes. The 1200 and 1300 Areas will be provided with sixteen 10,000 gallon stainless steel collection tanks where all low activity level waste streams will be collected and analyzed before disposal either to storage, uranium salvage, or to the tank farm neutralizers prior to disposal to the retention pond. The waste storage system will consist of (1) twenty 30,000-gallon, six 10,000-gallon, and six 5,000-gallon type 347 stainless steel tanks for radioactive wastes; (2) seven 35,000-gallon concrete-mild steel lined tanks for storage of concentrated wastes containing small quantities of activity; and (3) five 100,000-gallon mild-steel lined tanks for the same purpose as the 35,000 gallon tanks. Estimated volumes of waste streams discharged from the 1200 and 1300 Areas are shown on drawings TD-388, TD-418, and TD-419.

The waste disposal system will include two 7,000-gallon neutralizers and one evaporator in the tank farm, plus two evaporators and one neutralizer to be used in case of an overload on the tank farm system located in the 1200-1300 collection tank area. A retention pond consisting of six 1,000,000-gallon bays, constructed of concrete will be provided for the use of the entire 1000 project. The combined waste system is shown schematically on TD-420. The capacities for waste collection and storage outlined above are based on processing 600 grams 25 per day and 16 kg Th/day for 365 days per year.

A central laboratory will serve the 1200, 1300, 1400 and waste disposal areas. A one story U-shaped building is planned. Facilities will be installed for general chemical analyses, active chemical analyses requiring shielding, mass spectrographic determinations on uranium, and counting. The question of the number of mass spectrographs required has not been decided. Studies of the number of samples to be processed, the number of personnel required, and preliminary exhaust hood requirements have been made. The laboratory building is shown on TD-384.

The canal for storage and cooling of 25 fuel assemblies, shim safety and control rods, and 23 production rods has received only brief consideration. It is estimated that a canal 150 feet long by 10 feet wide and 20 feet deep with approximately 16 feet of water will be required. Handling and sawing facilities have not been investigated.

Three canyon concepts for Building 1205 have been developed and are shown on drawings TD-415, TD-402, and TD-403. Concept #1, shown on TD-415, similar to Hanford canyons, has been favored during preliminary discussions. Four studies of a building in which the Buildings 1205, 1305, 1405, 1706, change house, mechanical and instrument shops, and offices are combined have been made; the drawings are now in the process of being checked. Wall thicknesses for shielding hot cells have not been accurately determined, but the following wall thicknesses have been estimated:

8' concrete - Dissolvers through first cycle extraction cells

5' concrete - Second cycle extraction, hot section of solvent recovery, hot salvage cells

2' concrete	- Third cycle extraction, cold solvent distillation and storage
Normal wall thickness	- Fourth cycle extraction precipitation and ignition areas

General site plan and security requirements have been considered and have been reviewed in the Feasibility Report on Clinton High Flux Pile, Mon T-433, issued on December 1, 1947.

The ventilation requirements of hot cells, general building areas, and the waste storage and disposal system, have been studied. A memorandum discussing these requirements will soon be issued. The exhaust air fan house and stack have been given very little consideration. Ventilation of various areas is summarized below:

General operating and office area	7 changes/hr.
Hot cells and pipe tunnels	15 changes/hr.
Some lab rooms with hoods	20 - 60 changes/hr.

A total of approximately 329,000 cfm air will be required for the combined 1200 and 1300 areas, with 120,000 cfm of this being discharged from the stack.

Common service buildings will serve the 1200, 1300, 1400, and waste disposal areas. This group will include a chemical storage building (TD-392), a mechanical and instrument shop (TD-400), and a change house (TD-401). All buildings will be single story, fire-proof structures.

Heavy isotope production, 25 recycling, and 25 inventory in the 1200 area canal and processing area have been reviewed in the Feasibility Report.

Service requirements such as steam, compressed air, filtered water, demineralized cooling water, building vacuum, and electrical power requirements, have been reviewed and summary reports written to the 12-1300 Area Design File.

3.2 Laboratory and Semi-Works "25" Development

3.21 General

A 25 recovery process which has so far provided satisfactory fission product decontamination and uranium recovery has been developed in the Laboratory and Semi-works and is now in the pilot plant development stage. This process, which follows after the used pile assemblies are cooled 145 days to permit decay of the chemically-inseparable, active 7-day U^{237} isotope, consists of dissolving the uranium-aluminum assemblies,

filtering, if necessary, to remove insoluble materials from the dissolver solution, and extracting and decontaminating the uranium in two continuous hexone extraction cycles, in which a fission product decontamination factor of 10^5 is required. The aqueous phase in both extraction cycles is 1.3M in $Al(NO_3)_3$, 0.05M deficient in HNO_3 (pH = 1.8) and contains the reductants, hydrazine (0.1M) and ferrous ammonium sulfate (0.05M). All operations to this point must be carried out behind massive shielding in a "canyon." An additional solvent extraction cycle is then carried out behind thin shielding for final purification and concentration of the uranium. Ammonium diuranate is then precipitated, and the pure uranium salt is converted to U_3O_8 for metallurgical reuse.

The investigation of process variables is being pursued in the laboratory and semi-works, and process modifications are continually being made as the improvements are demonstrated. Significant process developments of the last quarter are as follows:

> Laboratory work on separation of neptunium, a desirable process by-product, show that it can be satisfactorily extracted if the aqueous pH is 1.2 or lower and if the total hydrazine concentration is increased somewhat. It is not yet known whether fission product decontamination will be satisfactory under these conditions.
>
> In first cycle semi-works extraction, changing the continuous phase from aqueous to organic caused the uranium loss to increase from 0.01% to 0.06% and therefore the H.E.T.S. to increase from 1.4 to 1.7 feet, but did not affect fission product decontamination. This confirms previous scouting work.
>
> Second cycle semi-works decontamination factors range from as low as 20 or 30 to as high as 500, being dependent even at maximum semi-works' radiation levels on the relatively low activity of the feed. They are, therefore, minimum values which may be exceeded in operation at higher levels of activity. Laboratory experiments show that the second cycle decontamination may be improved if the aqueous HNO_3 concentration is about 0.1 or 0.2 molar, which is in contrast to findings for the first cycle, where a slight deficiency of HNO_3 (-0.05M) is optimum.

In the next quarter, semi-works demonstration of neptunium recovery will be made. Also data on the plutonium decontamination obtained in the solvent extraction cycles and on the effectiveness of diphenyl thiourea in promoting decontamination of ruthenium, the most difficultly removable of the fission products, will be forthcoming. In the laboratory, the stability of the hexone solvent in the Redox process under process levels of radiation will be tested, the activity source being either a

laboratory-dissolved solution of Hanford-irradiated metal or some Hanford metal solution from the pilot plant dissolver. Previous tests at other conditions have shown no gross decomposition at such radiation levels, but, at the request of the Redox people of Hanford, this more careful study will be made under actual process conditions.

3.22 Flowsheet Changes

No flowsheet changes were made in the last quarter, but a higher total hydrazine content in the extraction cycles (at least a sum total of 0.2M in both phases) and an adjustment in aqueous acidity (from pH of 1.8 to ca. 1.2) are being considered to make possible neptunium recovery. Also, the acidity in second cycle extraction may be adjusted to a few tenths free HNO_3 rather than to a 0.05M HNO_3 deficiency, since laboratory tests show that second cycle decontamination is improved at the higher acidity.

The use of "bare" solvent extraction cycles, that is, cycles containing neither oxidizing, reducing, nor complexing agents may prove to be adequate; since, with neither oxidizing nor reducing conditions, complete clean-up of activity has been obtained in semi-works and preliminary pilot plant runs with Clinton-slug activity. The necessity for such reducing agents can only be shown in the coming pilot plant Hanford-slug activity runs. Of course, in such bare cycle operation there would be no great neptunium recovery or plutonium decontamination. However, neptunium recovery may not be desired if larger quantities are separated in the Redox process; if they are not, neptunium may be obtained in the 25 process by addition of a suitable complexing agent. Plutonium decontamination in the 25 canyon extraction is of little consequence, since there is little plutonium present and it may readily be removed in the laboratory clean-up steps. The reducing conditions may be required in the pilot plant extractions with Hanford metal to direct the relatively large quantities of plutonium into a single waste which is to be stored in a special tank.

3.23 Metal Solution

Remote Control of Concentrations. Simple methods for empirically determining $Al(NO_3)_3$ and HNO_3 concentrations in the dissolver solution or hot process feed solutions using measurements of pH and specific gravity have been developed in the laboratory. The work of J. B. Chrisney, formerly of the Chemistry Division, who first determined the relationship of pH and specific gravity to $Al(NO_3)_3$ and HNO_3 concentrations in the system $Al(NO_3)_3$ - H_2O- HNO_3, has been extended to include acid-deficient $Al(NO_3)_3$ solutions containing uranium. Nomographs and graphs for the determination of the $Al(NO_3)_3$ and HNO_3 concentrations in these solutions have been prepared and are available upon request. A report of this work will be issued soon.

Semi-Works Demonstration. Installation of a cold semi-works scale dissolver to demonstrate accurate remote adjustment of acidity and $Al(NO_3)_3$ concentration by pH and specific gravity measurements is now complete. A scouting run, made mainly to gain familiarity with the operation, showed that final dissolver solutions which are HNO_3-deficient are easily attainable.

3.24 Crud Removal

The possible deposition of "crud" (coagulated fission products, dissolver corrosion products, silica, and other insoluble materials) in extraction column packing, thereby rendering the columns inoperable, has made necessary a process provision for removal of the "crud" by filtration. As a consequence of this laboratory work, MnO_2 was chosen as a filter aid, and in subsequent small-scale semi-works tests, a procedure using co-formed MnO_2, a porosity "G" stainless steel filter medium, and a preformed MnO_2 precoat 1/4" in depth was outlined for pilot plant operation. However, in pilot plant operation, filtration rates dropped during filtration to excessively low values, both with and without the use of MnO_2 filter aid, and made necessary cleaning the filter twice during the filtration step. Laboratory work was therefore resumed to develop better conditions for the use of MnO_2 as a filter aid, either as the co-formed or preformed type.

As a result of this laboratory work, the use of commercial MnO_2, screened to remove particles less than about 40 microns in size, appears much more attractive than the use of MnO_2 precipitated in solution under the best known conditions. Considerably faster filtration rates are possible with the screened MnO_2 because an "E" porosity sintered steel filter medium may be employed instead of the finer "G" porosity required for the co-formed MnO_2. Uranium hold-up on the commercial MnO_2 is negligible, being 0.16 mg. of uranium per gram of MnO_2 after five washes with an amount of wash just sufficient to cover the precoat. This hold-up is one twenty-fifth that on co-formed MnO_2 after much more washing. The commercial MnO_2 greatly retarded plugging of filter pores by silica-aluminum suspensoids. To further minimize filter plugging, it is recommended that additional screened MnO_2 be added to the solution in order to build up a cake which contains proportional amounts of MnO_2 and dissolver insolubles.

3.25 Semi-Works Column Operation

Semi-works column operation was slowed down in the last quarter by extensive equipment alterations. The first cycle columns were modified to make possible operation with either phase continuous, and to allow the testing of diphenyl thiourea as a specific ruthenium complexing agent in the stripping column. The temporary second cycle columns are now being replaced with new, more permanent columns.

Decontamination and Uranium Recovery with the Organic Phase Continuous. Four first cycle extraction runs with the solvent phase continuous yielded a slightly higher uranium H.E.T.S. but essentially the same decontamination when compared with the previous aqueous phase operation, (H.E.T.S.: 1.8 vs. 1.4 feet, corresponding to 0.05 vs. 0.1% U loss; gamma decontamination factor about 2×10^3). The columns may be run with either phase continuous with equal facility, but all future runs will again be made with the aqueous phase rather than the organic continuous because of its higher extraction efficiency.

In the last run of the series with organic phase continuous, eliminating the scrub feed but correspondingly increasing the extraction feed rate to maintain the same aqueous to organic ratio in the extraction section reduced decontamination by a factor of four, thereby giving some indication of the amount of decontamination effected by scrubbing.

Second Cycle Decontamination

A review of reported second cycle results to date shows large variations in the gamma decontamination factors (from as low as 20 to as high as 500 even under similar run conditions). These variations may be explained by the fact that since essentially complete clean-up of activity is being obtained in two solvent extraction cycles using Clinton-slug activity, the indicated second cycle gamma decontamination factors are very dependent on the sensitivity of the detection method employed. For instance, in semi-works operation, decontamination factors have never been more than about 30, based on ordinary counting measurements. However, the factors on these same solutions have been shown to be as high as 500 if the activity is concentrated 40-fold and measured in an ionization chamber. The gamma factors may therefore be even higher if more sensitive counting methods are developed or if a higher level of activity is employed.

In connection with second cycle decontamination, laboratory studies have shown the beta decontamination factor to increase from 40 to 90 as the acidity is increased from a 0.05M deficiency of HNO_3 to a 0.1M free HNO_3 concentration (aqueous phase is 1.4M in $Al(NO_3)_3$, 0.1M in N_2H_4, and 0.05M in Fe^{++}). This variation in the beta decontamination is only indicative of the variation in the more important gamma decontamination, but the beta measurements were made, since with Clinton-slug activity too few gamma counts are present after a second cycle extraction to be accurately detected. Nevertheless, on the basis of these experiments, the second cycle acidity will probably be adjusted to around 0.1M free HNO_3. It is important to note that these low factors together with the first cycle gamma decontamination factor of about 2×10^3 bring the overall two-cycle gamma factor up to or past the required value of 10^5.

Future Column Work

A series of runs is now underway to demonstrate a means for increasing ruthenium decontamination by complexing the ruthenium in the organic extract with s-diphenyl thiourea in order to hold the ruthenium in the organic phase in stripping. These runs are of a scouting nature

and are part of a program to investigate methods to make possible a one-cycle extraction process which will yield the required canyon decontamination of 10^5. Also this complexing procedure, if promising, will be available for the present two-cycle process if needed (ruthenium activity now constitutes about 85% of the γ activity present in the second cycle feed). Laboratory cold column operation has already shown that the use of diphenyl thiourea in the stripping column does not cause excessive crud formation and that complete removal of the uranium can still be effected without difficulty. After these runs, the use of sulfamic acid will be tried as a holding reductant in place of hydrazine because it is less hazardous than hydrazine. Runs to demonstrate neptunium separation and plutonium decontamination will then be made.

Semi-Works Hexond Pretreatment and Recovery

During the quarter the semi-works continued to gather data on hexone loss over the recovery process. The total average loss over both washing and distillation steps for the last 35 recovery runs has been less than 10%.

The problem of recovering hexone containing large concentrations (about 0.1N) of hydrazine and diphenyl thiourea (DPT), and therefore ruthenium activity since DPT is used as a ruthenium complexing agent, is holding up further hexone recovery. The only method found to date for satisfactory decontamination of this hexone is steam distillation from a caustic heel (various washing procedures are ineffective), but concentration of the activity in the still bottoms makes this method impractical in the present equipment. New shielded distillation equipment with facilities for easy removal of the hot waste is now being installed.

A brief study of the hexone evaporation loss from 55-gallon storage tanks revealed that 0.2 pound per hour evaporates with moderate suction on the tank vents.

3.26 Laboratory Decontamination and Distribution Studies of Neptunium Separation

It was discovered that in an oxidized first cycle, employing dichromate as the oxidizing agent, separation of any neptunium as a process by-product is not possible because, at the low acid concentrations necessary for adequate decontamination, the dichromate will not oxidize Np to the extractable Np^{+6}. But the reduced neptunium can be complexed and extracted, for instance with hydrazine, which is used also as a reducing agent. However, in this report period it has been found that hydrazine complexing also becomes less effective with decreasing acidity. As may be seen in Fig. 2, the pH must be about 1.2 or less to provide a satisfactory neptunium distribution coefficient. The effect of hydrazine concentration is such that if it is lowered, the pH must also be lowered to maintain a satisfactory distribution coefficient. For instance, if the total hydrazine concentration (sum of the molarities in both phases) is lowered from 0.5 to 0.12M, the extraction pH must be lowered from 1.2 to 0.85 to maintain the minimum desired neptunium distribution coefficient of 1. Thus, it appears that the process pH will have to be decreased from 1.8 to around 1.1 if some Np separation is to occur. This change will be made only if fission product decontamination is satisfactory at the lower pH. Work is also underway

Drawing # 5239

Fig. 2

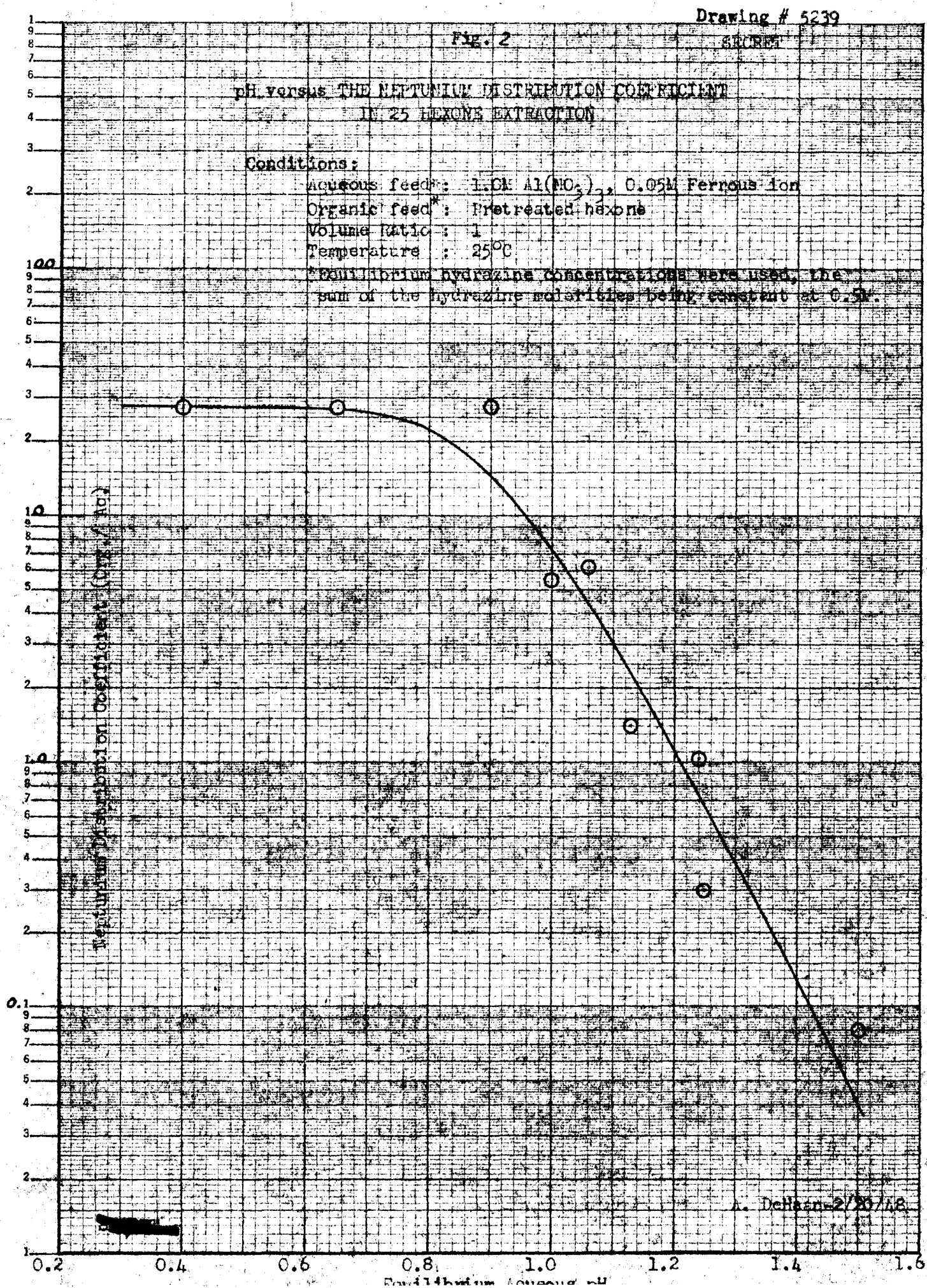
SECRET

pH versus THE NEPTUNIUM DISTRIBUTION COEFFICIENT
IN 25 HEXONE EXTRACTION

Conditions:

Aqueous feed*: 1.0M $Al(NO_3)_3$, 0.05M Ferrous ion
Organic feed*: Pretreated hexone
Volume Ratio : 1
Temperature : 25°C

*Equilibrium hydrazine concentrations were used, the sum of the hydrazine molarities being constant at 0.5M.



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seeking a more desirable complexing agent.

Uranium Complexing with Dibenzoyl Methane

Dibenzoyl methane is one of the various uranium complexing agents which are being investigated as they become known for the possibility of improving uranium-fission product separation. However, dibenzoyl methane is not promising because, although it increases the uranium distribution coefficient (3 to about 8 --org./aq.), it also increases fission product extraction so that there is a net loss in uranium-fission product separation.

Effects of Hexone Impurities on Uranium Extraction and Decontamination

A study of the effects of the small amounts of impurities present in commercial hexone, i.e., mesityl oxide (0.2 to 0.7%), mesitylene (0.05%), and methyl isobutyl carbinol (0.2 to 0.3%), on the uranium distribution coefficient and fission product decontamination is in progress. Preliminary findings using cerium tracer and a non-reduced feed show that the impurities in the listed concentrations have little or no deleterious effect on decontamination. However, uranium distribution is improved somewhat (25% or so) by removal of the mesitylene and mesityl oxide. It is planned to evaluate these effects under present flowsheet reducing conditions.

For detection of these impurities, the reducing strength of the hexone was found to be indicative of the presence of either the carbinol or mesityl oxide. The mesityl oxide content may be evaluated by determining the bromine number of the hexone. Refractive indices were of little value in detecting these impurities.

Laboratory Column Studies

In a 1-inch laboratory glass column packed with 3/16 inch Fenske rings, a scouting study has been made to determine the dependence of the uranium H.E.T.S. on (1) $Al(NO_3)_3$ and HNO_3 concentrations, (2) phase flow ratio, (3) total flowrate, and (4) interface position (aqueous or organic phase continuous). The results are summarized as follows:

1) At a flow ratio of one with the interface at the top of the column, the H.E.T.S. increased from about 0.9 feet to 5.2 feet when the $Al(NO_3)_3$ concentration was increased from 1.2M to 1.8M.

2) An increase in acidity raised the H.E.T.S. slightly, an effect which was nullified because the uranium distribution coefficient also increased.

3) Varying the flow ratio from 0.1 to 2 (org./aq.) had the effect of decreasing the H.E.T.S. if the aqueous was continuous and increasing the H.E.T.S. if the organic was continuous, the magnitude of these effects being small except in the region of flow ratios lower than 0.5.

4) For flow ratios between 0.5 and 2 (org./aq.) and regardless of the $Al(NO_3)_3$ salting strength or which phase is continuous, the H.E.T.S. increased linearly by about 0.1 ft. for every 15 gal./hr./sq. ft. increase in total throughput.

3.27 Laboratory Purification of "25"

A laboratory solvent extraction cycle (either batchwise or continuous) will be made to complete the purification of uranium from the second canyon cycle. In connection with U^{233} process development, complete clean-up of such uranium solutions by batchdiisopropyl ether extraction has been accomplished with negligible loss.

3.28 Decontamination of Waste Streams

A scouting study of fission product decontamination of the aluminum waste streams by various adsorption and precipitation methods is now being made. Dowex 50 resin will decontaminate 0.1M aluminum wastes by a factor of 100 or better for Ba and the rare earths, about 10 for Sr, 2 for Ru, Zr, and Cb, and less than 2 for Cs. Iron hydroxide is a very effective carrier and will decontaminate from Zr and Cb by a factor greater than 100, the rare earths and Sr by better than 50, Ru by about 5, but none from Cs. Other adsorbents such as bentonite, alumina, and charcoal, give gamma factors of 5 to 10 but have low capacities and cannot be regenerated as can the ion exchange resin. Cs, Sr, and Ru remain in all cases the activities most difficult to decontaminate. It is possible to precipitate the Al as $Al(OH)_3$ without carrying the Cs, Sr and Ru activities, but the problem of concentrating a very gelatinous precipitate is then introduced. Other aluminum precipitates will be investigated along with specific Cs, Sr, and Ru decontamination methods.

3.29 Packless Valve Development

The development of a packless valve for "25" process use is continuing with three commercial value companies, Hammel-Dahl, Fulton Sylphon, and Aloyco Steel Co. An altered Fulton Sylphon bellows valve has been operated through 10,000 flexes without leakage or bellows failure.

3.3 Pilot Plant Development (D. G. Reid)

The 1200 Area Pilot Plant, reported completed in the last report, has been tested and uranium previously bombarded at full Clinton Pile level has been processed. The testing was complete with only minor modifications in the equipment. So far there have been 5 runs made with inactive metal through one or the other extraction cycles, 6 runs with active

metal through the first cycle and 4 through the second cycle. The results have been generally satisfactory. Further details appear in the following tables. The results are discussed in the balance of this section.

The active runs were made under bare flowsheet conditions, which consist of 1.35M aluminum nitrate, at 0.05N basicity, and an organic to aqueous flow ratio of 2 with no oxidants, reductants, or complexants. The data obtained showed beta decontamination factors in the first cycle of 3×10^3 and gamma decontamination factors of 1.6×10^3 with total known uranium losses through two columns of extraction and two of stripping of 0.02%. Activity data from the second cycle show that the activity level of the product stream is too low to determine accurately decontamination factors using Clinton material, but these can be determined when processing of Hanford irradiated uranium is started in two months.

However, the present data permit the conclusion that a gamma decontamination factor of 2×10^4 to 4×10^4 <u>at least</u> is being attained in the pilot plant. Probably the decontamination factor is well above this, but definite proof cannot be established yet. Since the desired minimum decontamination factor in the first two cycles is 10^5, it is indicated that the present process is entirely satisfactory from this point of view.

Dissolution

The source of uranium for all the runs has been dissolving of uranium metal. For the three unirradiated metal runs and the first four active runs, dissolution was performed by using the theoretical amount of acid for the metal loaded. This type of dissolution resulted in solutions containing 0.4 - 0.5N excess acid, which in turn required that ammonium hydroxide be added during column feed makeup for neutralization to the proper basicity. Longer heating periods and higher temperatures were not successful in reducing the excess acidity in the dissolver. In the last four runs 100% excess metal has been kept in the dissolver and the reaction extended to a specific gravity of 1.83 instead of the former 1.78. Using this technique, the excess acidity has been reduced to an average of 0.15N. All previous dissolvers and dip tubes with a corrosion history have been fabricated entirely of 309S Cb stainless steel or equivalent. The pilot plant dissolver is fabricated with a body of 309S Cb stainless with 347 stainless dip pipes. Inspection of the dip pipes after dissolving four batches of uranium has shown some etching of the 347 stainless steel dip pipes.

Feed Makeup, First Cycle

Feed makeup is done by adding stock solutions of aluminum nitrate salt to dissolver solutions. Development of the technique has shown that the most suitable method of controlling this feed makeup to 1.35 ± 0.05M in aluminum and 0.05 ± 0.03N in free base is by accurate specific gravity measurements, close control of the acid balances, and the use of theoretical amounts for butting solutions. Using basic aluminum nitrate stock solution in this method, the conditions can be controlled so that no base is added to the shielded tanks, thus eliminating any pos-

Summary of First Cycle Runs

Flowsheet Conditions as Noted in Accompanying Figure

	Inactive			10% Clinton Activity			Full Clinton Activity		
Run	1D	4D	5D	1X	2X	3X	4X	5X	6X
Feed									
Al, M[a]	0.97	1.43	1.35	1.42	1.56[b]	1.39	1.41	1.37	1.36
Free Base, N	-	0.025	0.015	0.072	0.013	0.062	0.034	0.055	0.039
pH	1.06	1.52	1.44	1.84	1.30	1.81	1.83	1.80	1.85
U Losses, %									
Extraction	1.75	0.0087	0.013	0.0026	0.0008	0.0013	0.0013	0.0024	0.0009
Stripping	0.02	0.0086	0.06	0.0021	0.0003	0.0005	0.0013	0.0072[c]	0.0145[c]
D.F.									
Gross Beta	-	-	-	2.0×10^3	1.7×10^3	1.4×10^3	1.1×10^3	2.4×10^3	3.1×10^3
Gross Gamma	-	-	-	0.5×10^3	0.6×10^2	0.7×10^3	0.5×10^3	0.9×10^3	1.6×10^3
Ru	-	-	-	-	-	-	-	100	210
Zr	-	-	-	-	-	-	-	1.2×10^4	1.7×10^4
Pu	-	-	-	-	-	-	-	5.6	3.3

Notes:

(a) Recent information indicates these values may be approximately 6% too high because of uranium interference in analytical method.

(b) Value questionable.

(c) Stripped with 0.01N HNO_3.

Summary of Second Cycle Runs
Flowsheet Conditions as Noted in Accompanying Figure

	Inactive			10% Clinton Activity		Full Clinton Activity	
Run	2D	3D	4D	1X	2X	3X	4X
Feed							
Al, M[a]	1.30	1.25	1.42	1.54	1.49	1.46-	1.35
Free Base, N	0.025	-	0.03	0.056[b]	0.050[b]	0.044	0.018
pH	1.85	1.74	1.55	0.45	0.72	1.57	1.53
U Loss, %							
Extraction	0.05	0.003	0.02	0.0045	0.00065	0.0015	0.0007
Stripping	0.009	0.005	0.014	0.0083	0.0042	0.0022	0.0080
Feed (~ 34 g U/l cts/min/ml							
Beta	-	-	-	2.17×10^3	3.32×10^3	4.6×10^4	3.06×10^4
Gamma	-	-	-	17	16	188	165
Product (~ 50 g U/l cts/min/ml							
Beta	-	-	-	367[c]	350[c]	560[c]	510[c]
Gamma	-	-	-	2	5.2	7	2.7

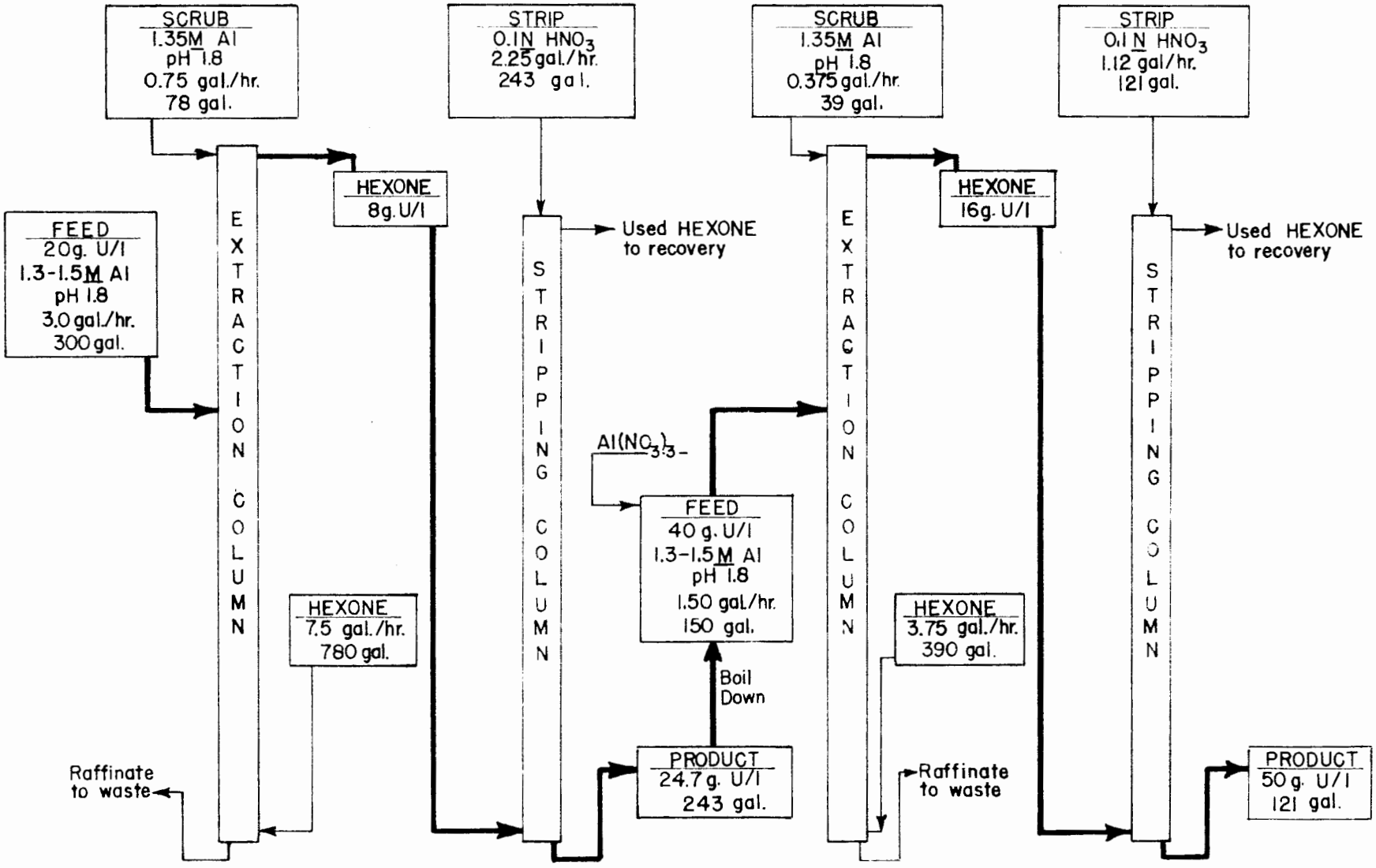
Notes:

(a) Recent information indicates these values may be approximately 9% too high because of uranium interference in analytical method.

(b) Free acid.

(c) These values to be compared with extracted unirradiated uranium of approximately the same age for which the beta rate is 350-500 cts/min/ml.

[redacted]



1200 AREA PILOT PLANT FLOWSHEET
2-15-48

FIGURE 3

Drawing # 5415

[redacted]

sibility of aluminum hydroxide precipitation. Sufficient time is allowed for equilibrium conditions to be established before analyzing and feeding the solution to the columns. The pH of the feed solutions requires 24 hours to reach equilibrium because of strong buffering action. The use of pH measurement as a process control is still under investigation. Acidity and alkalinity are determined by direct titration.

Crud Removal Filtration

The feed solution is filtered through a sintered stainless steel disc of porosity G having an area of 240 sq.in. without filter aid. An accumulation of solids builds up on the filter disc during filtration which is dissolved twice during each run with 5 gallon washes of 20% NaOH. The NaOH insoluble material built up in each run so that at the end of the 6th run when 1500 gallons of solution had been passed through the disc, washing with the NaOH wash did not improve filtration rates suitably. This necessitated the removal and disposal of the expendable filter disc element. During initial startup, tests were made on the equipment with 1 1/2 grams of MnO_2/liter co-precipitate in the column feed solution. Slow filtration rates and high uranium losses caused this procedure to be discontinued temporarily. Additional information will be obtained using MnO_2 as a filter aid in the coming period with particular emphasis on operability and decontamination factors.

Extraction and Stripping, First Cycle

Three 100-hour runs through two cycles of extraction and stripping columns using unirradiated uranium were made to test the operability of the columns. In these runs the flow measurement, column control, and analytical techniques were tested and improved. The data obtained during these runs show that the flows can be maintained by setting the column feed pumps to within 1% of the desired reading with maximum flow variation at any setting of 4% and an average deviation of 2%. A large amount of manpower was expended on the orifice type flow recorders with the result that they are moderately satisfactory in recording but as yet the readings are not entirely reproducible. The chemical data obtained during these runs show that satisfactorily low uranium losses can be obtained with 1.4M aluminum nitrate at .05N basicity with a flow ratio of organic to aqueous of 2.

The six subsequent active runs through two cycles of columns have shown that the total extraction and stripping uranium losses can be kept below .02% through all four columns. Four of these runs were made at full level Clinton activity which processed material having 2.1×10^9 beta counts/min/g U and 4.5×10^6 gamma cts/min/g U. Decontamination factors determined by counting techniques for the first cycle extraction columns were 3×10^3 for beta and 1.6×10^3 for gamma at the flowsheet conditions shown in Fig. 3 which had no oxidants, reductants, or complexing agents. The Ru decontamination factor under these conditions was 200, Zr was 1.77×10^4 and Pu was 4. Specific fission product analyses substantiated the fact that because the Ru activity in the metal processed was approximately 4.6% and the decontamination factor of only 200 was obtained, the major portion of the activity coming through the first cycle column was

Ru. This indicates that the total beta decontamination factor appears to depend almost completely on the Ru decontamination factor as previously concluded from semi-works scale experiments. Decontamination data show that there is a small trend toward higher decontamination as the salt concentration is lowered from 1.41 to 1.36. This also would have been predicted from the earlier semi-works experience.

The decontamination factors in the first cycle stripping column ranged between 1.2 to 1.7, substantiating previous data that essentially all activity is removed from the hexone at this stage. The losses in the stripping column for four runs averaged 0.0011% when 0.1N nitric acid was used for stripping, which increased to average 0.01% for three runs when the acid was reduced to 0.01N. This reduction in nitric acid concentration to the first cycle stripping column was necessary to eliminate the addition of ammonium hydroxide during the second cycle feed makeup. Stripping at a concentration of .03N nitric acid will be made during the coming period to determine losses at intermediate conditions.

Crud Formation in Columns

The operation of the columns has been quite satisfactory with no evidence of crud formation. At the end of the inactive runs, all columns had 350 hour-periods of operation and were dismantled for careful inspection of the packing. There was no sign of plugging, crud, or deposits of any sort on the packing. It appeared to be as shiny as when first put into the columns. The columns have operated a total of 700 hours processing active feed and no evidence of channeling or plugging has appeared. This absence of crud in the columns, it is felt, is due to the extreme care with which all process solutions are handled; all are filtered before entering the columns. As an added precaution, the solvent is contacted with scrub in a column packed with 3/16" Fenske rings just before entering the process. The accumulation of crud in this column is appreciable, requiring cleaning about every 4 - 6 days. The pneumatically operated specific gravity controller continued to give excellent remote control of the column interfaces.

Feed Preparation, Second Cycle

The second cycle feed is made up by butting the concentrate of the entire first cycle product stream. This step was greatly improved by the reduction in the nitric acid content of the strip in the first cycle. Uranium losses first measured were 0.05% during the concentration and were reduced to 0.03% by the installment of baffles in the vapor line. These small losses can be reduced in the 1200 Area by more suitable design of the plant equipment. There is some evidence that a slight decontamination factor is obtained during the concentration operation, but as yet the activity differential is so low that the evidence is only a trend.

Second Cycle Extraction Results

The beta activity in the second cycle product is below uranium background by a factor of 10 because of the separation of the uranium decay daughters from the parents by the extraction process. Inasmuch as the solution samples are counted about 4 days after leaving the column, a large, but as yet unknown, fraction of the counts is due to the uranium background which makes calculation of the second cycle decontamination factors unreliable. It is estimated, however, that the beta decontamination factor is at least 100. The gamma decontamination factors cannot be measured accurately with existing equipment due to the very low level of activity of 5-10 cts/min/ml in the product stream. More information on the second cycle decontamination factors can be obtained when Hanford irradiated uranium is to be processed in an estimated three months.

Feed Pumps

A test has been made of feeding the columns by the use of a liquid piston pump in which the check valves were the only working portions of the pump exposed to the process solutions. The pump gave only partial satisfaction inasmuch as the weak portion of the pump, the check valves, are still in contact with the active solutions and the improvement in maintenance was only minor. The metering of the flow of active solutions continues to be a problem.

HETS Values

Extraction HETS values have been calculated for both the 2" and 3" columns packed with 1/4" x 3/8" Raschig rings. The calculations show that the HETS value is 2.46 feet with a probable error of 0.14 feet. This HETS value does not vary appreciably in successive runs in the same column or between the 2" and 3" columns.

Waste Disposal

The wastes from the columns are disposed of by neutralizing the extract raffinates containing principally aluminum nitrate, fission products, and a small amount of hexone by adding sufficient caustic to give a basic solution. The uranium is disposed of through the standard medium of complexing with soda ash. No trouble has been experienced with any of these steps as long as an excess of caustic or soda ash is maintained.

Solvent Treatment and Recovery

Pre-treatment of commercial hexone consists of washing with sodium dichromate and batch distilling from a caustic heel. This gives a product with a reducing power of 0.004N and an acidity of 0.004N in

acid as acetic acid. The recovery of the hexone was first done batch-wise from a caustic heel which yielded slightly better material. The equipment was later changed to a continuous type distillation, plus an installation of a packed section for reducing entrainment and has yielded a product averaging 0.002N in reducing power and 0.002N in acidity.

The residual activity in the used hexone has been mainly beta and has given no health hazard difficulties during the solvent recovery operation. The used solvent reservoir has had an increase in activity from mixing the various types of hexone, resulting in material with 660 beta cts/min/ml. This activity was reduced by a factor of 6 by remote control washing with a 1/20 volume of 10% caustic.

Program

The program for the coming period includes investigation of various reducing agents and complexants on the effect of fission product and Pu decontamination factor and uranium recovery. In the next period it is hoped that the flowsheet will be sufficiently established in generalities to permit investigation of minor process questions, which is scheduled to start using Hanford material on approximately May 1, 1948. Among the various materials to be tested are sulphamic acid and ferrous ammonium sulphate as reductants; 2-Hexyl-Pyridine as a uranium complexant; and diphenyl-thio-urea as a scrub in the stripping column for improving Pu decontamination.

4. 23 Separations Process Design and Development

4.1 Design Progress (J. F. Huffman)

Design work has been concerned chiefly with the development of general information for the 1300 area and of supporting information for the selection of a 23 process. Since a 23 process has not been definitely selected as yet, no detailed process studies or equipment flowsheets have been made. This work must be done before a process design report is prepared. Detailed information concerning the 1300 area has been presented in nineteen Technical Division drawings and twelve memoranda to the 12-1300 area Design File. Information about the auxiliaries and services which are shared with either the 1200 area or the entire 1000 Project is given in the drawings and memoranda concerning the 1200 area.

The maximum expected delivery of irradiated thorium from the Research Pile to the 1300 area is 8 kg. per day, and it is anticipated that the thorium deliveries from other piles, if any, will be about the same. Consequently, the design capacity of the 1300 area has been set at 15 kg. per day. A typical delivery of thorium will contain about 75 g of 23 and 13 and 2-1/4 g of fission products per 50 kg of irradiated thorium. The shortest cooling period considered at the present time is eight months. With this cooling period about 100 mg of 13 are present, i.e., seven watts of 13 activity: the associated fission product activity is about four watts. The corresponding process requirements are given

in the following tabulation:

Total loss of 23, per cent	0.1
Thorium reduction	10^6
13 decontamination	2×10^5
Fission product decontamination	10^6

The decontamination requirements are designed to make it possible to carry 100 g of 23 product in a container having lead walls one inch thick. This purity appears to be satisfactory for production quantities of 23. Whenever higher purities are needed, the additional purification operations will be made by the investigators concerned.

When the Research Pile is operating at 30,000 kw, the annual production of Pa^{231} is estimated to be less than one gram. Since it appears that Pa^{231} can be produced more efficiently by the irradiation of Th^{230}, it is recommended that no facilities be provided for its recovery. The concentration of U^{232} and U^{234} in the 23 product, based on the production of these isotopes in the Research Pile, will probably be less than one per cent.

The purity of thorium metal used is dictated by the purity requirements of the 23 product and by the neutron economy, especially if breeder piles are involved. A brief study of the irradiation products of the impurities in natural thorium indicates that no serious neutron poisons are produced. In addition, it is anticipated that these impurities and their irradiation products will follow the thorium through the 23 recovery operations and that significant concentrations of them will not appear in the product. In connection with the Research Pile, the presence of 5 ppm of natural uranium in the thorium metal would result in a concentration of about 0.5% U^{236} in the 23 product which is acceptable. In general, the thorium metal produced at Iowa State College from the unextracted nitrate (Lindsay Light and Chemical Company) appears to be satisfactory. Thorium metal from the extracted nitrate might be necessary for breeder piles and for the production of very pure 23.

A batch 23 process using diisopropyl ether has been developed, and the development of a continuous process using either diisopropyl ether, dibutyl cellosolve, or hexone is in progress. Chemical flowsheets for these processes are given in the following drawings.

Technical Division Drawing Numbers	Subject
343, 344, 345, 346, 347	Batch 23 Process using Diisopropyl Ether
353, 354, 355, 356	Continuous 23 Process using Diisopropyl Ether
333, 334	Continuous 23 Process using Dibutyl Cellosolve
349, 350	Continuous 23 Process using Hexone

The batch process consists of five major operations: (1) jacket removal and metal solution; (2) first extraction cycle; (3) scrub recovery cycle; (4) second extraction cycle; and (5) final clean-up operations. The first extraction cycle consists of nine equal-volume extractions, two 1/12 volume scrubs, and four 1/4 volume strippings operations. The recovery of 23 from the scrub solution is effected with about eight equal-volume extractions and four 1/4 volume strips. The principal difference between the batch and continuous processes centers around the first extraction and scrub cycles. With the continuous process the extraction, scrubbing, and stripping operations are accomplished with packed columns. The height of the extraction-scrubbing and stripping columns will probably be 35-40 and 18-20 feet, respectively. Both processes require two cycles to effect the desired decontamination, and in each case the proposed second cycle is a batch operation using diisopropyl ether.

Three classes of waste solutions, low, intermediate and high activity, are associated with each process. The 1300 area waste disposal system has been combined with those for the 1100 and 1200 areas. A discussion of the combined waste disposal system is presented in connection with the report concerning the 1200 area. In general, the low activity wastes are collected, analyzed for uranium and activity and discharged to the retention pond; the intermediate and high activity wastes are usually neutralized, concentrated, stored for decay and eventually discarded.

All of the operations in connection with the recovery and decontamination of 23 are carried out in the 1305 building. Proposed layouts of the 1305 building for the batch and continuous processes have been presented in TD-428 and TD-425, respectively. The overall length of the building is 220 feet for the batch and 200 feet for the continuous process. The overall width and height are 58 and 48 feet, respectively. The estimated investment costs of the 1305 building and its associated equipment for each process is given in the following tabulation.

	Batch	Continuous
Building	$ 668,000	$ 650,000
Equipment	950,000	850,000
	$1,618,000	$1,500,000

Since these cost estimates may be in error by more than $100,000, the apparent differences are not significant.

The auxiliaries and services required by the 23 process can be divided into two classes: (1) those which are shared with the 1200 area and (2) those which are shared with the entire 1000 Project. A preliminary survey of the requirements for each process indicates that the differences are not significant. The allocated investment cost of the auxiliaries and services which are shared with the 1200 area is about $1,250,000. The allocated investment cost of those which are shared with the entire 1000 Project is about $1,000,000. This figure includes a share of all general charges by the contractor for the 1000 Project. Since these investment costs are about 50% greater than those for the 1305 building and its associated equipment, the apparent differences mentioned in the preceeding paragraph are even less significant.

A detailed investigation of all operating costs associated with 23 recovery has not been made; however, it appears reasonable to assume that the operating costs in connection with the auxiliaries and services are the same irrespective of whether the batch or continuous process is used. The number of man hours of labor required with both processes differs somewhat; however, since only a small number of operators is necessary in the 1305 building at any given time for either process, practical considerations require a staff of about nineteen operators for each process. The estimated annual cost of process chemicals is given in the following tabulation.

Batch Process using Diisopropyl Ether	$15,500
Continuous Process using Diisopropyl Ether	4,600
Continuous Process using Dibutyl Cellosolve	16,400
Continuous Process using Hexone	6,200

The cost of chemicals used in connection with solvent pretreatment and recovery is a minor item in each case, probably less than $100 per year. It is difficult to determine the cost of chemicals for decontamination, but a rough estimate indicates that the annual cost for each process may be about $30,000. In general, it appears that there are no major differences in operating costs with either the batch or the continuous process.

Since estimated costs for the batch and continuous processes are essentially equal, the choice between them must be based on process considerations such as reliability, flexibility and future possibilities. Preliminary results at X-10 and elsewhere on the project indicate that the desired recovery and decontamination can probably be obtained with either process.

In general, it is likely that the continuous process will be more satisfactory over a long period of time.

4.2 Laboratory and Semi-Works "23" Development

4.21 General

The "23" process for separation of U^{233} from thorium, fission products, and Pa^{233} includes the following steps: dissolving the thorium metal; filtering, if necessary, to remove insoluble materials; a batch or continuous solvent extraction cycle (extracting the 23 into diisopropyl ether or dibutyl cellosolve, or possibly hexone, scrubbing the extract to remove part of the contaminants, and stripping the 23 into water) in which a separation of 23 from the contaminants by a factor of at least 10^3 is obtained. To this point the process is carried out behind heavy shielding in a canyon. The partially decontaminated 23 in the stripping solution is then removed to a laboratory where, in a diisopropyl ether extraction cycle behind thin shielding, the separation from the contaminants is completed. The purified 23 is then to be precipitated as ammonium diurenate and ignited to U_3O_8.

In the semi-works, a batch extraction process has been shown to give adequate decontamination with 23 losses less than the maximum permissible of 0.1%. Investigations toward making the canyon solvent extraction cycle continuous using as the solvent either diisopropyl ether or dibutyl cellosolve have been going on for about six months. Both of these solvents give more than adequate decontamination and reduce 23 losses to less than 0.01% in columns about 20 feet long which are packed with 3/16" helices. However, with either solvent the columns become plugged after a short period of operation (100 hours or less) because an insoluble material ("crud") deposits in the Fenske packing, despite filtration and prescrubbing of feeds. Preliminary work with larger Raschig ring packing, which is much less likely to become plugged than is Fenske packing because of its larger voids, show it to be usable with either solvent from the standpoint of producing the required 23 recovery in reasonable height columns, calculated extraction heights of from 25 to 40 feet being required. Therefore, work of the next quarter will be to demonstrate satisfactory operation of columns packed with Raschig rings.

4.22 Laboratory Continuous Column Extraction Studies

The problem of crud deposition in columns has been studied in the laboratory during the past quarter from two points of view: (1) prevention of its formation, and (2) substitution of larger packing which will allow crud to pass through the column. The latter was successfully resorted to in the 25 process.

Use of Filtered and Pre-Scrubbed Feeds to Prevent Crud Formation

The methods tried for preventing crud deposition in 3/16" Fenske packing have been filtration of all feeds and pre-scrubbing of all feeds in short, packed pre-scrubbers in the hope that crud will form in them rather than in the column. With filtered feeds and dibutyl cellosolve solvent, sufficient crud still collected in 90 hours of operation at near flooding flowrates to plug the column. Somewhat longer operation, but undoubtedly still far short of a practical time, would be expected at lower flow rates. In a run using Fenske packing with diisopropyl ether, in which both filtered and pre-scrubbed feeds were used, deposition of a gelatinous crud just above the feed plate occurred after 30 hours of operation. This work clearly indicates that Fenske packing is unsatisfactory for plant scale operation. The main constituents of this crud were aluminum, silicon, magnesium, and considerable amounts of the components of stainless steel (iron, chromium, nickel, and manganese).

Use of Raschig Ring Packing (1/4 x 3/8-inch)

This packing has been investigated to determine if adequate 23 recovery can be achieved in extraction sections of reasonable height (40' or less), and its value in eliminating crud deposition has not yet been tested. In 1-1/2-inch Pyrex glass columns, the 23 HETS was found to increase gradually from 5.1 to 8.6 ft. for either dibutyl cellosolve or diisopropyl ether as the total throughput was increased from 5 to 80% of the flooding throughput (600 gal/hr/sq. ft. for the dibutyl cellosolve

KEUFFEL & ESSER CO., N. Y. NO. 359-11
10 × 10 to the ½ inch. 5th lines accented.
Engraving 7 × 10 in.
MADE IN U.S.A.

Drawing # 5240

SECRET

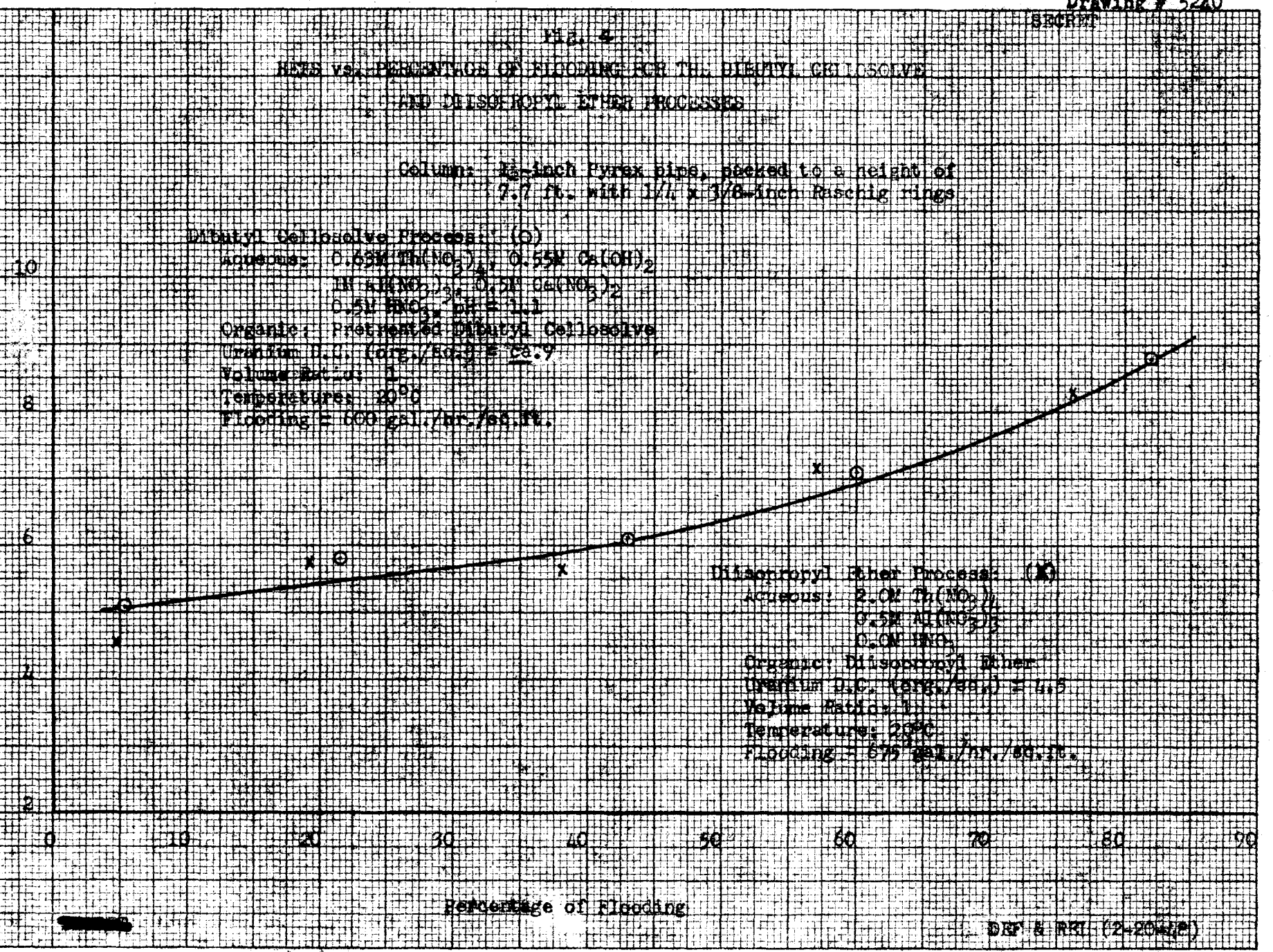
Fig. 4

HETS vs. PERCENTAGE OF FLOODING FOR THE DIBUTYL CELLOSOLVE AND DIISOPROPYL ETHER PROCESSES

Column: [redacted]-inch Pyrex pipe, packed to a height of 7.7 ft. with 1/4 x 3/8-inch Raschig rings

Dibutyl Cellosolve Process: (O)
Aqueous: 0.63M $Th(NO_3)_4$, 0.55M $Ca(OH)_2$
1M $Al(NO_3)_3$, 0.5M $Ca(NO_3)_2$
0.5M HNO_3, pH = 1.1
Organic: Pretreated Dibutyl Cellosolve
Uranium D.C. (grs./sq.ft) = [redacted].9
Volume Ratio: 1
Temperature: 20°C
Flooding = 600 gal./hr./sq.ft.

Diisopropyl Ether Process: (X)
Aqueous: 2.0M $Th(NO_3)_4$
0.5M $Al(NO_3)_3$
0.0M HNO_3
Organic: Diisopropyl Ether
Uranium D.C. (grs./sq.ft) = 4.5
Volume Ratio: 1
Temperature: 20°C
Flooding = 675 gal./hr./sq.ft.



[redacted]

DEF & RKI (12-20-46)

system and 675 for the diisopropyl ether system) (see Fig. 4). It is recommended that columns be designed for operation at a total throughput which is less than 50% of the flooding throughput, corresponding to a maximum HETS of 6.4 ft. The high uranium distribution coefficient (9--org./aq.) present in the dibutyl cellosolve runs cannot be consistently obtained in all runs, occasionally dropping under apparently identical solution conditions to as low as 5.5. Therefore, for safety reasons it would now be necessary to prescribe extractions sections of about 35 to 40 ft. for either solvent for operation at less than 50% of the flooding throughput. Next quarter, it is planned to make runs with both solvents to demonstrate satisfactory column operation for long periods of time using the Raschig ring packing.

4.23 Laboratory Decontamination and Distribution Studies

The effects of $Th(NO_3)_4$ concentration and HNO_3 acidity on 23 extraction and decontamination using dibutyl cellosolve have been studied in the laboratory and extraction conditions outlined for a process which does not contain an MnO_2 precipitation step. The inclusion of the MnO_2 step to aid in crud removal or to improve decontamination from protoactinium makes necessary very complex extraction conditions. This laboratory study disclosed that the 23-fission product separation is very dependent on pH but essentially independent of the $Th(NO_3)_4$ concentration. The optimum pH range for high decontamination and 23 recovery was determined to be 0.8 to 1.0 (see Fig. 5). The following are the recommended salt and acid conditions for the feed and scrub:

Feed: 2.0M $Th(NO_3)_4$
0.1 to 0.3M in free HNO_3

Scrub: 2.4M $Al(NO_3)_3$ at a pH of 1.8 (0.85M deficient in HNO_3)

The required ratio of scrub to feed is between 0.3 and 0.4 and produces an extraction aqueous phase of the following approximate composition: 1.6M $Th(NO_3)_4$, 0.55M $Al(NO_3)_3$, and a 0.05M HNO_3 deficiency (pH = 0.8 to 1.0). The ratio of aqueous to solvent in the extraction section is one. Under these conditions, the uranium distribution coefficients in extraction is at least 8 in favor of the cellosolve, the beta fission product decontamination factor 1×10^4, and the amount of 23 recycled in scrubbing about 5%. These conditions will soon be tried in continuous column runs.

4.24 Laboratory Purification of 23

Using batch diisopropyl ether extraction, a total of 6350 mg. of 23 was isolated and concentrated from strip product solutions during the past quarter. Of this total, 6080 mg. was concentrated with a loss of 0.036% from the product of the Chemistry Division's dibutyl cellosolve columns and 270 mg. was recovered with a loss of 0.072% from the strip solution of the Technical Division laboratory columns. These losses are much higher than the usual 0.002% losses because of the formation of large amounts of crud in extraction and the presence of oily organic decomposition products.

An alternate laboratory clean-up cycle employing TTA (thenoyl trifluoro acetone) complexing and extraction into benzene appears promising for either 23 or 25 clean-up since laboratory results show a ruthenium decontamination factor of 60-100 with a uranium distribution coefficient of 20-25 in favor of the organic (benzene, 4.0M in TTA). Stripping the uranium from the extract with water is difficult unless the extract is diluted 6-fold with benzene. Attempts are now being made in the laboratory to adapt the TTA process to packed column extraction.

Laboratory work on final 23 or 25 purification methods was curtailed during the quarter by the installation of three stainless steel canopy hoods.

42.5 "23" Equipment Design

Design work for equipment for the 23 pilot plant to be located in the proposed new solvent building, 706-HB, was started during the quarter by the Process Design Section.

5. Pile Metallurgical Development

5.1 Fabrication of Uranium-Aluminum Alloy

Melting and Casting

Activity in this field has largely been devoted to production of slugs containing enriched uranium for radiation at Hanford. The melting and casting part of this operation is completed for the material available. Twelve remelt heats have been returned to Y-12 for separation of the uranium due to the inadvertent use of Al-Si alloy instead of high purity aluminum in making up of these slugs.

The average recovery of these reduction heats was 98.3%. Seven analyses have been received on remelt heats. The compositions are within specifications and average 4.30% uranium.

Rolling and Cladding

Difficulties have been encountered with blistering of the 2S aluminum covering the sandwiches. This occurs mostly at the ends of the cores and is due to poorly fitting frames. The most recent work has been directed toward elimination of the use of frames. Several acceptable sheets have been produced in this way and certain conclusions can be drawn. 1) A reduction of at least 5 to 1 is necessary to bond edges and ends completely, and 2) beveling the corners of the cores eliminates the corner blisters in the final sheet.

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Drawing # 5156

FIG. 5

EFFECT OF pH ON "23" EXTRACTION WITH DIBUTYL CELLOSOLVE

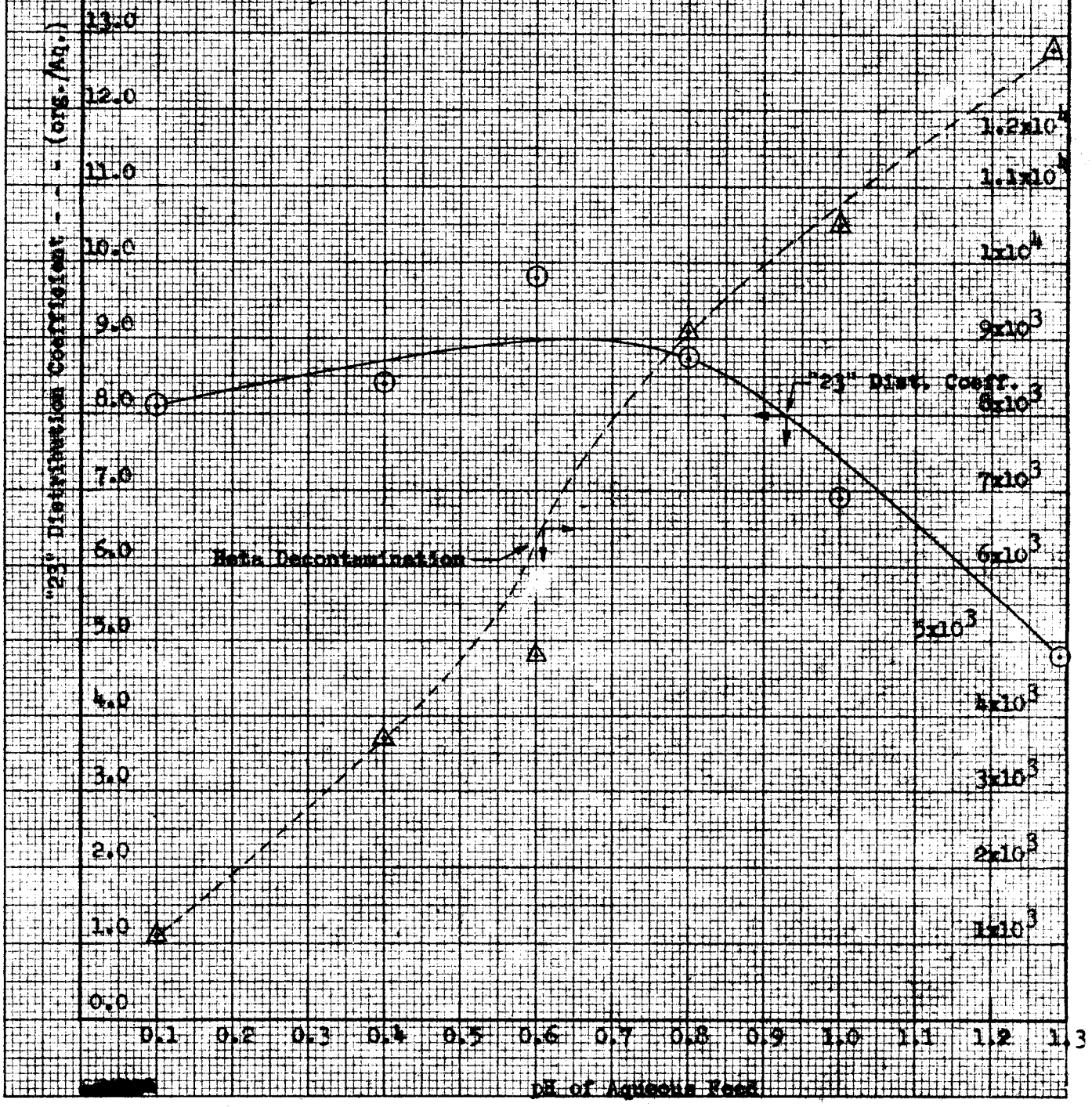
Conditions:

Aqueous feed: 2M $Th(NO_3)_4$
Ca. 0.05 M Uranium
Variable pH

Organic feed: Dibutyl Cellosolve (1 equal-volume pass made)

Temperature : Room Temperature

Activity : 1-slug, bombarded 3 yr., cooled 3 mo.



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10 × 10 to the ½ inch, 5th lines accented.
Engraving, 7 × 10 in.
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5.2 Fabrication of Uranium-Aluminum Fuel Rods

Jigging for Brazing

The problem of devising suitable jigs for use in furnace brazing of the active assemblies has been investigated further. Graphite has been found previously to be satisfactory, except that it is attacked by oxidation. Coatings of various phosphates have been found to give some protection to the graphite, but tend in turn to be attacked by the brazing fluxes used. Within the quarter just completed it has been found possible to substitute a Mg-Al silicate material known commercially as "Lava" for the graphite. This material is much more stable than the graphite with respect to attack by flux or air. The design of the jog has had to be changed so that it now consists of two slabs of Lava resting on an evenly machined third slab also of Lava. Further work on this technique is planned.

Flux

Efforts have been made to find a brazing flux which does not contain strongly neutron absorbing elements such as lithium or chlorine. During the quarter, two lithium-free brazing fluxes said to be suitable for furnace brazing aluminum have been received from commercial sources. Unfortunately, neither is satisfactory. One of the fluxes was useful for torch brazing, but the other was useless even for this application.

Two lithium-free brazing fluxes, said to be suitable for furnace brazing aluminum, have been received from two commercial sources. For more than a year, both suppliers have maintained that such a material had not as yet been made. Unfortunately, neither shows even the least promise for furnace brazing. We have been successful in torch brazing with the aid of one of these and have failed with the other.

Dip Brazing

A tank for dip brazing has been fabricated from the corrosion resistant alloy, Hastelloy C. This tank is now being built into the dip brazing unit previously provided with a graphite flux tank. Charging arrangements have been provided which will permit controlled immersion and removal of assemblies from molten flux.

5.3 Fabrication of Control Rod Units

Alclad Thorium

Work on the Al cladding of thorium has continued. One of the difficulties is caused by the difference in the plastic constants of the two metals. Any appreciable reduction by rolling of sandwiches results in a thick edged core and distorted aluminum surface. To avoid this, the thorium will have to be rolled to approximately the final core thickness before aluminizing and cladding. This difference in properties

is also apparent in the curving of the clad plate as fracture of the aluminum tends to occur at the core edge. This tendency is decreased somewhat by using a tapered edge.

Another peculiarity of alclad thorium is the existence of a thin brittle compound layer at the interface. The brittleness of this layer may not be undesirable if the cladding is perfect, but if any void exists there is a tendency for separation along this layer. This property becomes apparent when rolled plates are cut open.

Various intermediate coatings have been applied in an effort to eliminate the formation of the brittle layer. With Al-Si eutectic coatings no improvement was observed. Copper coatings oxidized excessively in the rolling preheat and the subsequent adhesion was poor. These coatings were applied by electroplating and by dipping in molten NaCl-CuCl. The first of these methods worked better. Dipping in molten Cu was tried but this resulted in rapid solution of thorium in the copper.

Thorium was also coated with various Cu-Al alloys by dipping. However, the clad samples still showed the presence of a brittle alloy.

Tests were made with zinc-coated thorium produced by dipping into molten Eutectic No. 190 flux. These did not bond well due probably to excessive oxidation of the zinc despite low rolling temperatures.

Additional tests are planned with silver coated thorium.

Several other experiments in cladding were tried. In one of these the thorium was perforated before aluminum dipping. This provides additional anchorage at the perforations. In another, cladding was tried without the use of a frame around the core. This was not very successful.

In some of the experiments, laminations in the thorium were revealed. These should be eliminated by improved techniques in production of thorium sheet.

If it will not be possible to eliminate the brittle layer, other methods may be developed. Perforation of the thorium seems beneficial. Another possibility may be in the use of powder metallurgy in which thorium granules would be embedded in an aluminum matrix.

Alternate Design of Thorium Unit

An aluminum mock up is to be brazed shortly to demonstrate whether an assembly of flat thorium sandwiches can be brazed to curved side plates to make a unit suitable for the control rods.

5.4 Fabrication of Beryllium

Mechanically Joined Assembly

A beryllium assembly in which the slabs have been bolted together with beryllium bolts has been fabricated and end boxes attached. This assembly has been measured and found to be reasonably close to the design

tolerances. It is ready for functional tests, after which it will be again inspected dimensionally. Difficulties in machining these beryllium pieces were due in some measure to inadequate capacity of machine tools. When more adequate tools are available, a complete report can be written covering optimum machining technique. This method of joining is of special interest in that it avoids or reduces the seriousness of several problems appearing in the brazing of assemblies, such as spacing, alignment, and flux entrappment. Less machining is required for this method of assembly than for joining by brazing. The problem of threading the slabs (female) and the beryllium bolts (male) is not nearly as serious as the problems which are peculiar to the brazing method.

The successful fabrication of this, the first full-sized beryllium assembly, shows clearly the feasibility of this method for fabrication of all the beryllium.

Assembly by Brazing

The pot furnace installation is now complete and work has been started on the dip-brazing of larger sections of beryllium. The first trials involved the use of Al-Si alloy covered with Airco flux. Tests on the removal of aluminum from the holes in the beryllium show promise. To accomplish this, the excess aluminum is pushed out by means of rods immediately after removal from the bath. A 3" x 6" x 5", five-sectioned unit has now been processed.

Some difficulty is being experienced in flux seepage through the glazed tercod crucible. This is obviously undesirable and may necessitate the use of some other material as a container.

Samples of beryllium sprayed with Al-Si eutectic alloy were received recently. The coating adheres strongly but can be separated mechanically as there is no metallurgical bonding. Attempts were made to join slabs by application of heat and moderate pressure both in air and in a Pb bath. The latter method was the better of the two, but is not satisfactory as the presence of oxide in the coating interferes with the alloying process.

Machining of Beryllium

Trials were made in the deep drilling of beryllium with varied success. An 11/64" hole was drilled through an 6" length using high speed, slow feed and no lubrication. Difficulty is caused by fluting wear, seizing and breaking of the drills. One carboloy drill was tried unsuccessfully but others are now here. As the length:diameter ratio is large, alignment is difficult.

The new hydraulic feed rate control method developed at Farmingdale Aircraftsman Corporation is being investigated.

It has been noted earlier that thin abrasive discs cut beryllium better than thick discs. In the use of these, moderate pressure is

recommended to avoid disc deformation which causes non-linear cutting and disc breakage.

In the cutting of thin beryllium sections, lateral support minimizes warping and cracking of the metal.

Strength Characteristics of Beryllium

Thermal cracking tests were made by the induction heating of centrally water-cooled beryllium cylinders. The results indicate that the extruded beryllium has adequate lateral strength for use in the new pile. The test conditions were more than twice as severe as expected in the pile, but no cracking resulted.

Adequate strength was also shown in the test which involved the pressing of thin transverse sections between surfaces having known radii of curvature.

Acceptance Testing

Some of the beryllium extrusions received recently were found to be unsatisfactory because of the presence of thin longitudinal inclusions which produce a fibrous fracture. These can be detected by end-cropping and deep etching with dilute sulfuric acid. They can also be detected by fracturing transverse sections or by heating to a red heat and quenching.

5.5 Inspection

Uranium-Aluminum Alloy Sandwiches

A sandwich in which the core was brazed to the ends of the frame prior to rolling for cladding was received from the rolling mill. Annealing showed blisters at each of three corners. Metallographic examination showed a rounding of the lateral core edge and good bonding. A section through one of the blisters revealed porous core alloy. This may explain the usual occurrence of blistering on both sides, within the core area, and the presence of such blisters may be more properly attributed to the core alloy condition at that point than to rolling technique. Future examination of such blisters will include more detailed examination of core alloy in the vicinity of the blister.

Several U-Al alloy sandwiches were received from the rolling mill in which no frames were used. The initial core and cladding thicknesses were each 1/16" instead of the previously used 1/8". The rolling direction was reversed after each pass to minimize end bulging.

The usual anneal produced no blisters. The edges, however, were not well bonded and could be separated mechanically to about 1/2" within the core area. The central portion was well bonded. Prolonged annealing did not improve the edge bonding. Samples examined metallographically after dilute caustic etching appeared sound. However, when strong caustic pickling was tried the line of separation was revealed in the aluminum. Another test

which might be termed semi-destructive involves bending and straightening the plate causing edge separation in poorly bonded plates. Intermediate initial thicknesses will be tried to obtain better edge bonding.

It has been found that surface irregularities can be detected more readily if a Dykem coating is applied. This coating is dark blue in color and has a glossy finish, producing greater contrast between the principal and secondary reflected rays at regions where defects occur. Projections much less than .0005" in height can be detected in this manner.

X-Ray Inspection

The equipment for this work has been temporarily set up and protected with lead to bring radiation outside the cabinet to tolerance levels. The equipment has been used to locate cores in clad U-Al sandwiches, and study the segregation and shrinkage cavities in U-Al alloy ingots.

Exposure techniques have been developed for U-Al alloy, beryllium, and U-Al alloy sandwiches.

Dimensional Inspection

All brazed assemblies of fuel or control rod designs are being inspected as a routine matter and the records of these results, when available in sufficient numbers can be used to determine probable deviation from mean dimensions, number of rejections to be expected for a given set of tolerances, or the tolerances that will produce a given percentage of acceptable assemblies.

Miscellaneous Tests

A small amount of work has been done on Bi-Al bonding as a result of Spinrad's interest in Bi as a reflector material. These two metals are insoluble but can be made to bond by ternary alloy formation. One such alloy is the Bi-Zn-Al ternary. Bonding of this type can be obtained through the use of Alcoa X-62 flux which deposits Zn on the Al. This flux, however, contains boron, which would be undesirable if flux inclusions were present. If further work is contemplated, better fluxes could be formulated.

6. Miscellaneous

6.1 Heat Transfer Research (R. N. Lyon)

As indicated at the end of the last quarter, the liquid metal research has been divided into three categories: 1) Theoretical heat transfer considerations, 2) Experimental determination of heat transfer, and 3) Testing of liquid metal handling techniques.

Experimental and Handling

The testing of handling techniques has been limited during this quarter to experience in cleaning NaK out of the plugged natural convection system, and to transferring NaK from one vessel to another. The use of low pressure steam for removing the alloy from walls of equipment, as suggested by other handlers, has been found quite safe and easy. Wet steam caused no difficulty, as long as the flow was rapid enough to exclude air and to carry away the hydrogen which was formed. A quiet flame, presumably of hydrogen, was observed at the outlet of equipment for a short time after the steaming operation was begun.

Transfer from one tank to another was easily accomplished using copper tubing and dry argon under pressure. The tank level indicator operated perfectly, and no plugging of the gas bleed line to the alloy outlet pipe occurred.

Most of the major pieces for the experimental equipment have been built, and actual installation has been begun.

Theory

The theoretical investigation of heat transfer to liquid metals is now relatively complete. Integration of the general equation has been accomplished using the actual velocity distribution data of Nikuradse, and a simplified approximate heat transfer equation has been worked out for liquid metals. The development of the general equation and the approximate liquid metal equation is carried out in the following manner:

We first establish a definition for the heat transfer coefficient and then proceed to set up a rigorous equation for it in the case where physical properties are unaffected by temperature and where heat flow through the wall surface is uniform around the tube and along its length. Later we consider in a cursory way the possible effects of changes in physical properties along temperature gradients. The development is carried out for the situation far enough from the entrance to have established the ultimate velocity and temperature profiles.

No consideration is given here to the case where the fluid does not wet the wall, although experimental work with mercury at General Electric and elsewhere has indicated that this situation is important.

Definition of h

The heat transfer coefficient, or unit conductance of a flowing system, h, is defined as the heat flux normal to the wall surface, divided by difference in temperature between the wall surface and the flow mean temperature.

$$h \equiv \frac{q_w}{A_w (t_w - t_m)} \qquad (1)$$

q_w is the heat per unit time normal to the wall surface; A_w is the area of the wall surface across which the heat flows; t_w is the temperature of the wall surface; and t_m is the flow mean temperature of the fluid.

The flow mean temperature, sometimes called the mean temperature, the bulk temperature or the mixed mean temperature, is the temperature which would be obtained by cutting off the pipe at the distance along the pipe in question and catching all the flowing fluid in a vessel where it is completely mixed. It is the average weighted by the quantity of fluid flowing at each temperature across the tube.

Our problem now consists of finding the ratios of $\frac{q_w}{A_w}$ to $(t_w - t_m)$ with various fluids flowing at various velocities in various sizes of pipes, where the quantities in question are constant with respect to time, and symmetrical about the central axis.

Determination of $(t_w - t)$

To find h, we must first obtain an expression for $(t_w - t)$ in terms of $\frac{q_w}{A_w}$, where t is the temperature at any distance, r, from the center of the tube. By appropriate integration, the weighted average $(t_w - t_m)$ can then be found.

We start with the equation

$$\frac{dt}{dr} = \frac{q}{A\bar{K}} = \text{rate of temperature increase at distance r from the tube center as one moves away from the center.} \qquad (2)$$

q is the radial heat flow, at distance r, toward the tube center, and A is the area across which the heat is flowing at distance r from the tube center. If we consider, henceforth, only a unit length of tube, A becomes numerically equal to $2\pi r$.

$\bar{K}$ is the total conductivity of the fluid in the direction of heat flow. It includes both molecular conductivity and eddy transfer. It can include any other conduction mechanism which affects heat flow to the first power of the temperature. Radiant transmission which is affected by the temperature to a higher power is assumed to be negligible. $\bar{K}$ is not a constant, but varies with the distance r.

We are now interested in finding an expression for $\frac{q}{A}$ in terms of r or $\frac{r}{r_w}$ where r_w is the radius of the tube.

Since A is numerically equal to $2\pi r$, when a unit length of tube is considered,

$$\frac{A}{A_w} = \frac{r}{r_w}$$

or

$$A = A_w \frac{r}{r_w} \qquad (3)$$

q will be found by summing up the temperature increase per unit length of pipe, $\frac{dt}{dz}$, of all the flow filaments of fluid within the ring of radius r and multiplying by the specific heat, c, and density of the fluid. In addition, we must multiply each filament by its velocity, u, since the velocity determines the relative ability of a given filament to carry heat axially.*

$$q = \int_0^{r_2} 2\pi r_1 \, c\rho \, u_1 \frac{dt}{dz} \, dr_1$$

The subscripts 1 and 2 indicate the order of the limits. That is, r_2 is the limiting value of r_1. Later r_3 will be seen to be the limit of r_2. Numerical subscripts on other variables carry a similar significance. For example, u_1 is velocity as a function of r_1 and if, as occurs below, S is defined as $\frac{r}{r_w}$, $S_2 = \frac{r_2}{r_w}$, or S_2 is the limit of S_1. This nomenclature is resorted to because the development involves three successive integrations all with respect to distance from center of the tube.

$\frac{dt}{dz}$ will be seen to be independent of r, since the temperature profile under our assumptions will not change as one moves along the pipe. When we integrate all the way out to the wall, r_w, we obtain

$$q_w = 2\pi c\rho \frac{dt}{dz} \int_0^{r_w} u_1 \, r \, dr = \pi r_w^2 \, c\rho \, u_m \frac{dt}{dz}$$

---

* Since our assumption of uniform temperature profile along z leads to a constant value of $\frac{dt}{dz}$, it also indicates constant heat flow by conductance for a given filament in the axial direction. A heat balance around a small volume, (dr, dθ, dz) indicates by inspection, therefore, that the net heat introduced by molecular and eddy conductivity along r can only leave as increased heat content of the liquid passing through the volume in the z direction. It is also clear that since we have assumed steady state, all of the temperature rise of the fluid passing through this small volume must be accounted for by heat entering along the radius. No heat is assumed to flow circumferentially, since the system is symmetrical about the z axis. (See Figure 6)

Drawing # 5416

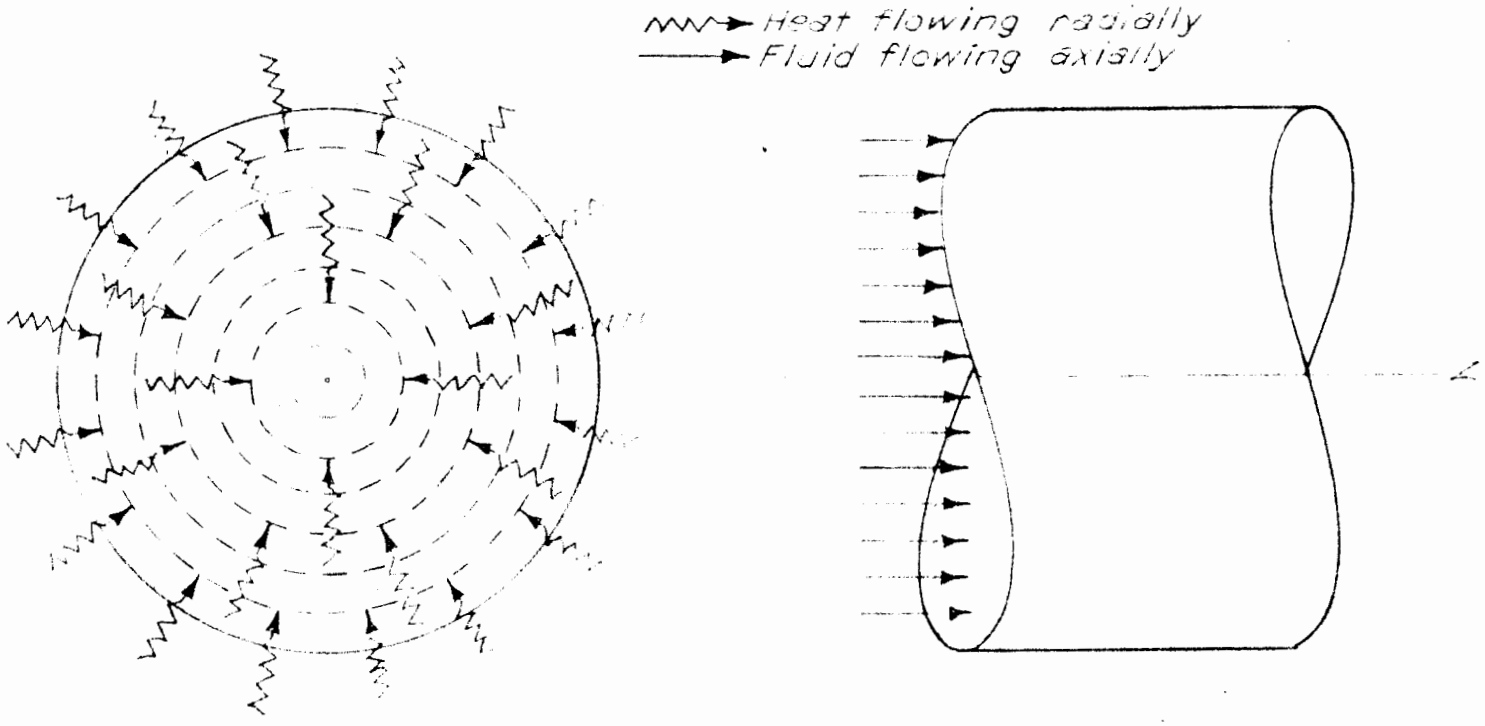


FIGURE 6

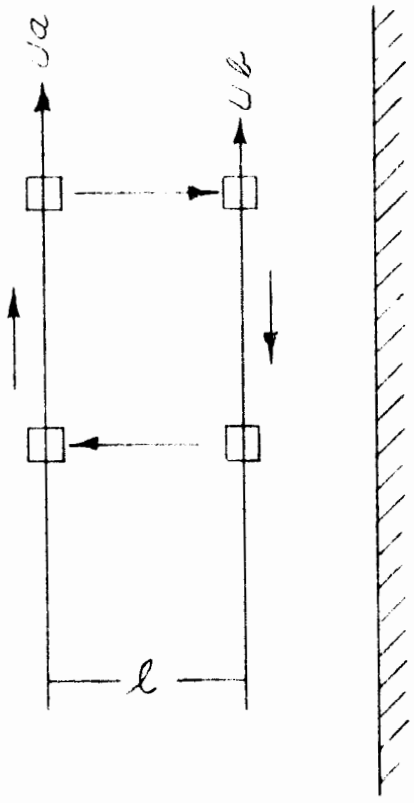


FIGURE 7

In this step we have defined u_m mathematically as

$$u_m = \frac{2\pi \int_0^{r_w} u\, r\, d\, r}{2\pi \int_0^{r_w} r\, d\, r}$$

Physically, u_m is simply the average velocity of the fluid in the tube. The ratio of q to q_w, then, is

$$\frac{q}{q_w} = \frac{2\pi c \rho \frac{dt}{dz} \int_0^{r_2} u_1\, r_1\, d\, r_1}{\pi c \rho \frac{dt}{dz}\, u_m\, r_w^2}$$

$$= 2 \int_0^{r_2/r_w} \frac{u_1}{u_m} \cdot \frac{r_1}{r_w}\, d\, \frac{r_1}{r_w}$$

or

$$q = 2q_w \int_0^{S_2} \frac{u_1}{u_m} S_1\, d\, S_1 \qquad (4)$$

if we define S as $S = \frac{r}{r_w}$.

Substituting equation (4) for q and equation (3) for A in equation (2) for $\frac{dt}{dr}$, we obtain

$$\frac{dt}{dr} = \frac{2q_w \int_0^{S_2} \frac{u_1}{u_m} S_1\, d\, S_1}{A_w\, S_2\, \bar{K}}$$

Integrating both sides of the preceding equation from r_3 to the wall, $r_2 = r_w$ gives us an equation for $t_w - t_3$;

$$t_w - t_3 = 2 \frac{q_w}{A_w} \int_{r_3}^{r_w} \frac{\int_0^{S_2} \frac{u_1}{u_m} S_1 \, d S_1}{S_2 \bar{\bar{K}}} d r_2$$

or (5)

$$t_w - t_3 = D \frac{q_w}{A_w} \int_{S_3}^{1} \frac{\int_0^{S_2} \frac{u_1}{u_m} S_1 \, d S_1}{S_2 \bar{\bar{K}}} d S_2$$

where $D = 2 r_w$ or the diameter of the tube and $S_w = 1$

Determination of h and Nu

$(t_w - t_m)$ can now be found by averaging $(t_w - t_3)$ from the center to the wall using the weight factor $2\pi r_3 u_3$ and integrating with respect to r_3

$$t_w - t_m = D \frac{q_w}{A_w} \frac{\int_0^{r_w} 2\pi u_3 r_3 \int_{S_3}^{1} \frac{\int_0^{S_2} \frac{u_1}{u_m} S_1 \, d S_1}{S_2 \bar{\bar{K}}} d S_2 \, d r_3}{\int_0^{r_w} 2\pi r_3 u_3 \, d r_3}$$

$$= 2D \frac{q_w}{A_w} \int_0^{1} \frac{u_3}{u_m} S_3 \int_{S_3}^{1} \frac{\int_0^{S_2} \frac{u_1}{u_m} S_1 \, d S_1}{S_2 \bar{\bar{K}}} d S_2 \, d S_3$$

If we now substitute into equation (1), we obtain:

$$h = \frac{\frac{q_w}{A_w}}{2D\frac{q_w}{A_w}\int_0^1 \frac{u_3}{u_m} S_3 \int_{S_3}^1 \frac{\int_0^S \frac{u_1}{u_m} S_1 \, d S_1}{S_2 \bar{K}} d S_2 \, d S_3}$$

or

$$\frac{hD}{k} = Nu = \frac{\frac{1}{2}}{\int_0^1 \frac{u_3}{u_m} S_3 \int_{S_3}^1 \frac{k}{S_2 \bar{K}} \int_0^{S_2} \frac{u_1}{u_m} S_1 \, d S_1 \, d S_2 \, d S_3} \qquad (6)$$

Nu is the Nusselt dimentionless modulus defined as: $Nu = \frac{hD}{k}$, and k is the molecular thermal conductivity of the fluid.

Limiting Value of Nu

A number of interesting points are disclosed in equation (6). With it can be shown the attacks made by von Karman and Martinelli. The latter used the approximation that $\frac{u}{u_m} = 1$ over the entire range except for a negligible region near the wall, for the first integration, but apparently not for the third. If we use the assumption $\frac{u}{u_m} = 1$ for both the first and third integration equation (6) reduces to:

$$Nu = \frac{hD}{k} = \frac{\frac{1}{2}}{\int_0^1 S_3 \int_{S_3}^1 \frac{k}{S_2 \bar{K}} \int_0^{S_2} S_1 \, d S_1 \, d S_2 \, d S_3} \qquad (6a)$$

The only problem in solving this equation is the evaluation of $\bar{K}$ the apparent conductivity of the fluid as a function of S, the normalized distance from the center. In most situations which actually arise, $\bar{K}$ will be equal to the molecular conductivity plus the conductivity due to eddying; or $\bar{K} = k + E$ where E is the point value of eddy conductivity in units of heat/(time)(area)(temperature/length).

It is seen that where the fluid has a very high molecular conductivity the apparent conductivity, $\bar{K}$, approaches k, and equation (6a) may be simplified still further to give:

$$\frac{hD}{k} = \frac{\frac{1}{2}}{\int_0^1 S_3 \int_{S_3}^1 \frac{1}{S_2} \int_0^{S_2} S_1 \, d S_1 \, d S_2 \, d S_3} \tag{6b}$$

The integrations in this equation can be carried out very simply as shown:

$$\frac{hD}{k} = \frac{\frac{1}{2}}{\int_0^1 S_3 \int_{S_3}^1 \frac{1}{S_2} \cdot \frac{S_2^2}{2} \, d S_2 \, d S_3}$$

$$= \frac{\frac{1}{2}}{\int_0^1 S_3 \left(\frac{1}{4} - \frac{S_3^2}{4}\right) d S_3}$$

$$= \frac{1}{2\left(\frac{1}{8} - \frac{1}{16}\right)}$$

$$\frac{hD}{k} = 8$$

This is the limiting value of Nu when k becomes large using the approximation $u = u_m$ over the entire radius of the tube.

Martinelli in his papers failed to extend his work to this limit. His method, while directed toward the same objectives and following the same reasoning is markedly different in details and obscures the simplicity of the calculation of the limit. He does refer to a limiting value obtained from Norris and Streid's work with laminar flow. As would be expected, this gives a considerably lower limiting value for Nu, than for turbulent flow.

He has now issued a correction which in addition to rectifying an error lists values of Nu at Pr = 0 as ranging from 7.02 to 7.27 at Re of 10^4 to 10^6.

Calculations by graphical integration of the limiting case using actual experimental results of Nikuradse for $\frac{u}{u_m}$ at various values

of S gives the result that at Re = 4000*, Nu = 6.84, and at Re = 3,240,000, Nu = 7.15.** We see, therefore, that the limiting value for Nu is roughly 7 and that the error of using the approximation $u = u_m$ is about 14% in the limiting value.

Diffusivity and Comparison with Fluid Friction

Molecular conductivity, k, has the units of (heat)/(time)(area)(temperature/length). If we divide k by $c\rho$, the volume heat capacity, we obtain the molecular diffusivity of heat, $\frac{k}{c\rho}$. This property has the units, area per time.

Similarly, $\frac{E}{c\rho} = \epsilon_H$, the eddy diffusivity of heat.

We now follow a classical line of reasoning parallel to that worked out originally by Prandtl for momentum.

We have already seen that

---

* Re stands for the Reynolds Modulus

$$Re = \frac{u_m D \rho}{\mu}$$

where ρ is density, and μ, viscosity.

Fluids flowing with a value of Re below about 4000 often move in a laminar fashion rather than turbulently. The general development in this report holds for both laminar and turbulent flow, but the numerical results are calculated only for fluids in turbulent motion. A Reynolds Modulus of 3,240,000 is considerably above that usually found in ordinary heat transfer equipment.

** These calculations are straightforward, graphical integrations and are not shown here because of space limitations, and since they follow the obvious course. Examples will be given in the final report on this problem.

$$\frac{dt}{dr} = \frac{q}{A\,\bar{K}}$$

or

$$\frac{q}{A} = \bar{K}\,\frac{dt}{dr} = (k + E)\,\frac{dt}{dr} \qquad (2)$$

where q flows toward the center of the pipe. This may also be written

$$\frac{q}{A\,c\rho} = \left(\frac{k}{c\,\rho}\right) + \epsilon_H\,\frac{dt}{dr}$$

The heat, q_E, which flows by virtue of eddying only, can be given by

$$\frac{q_E}{A\,c\rho} = \epsilon_H\,\frac{dt}{dr} \qquad (7)$$

In the eddying stream, let us consider two filaments parallel to the axis and a distance ℓ apart.

The temperature difference between the filaments is $\ell\,\frac{dt}{dr}$ and the velocity difference is $\ell\,\frac{du}{dr}$.

If a lump of fluid retains its identity while moving between the filaments, it will carry an available heat proportional to $\ell\,\frac{dt}{dr}\,c\rho$. Upon entering the new filament, the lump will displace another lump which must ultimately return to the first filament as shown in Figure 7. If two continuous streams of lumps flow between the two filaments we have what appears to be an eddy moving downstream at a rate between those of the two filaments. The speed with which the eddy spins, and hence the velocity with which the lumps move from one filament to the other will be proportional to the difference between the velocities of the two filaments.

Hence we may write,

$$\frac{q_E}{A\,c\rho} = -B_H\,\ell^2\,\frac{dn}{dr}\cdot\frac{dt}{dr}$$

where B_H is some proportionality constant. Let us now define ℓ_H so that $B_H = -1$. That is, ℓ_H is defined by the equation:

$$\ell_H \equiv \sqrt{\frac{q_E}{A\,c\rho}\cdot\frac{-1}{\frac{du}{dr}\cdot\frac{dt}{dr}}}$$

or

$$\frac{q_E}{A\,c\rho} = \ell_H^2 \frac{du}{dr} \cdot \frac{dt}{dr}$$

Remembering Equation (7), we may also write

$$\epsilon_H = \ell_H^2 \frac{du}{dr}$$

Similarly for momentum transfer, the shear τ_E due to eddying in units of force per unit area is proportional to the rate of momentum transfer.

$$\frac{\tau_E}{\rho'} = B_M \ell^2 \frac{du}{dr} \cdot \frac{du}{dr}$$

and if ℓ_M is defined so that $B_M = 1$,

$$\ell_M^2 \frac{du}{dr} = \epsilon_M$$

the eddy diffusivity of momentum. ρ' indicates the density in mass units, that is $\rho' = \frac{\rho}{g}$, where g is the gravitational constant.

ℓ_M and ℓ_H are known as mixing lengths and may be thought of either as the effective eddy diameters or as the effective distances a discrete lump of fluid travels before losing its identity with respect to momentum and heat. They are also sometimes referred to as the mean free path of the lump.

If ℓ_M and ℓ_H are identical, then $\epsilon_M = \epsilon_H$. Conflicting evidence exists to date on this question. Martinelli, following a suggestion of Boelter, sets

$$\frac{\epsilon_H}{\epsilon_M} = \alpha$$

and assumes α is a constant.

We will also assume α is a constant. There is so little evidence regarding this quantity that if we are to obtain any kind of a solution we are forced to this assumption. We will carry it as a constant through the final calculations and into the simplified equation. There, until its true nature becomes known, we will assume it is unity in the calculation of problems.

We may now rearrange equation (6) to get

$$Nu = \frac{\frac{1}{2}}{\int_0^1 \frac{u_3}{u_m} S_3 \int_{S_3}^1 \frac{\frac{k}{c\rho}}{S_2\left(\frac{k}{c} + \alpha \epsilon_M\right)} \int_0^{S_2} \frac{u_1}{u_m} S_1 \, d S_1 \, d S_2 \, d S_3}$$

or

$$Nu = \frac{\alpha Pr}{2 \int_0^1 \frac{u_3}{u_m} S_3 \int_{S_3}^1 \frac{1}{S_2\left(\frac{1}{\alpha Pr} + \frac{\epsilon_M}{\nu}\right)} \int_0^{S_2} \frac{u_1}{u_m} S_1 \, d S_1 \, d S_2 \, d S_3} \qquad (6e)$$

where Pr is the Prandtl modulus, $\frac{c\mu}{k}$, and where ν is the kinematic viscosity, $\frac{\mu}{\rho}$.

Friction Velocity - Additional Forms of Equation (6)

The Fanning friction factor may be defined by the equation:

$$f \equiv \frac{g\, r_w}{\rho\, u_m^2} \cdot \frac{dp}{dz}$$

or

$$\frac{dp}{dz} = \frac{\rho f\, u_m^2}{g\, r_w}$$

But also

$$\frac{dp}{dz} = \tau_w \frac{2\pi r_w}{\pi r_w^2} = \frac{2}{r_w} \tau_w$$

Hence

$$\frac{2}{r_w} \tau_w = \frac{\rho f\, u_m^2}{g\, r_w}$$

or

$$\frac{\tau_w}{\rho} g = u_m^2 \frac{f}{2}$$

and

$$\sqrt{g\frac{\tau_w}{\rho}} = u_m\sqrt{\frac{f}{2}}$$

This may also be written

$$\sqrt{\frac{\tau_w}{\rho 1}} = u_m\sqrt{\frac{f}{2}} \tag{9}$$

In the preceding development,

P is the static pressure of the fluid

τ is the shear

$\sqrt{\frac{\tau_w}{\rho 1}}$ is frequently referred to as the friction velocity, designated by v*.

From Equation (9) we learn that the friction velocity is the mean velocity times the square root of half the friction factor.

Two dimensionless terms involving velocity and distance to the wall can now be defined

$$u^+ \equiv \frac{u}{v^*} = \frac{u}{u_m}\sqrt{\frac{2}{f}} \tag{a}$$

and if y is the distance from the wall,

$$y^+ \equiv \frac{y}{\nu} v^* = \frac{y}{r_w} \quad \frac{Re}{2}\sqrt{\frac{f}{2}} \tag{b}$$

Re is of course the Reynolds modulus,

$$\frac{D\, u_m \rho}{\mu} \quad \text{or} \quad \frac{2\, r_w\, u_m}{\nu}$$

It will be observed that

$$\frac{u}{u_m} = u^+\sqrt{\frac{f}{2}}$$

Substituting into Equation (6e) we obtain

$$Nu = \frac{\alpha \, Pr}{f \int_0^1 u^+ \, S_3 \int_{S_3}^1 \frac{R}{S_2} \int_0^{S_2} u^+ \, S_1 \, d \, S_1 \, d \, S_2 \, d \, S_3} \qquad (6f)$$

where $R = \dfrac{1}{\frac{1}{\alpha Pr} + \frac{\epsilon_M}{\gamma}}$

Evaluation of u^+ and y^+

Nikuradse, on whose data we plan to rely in large part for the ultimate calculation, obtained an approximate general equation for u^+ in terms of y^+ which holds reasonably well in the turbulent core of the fluid stream for all Reynolds moduli:

$$u^+ = 5.5 + 2.5 \ln y^+$$

where "ln" indicates the natural logarithm.

This may also be written

$$\frac{u_{\mathfrak{c}} - u}{v^*} = -2.5 \ln \frac{y}{r_w}$$

where $u_{\mathfrak{c}}$ is the velocity at the center line since

$$\frac{u_{\mathfrak{c}} - u}{v^*} = u^+_{\mathfrak{c}} - u^+$$

and

$$u^+_{\mathfrak{c}} = 5.5 + 2.5 \ln y^+_{\mathfrak{c}}$$

A more accurate but complicated expression has been developed by Wang.

Von Karman has proposed the following expressions for u^+ vs y^+ in the transition and laminar regions:

$$u^+ = -3.05 + 5.00 \ln y^+, \text{ in buffer region} \tag{11}$$

$$u^+ = y^+, \text{ in laminar layer} \tag{12}$$

He believes that the laminar layer extends from the wall to $y^+ = 5$, and that the buffer or transition region extends from $y^+ = 5$ to $y^+ = 30$.

These equations are shown plotted in Figure 8. In Figure 9 are given the corresponding $\underline{u}$ vs S curves for Re = 4000 and Re = 3,240,000 which are the u_m limits of Reynolds range measured by Nikuradse.

Determination of R - Effect of Prandtl Number

R has been defined in the equation

$$R = \frac{1}{\frac{1}{\alpha \, Pr} + \frac{\epsilon_m}{\nu}}$$

Aside from α, which we are simply carrying along, the only unknown term is ϵ_m or $\frac{\epsilon_m}{\nu}$. This can be obtained by differentiating u^+ with respect to y^+, or directly from Nikuradse who has tabulated experimental values of

$$\frac{\epsilon_m}{r_w \, v^*} \quad \text{or} \quad \frac{\epsilon_m}{\nu} \cdot \frac{\alpha}{Re \sqrt{\frac{f}{2}}}$$

This is obtained by multiplying and dividing by 2 and ν.

The differentiation of u^+ with respect to y^+ using equations (a) and (b), discloses several interesting points.

$$\frac{du^+}{dy^+} = \frac{\nu}{\frac{\tau_w}{\rho l}} \cdot \frac{du}{dy}$$

But

$$\frac{\tau}{\rho l} = (\epsilon_m + \nu) \frac{du}{dy} = \frac{\tau_w}{\rho l} \, S$$

Hence

$$\frac{1}{\frac{du^+}{dy^+}} = \frac{\frac{\epsilon_m}{\nu} + 1}{S}$$

Drawing # 5417

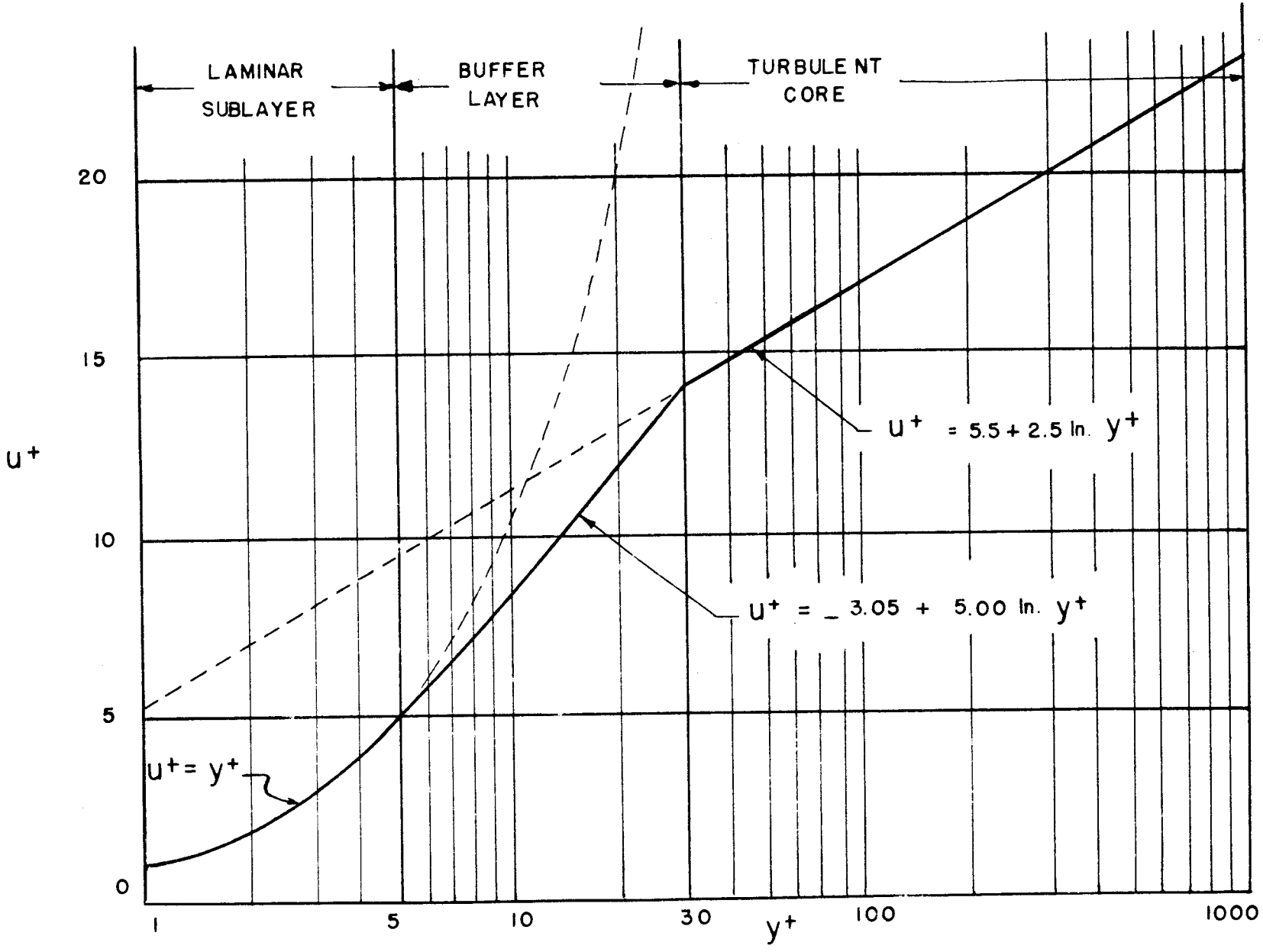


Fig. 8 GENERALIZED VELOCITY DISTRIBUTION FOR TURBULENT FLOW IN TUBES

KEUFFEL & ESSER CO. N. Y. NO. 359-91
Semi-Logarithmic, 5 Cycles × 70 to the inch, 5th lines accented
MADE IN U.S.A.

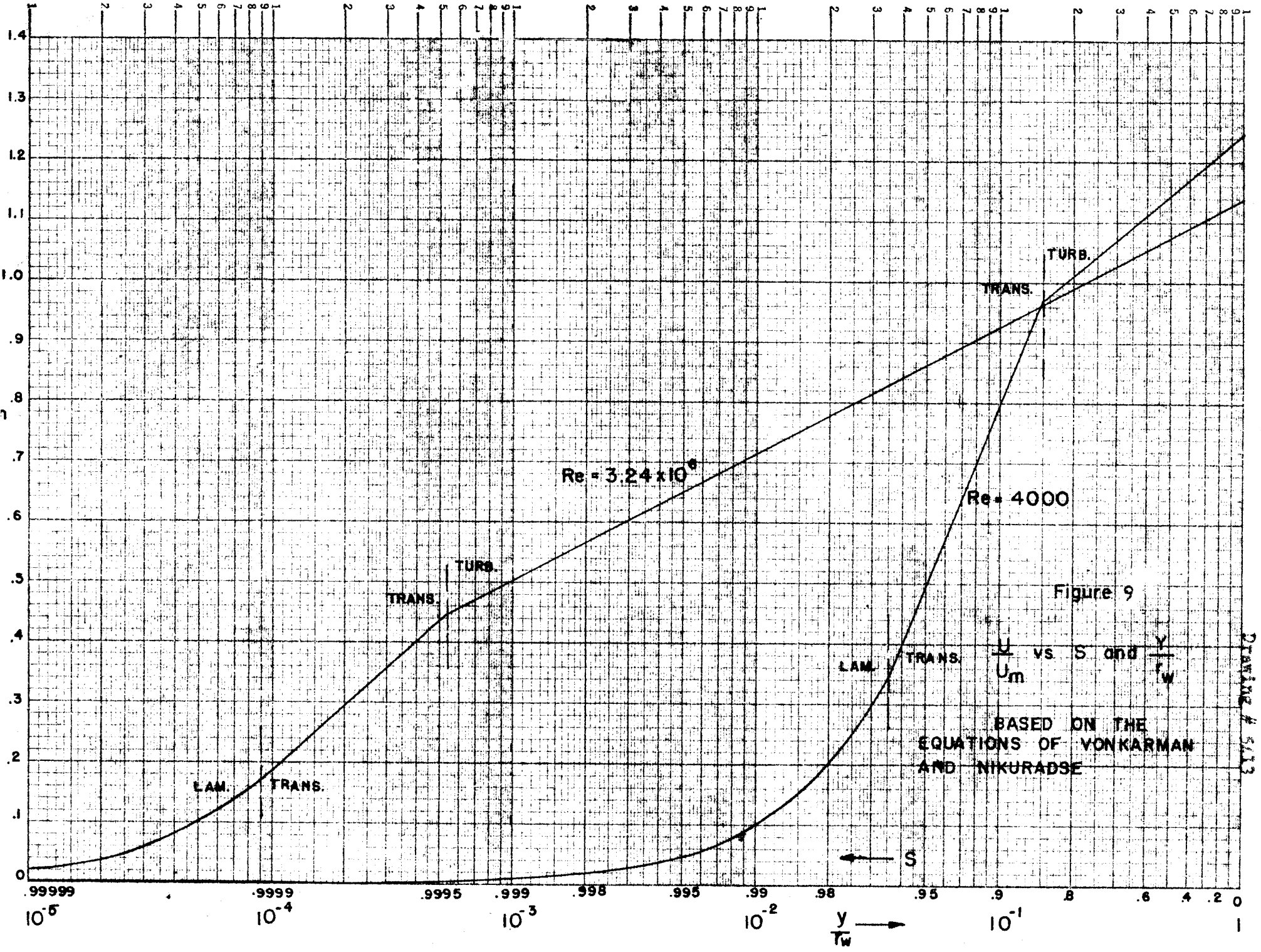


Figure 9

Drawing # 5113

In order to show how the heat conductance of the system changes in the neighborhood of the wall, we present Figure 10. The heat conductance is proportional to the group

$$\frac{1}{R} = \frac{1}{\alpha Pr} + \frac{\epsilon_m}{\nu}$$

the term ϵ_m/ν can be evaluated from the above expression for $\frac{du^+}{dy^+}$, assuming δ to be very close to unity (near the wall) and using Eq. (12) and (11) to calculate values of $\frac{du^+}{dy^+}$ in the laminar and buffer regions respectively, and the corresponding equation

$$u^+ = 5.5 + 2.5 \ln y^+$$

of Nikuradse in the turbulent core. The four lower curves of Fig. 10 result. By assuming a value of 0.005 for the Prandtl number (typical of alkali metal alloys) and combining 1/Pr with ϵ/ν the two upper curves are obtained. (It is also assumed that the uncertain constant α is unity.) We see that when Pr is large $\frac{\epsilon_m}{\nu}$ is controlling in the sum

$$\frac{1}{\alpha Pr} + \frac{\epsilon_m}{\nu}$$

even at very low values of y^+. At low Prandtl number, the eddy diffusivity does not become controlling until one is considering a region in the tube which is well inside the turbulent region.

Since the velocity distribution data from the Nikuradse's and Reichardt's experiments are in excellent agreement throughout the turbulent region, we can proceed in the case of low Prandtl number with considerable confidence. The analogy between heat transfer and momentum transfer is peculiarly adaptable to predicting the heat transfer coefficient for liquid metals, but the method cannot be applied with confidence to materials of high Prandtl number because in this case the importance of turbulence extends right up to the viscous layer, and may even extend into it.

It is much more reasonable to obtain a velocity distribution in the laminar layer along the wall using heat transfer data for high Pr materials than to estimate their heat transfer properties from a few data near the high velocity edge of the laminar region. Further mention of this will be made later.

A New Heat Transfer Correlation

We now see that if data are available on the velocity distribution in the turbulent core of a well-defined fluid in a round tube at various Reynolds numbers, we can evaluate the integrals in Equation (6), or a modification such as Equation (6f). Once this is possible, we can obtain values for Nu at various values of α Pr and Re. It should be remembered

Drawing # 5418

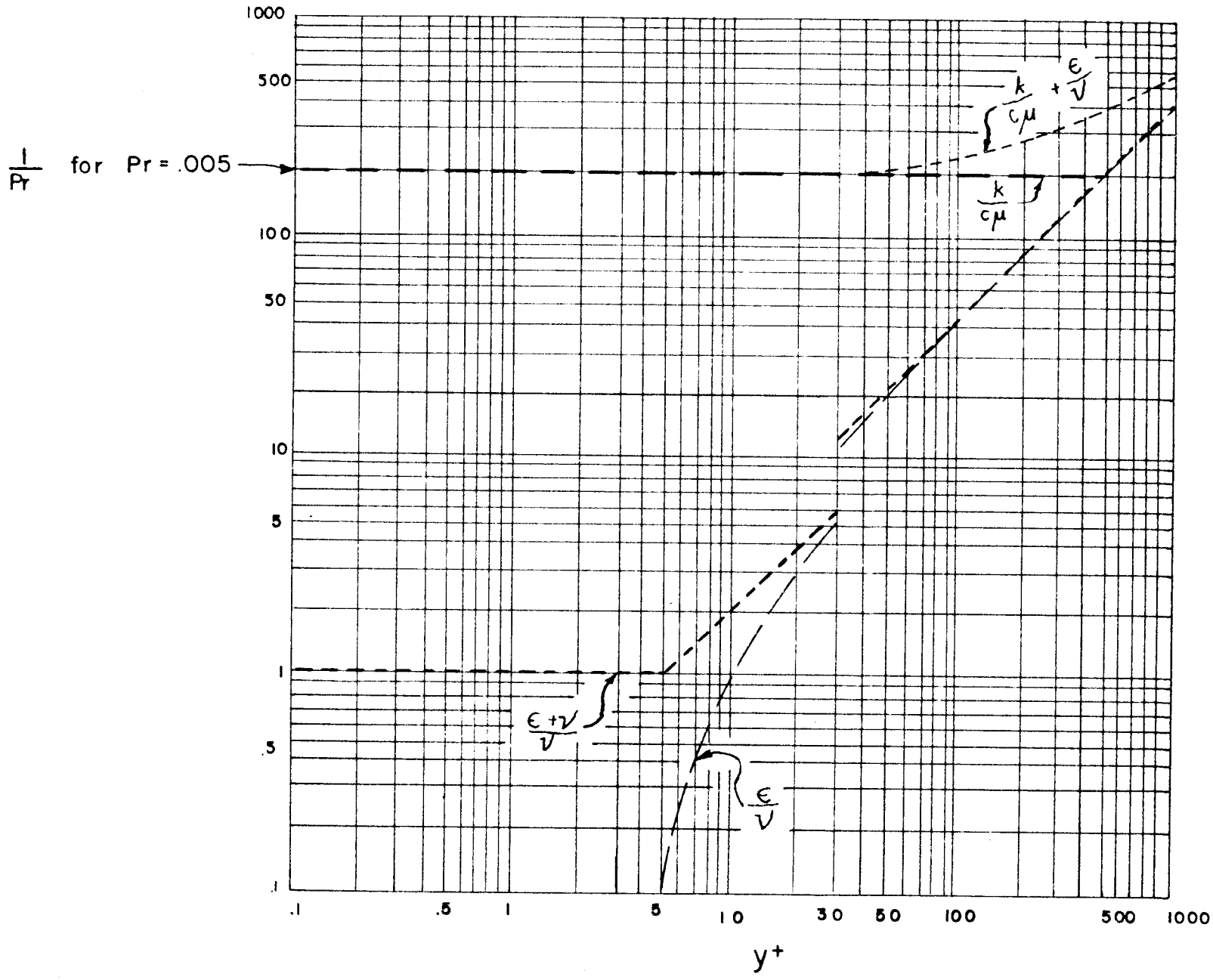


Fig. 30 EDDY AND MOLECULAR DIFFUSIVITY vs y^+

that f, the friction factor, is a function of Re alone. Either the general approximations for u^+ as a function of y^+ may be used in these calculations, or we may use the actual experimental results. Martinelli chose the former in his development which differs from, but parallels the one here. Von Karman as a predecessor of Martinelli also used the u^+ equations, and ignored molecular conductivity in the turbulent region.

We use the data of Nikuradse directly by means of graphical integrations, and hence we avoid any errors due to the inaccuracy of the generalization. The resulting set of computed values of Nu will evidently be functions of those quantities which appear in Eq. (6f), which can be combined into the familiar dimensionless groups

$$Re = \frac{D\,u\,\rho}{\mu} = \text{Reynold's number}$$

$$Pr = \frac{C_p\,\mu}{k} = \text{Prandtl number}$$

By trial we find that when we plot the values of Nu against the product of $\alpha \cdot Pr \cdot Re$, the points fall close to a single line regardless of Reynolds number. We have already indicated that the lower limit of Nu is about 7, and we find that as $\alpha \cdot Pr \cdot Re$ becomes large, with Pr remaining less than 1, Nu approaches $.023(\alpha \cdot Pr \cdot Re)^{.8}$ All of the points appear to fit reasonably well the equation

$$Nu = 7 + .05(\alpha \cdot Pe)^{.5} + .023(\alpha \cdot Pe)^{.8} \tag{13}$$

where Pe, the Peclet modulus may be thought of as the product of the Prandtl and the Reynolds moduli, or

$$Pe = Pr \cdot Re = \frac{C\,D\,u_m\,\rho}{k}$$

It is found that values calculated by Martinelli fall very close to this line as well. The agreement is shown in Figure 11.

This correlation will be expected to be best for fluids like liquid metals which have a low Prandtl number, since for such fluids the molecular conductivity of the fluid is so high that heat flows readily through even a relatively thick laminar film at the wall. The exact velocity distribution is hence unimportant, and the uncertainties resulting from our poor knowledge of the relation between u^+ and y^+ in the laminar and buffer layers cause little error in the integration of Eq.(6f). Fluids of high Prandtl number, on the other hand, do not fit the correlation very well.

It will be observed that the viscosity, μ, does not appear in Eq. (13). From this and the fact that viscosity is usually the property

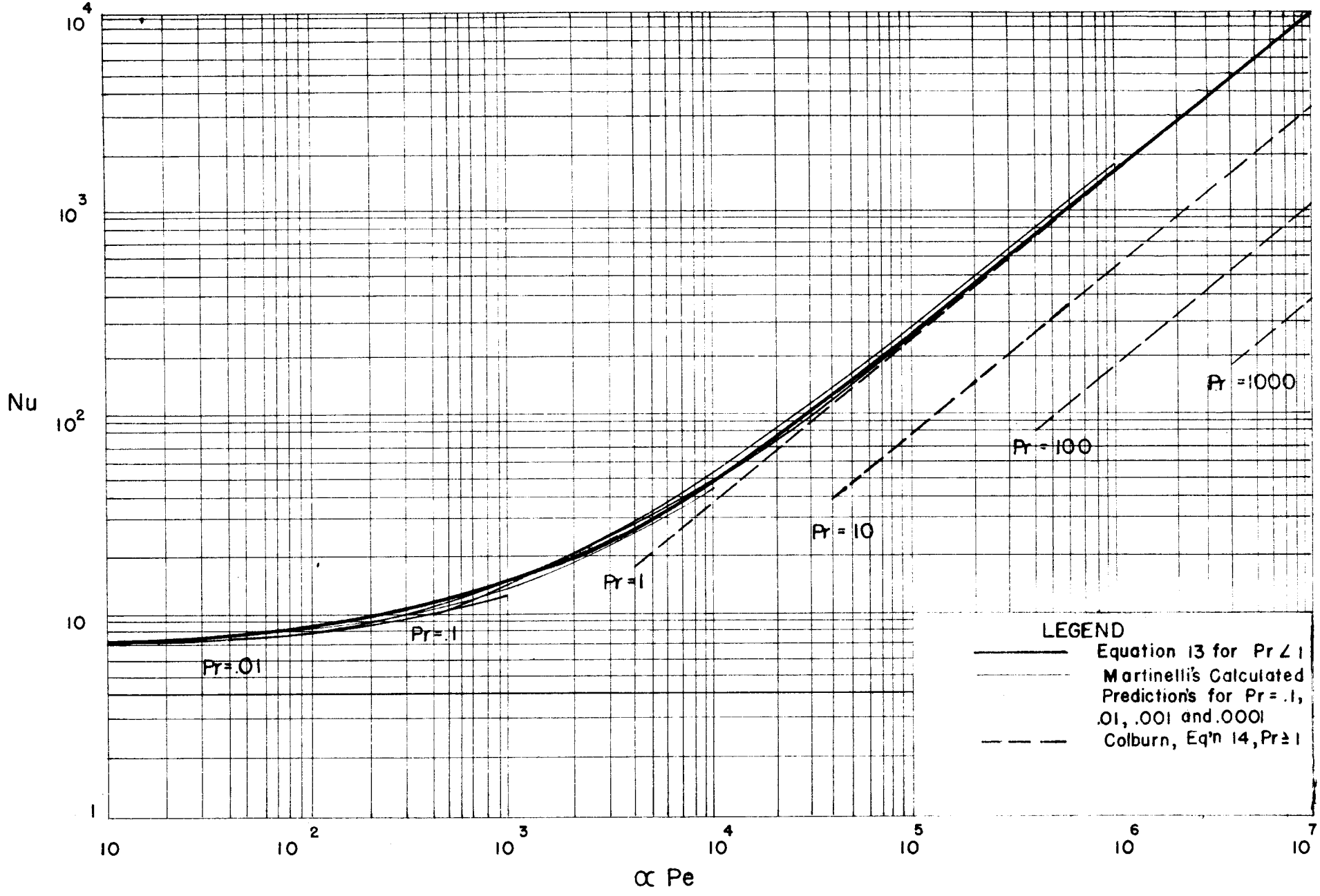


Figure PROPOSED EQUATION AND MARTINELLI'S RESULTS

most sensitive to temperature change, it may be argued that our assumption of no significant property change within the fluid is not a misleading one. Support is given this argument by recognition of the high heat transfer coefficients which will be obtained, so that the actual temperature difference between the wall and the stream center will be relatively small for ordinary heat fluxes.

Applications of Theory

Numerous problems can be attacked using the approach developed here. As already shown, the theoretical relatively rigorous equation (6f) can be evaluated, using actual experimental velocity data, to give values of Nu at low Pr as a function of α Pr and Re, and these in turn yield an approximate equation for Nu at low Pr as a function of α Pe. Experimental work with NaK at another laboratory is reported confidentially to check the predicted heat transfer coefficient within about 10%.

The approximate equation is useful in predicting the relationship between the heat transfer coefficient, and the velocity. Such a relationship is helpful in evaluating experimental heat transfer data from double tube exchangers where only the fluid temperatures are known, since it is obvious from the curve in Figure 11 that in general the h for liquid metals does not vary as the .8 power of u. A variation from the usual Wilson plot method is needed, based initially on the h change with u shown by our development.

It has been mentioned that the velocity distribution is not well established in the buffer or laminar region. Heat transfer coefficients predicted by von Karman and Martinelli on the momentum - heat analogy are low when compared with experimental results, at Pr above about 100. This discrepancy may be due to inadequate information regarding the velocity distribution, and hence the value for the eddy diffusivity, in the buffer region and particularly in the laminar region. It is obvious from the definition of R that at high Pr numbers very small values of eddy diffusivity may have large effects on the heat transfer coefficients.

It is believed that an equation for Nu, similar to the equations 6 but somewhat simpler in form, can be developed for the case of high Prandtl modulus using a number of assumptions. One such assumption for this case would be that the temperature drop from the wall to the edge of the turbulent region is large compared with the temperature differences across the turbulent core. Such an equation would of course contain a term involving R or

$$\frac{1}{\frac{1}{\alpha \, Pr} + \frac{\epsilon_m}{\nu}} .$$

For the case of Prandtl modulus greater than one, experimental data have generally been found to fit Colburn's empirical equation $Nu = .023 \, Re^{.8} \, Pr^{.33}$. If we equate his expression with our simplified

high Pr expression for Nu, we should obtain an equation involving Re, Pr, R, and S. From this equation it should be possible to develop a relationship between R and S with Pr and Re as parameters. For a given value of Pr, values of $\frac{\epsilon_m}{\nu}$ can be calculated from corresponding values of R. $\frac{\epsilon_m}{\nu}$ will be recognized as the ratio of eddy diffusivity of momentum to molecular diffusivity of momentum.

Knowledge of the values of $\frac{\epsilon_m}{\nu}$ with respect to S gives us a method of computing the velocity distribution in the laminar and buffer regions at points very close to the wall.

Such velocity distribution information has applications in hydrodynamics which in turn may lead to better understanding of flow and heat transfer in non-tubular channels, the estimation of end effects, and a better understanding of extraction and absorption phenomena.

The theory opens the way for study of other diffusional problems such as heat transfer between parallel plates or in annuli where only one side is heated. The existence of a partial diffusional barrier in the center of such a stream is indicated by the minimum in eddy diffusivity at the center of a tube reported by Nikuradse. The general approach used in developing Eq. (6) can be used to develop similar equations for annuli and other shapes where this partial barrier occurs. From the results of such a study a better method of correlating experimental data may be found.

Most important of all, the development and its results give a better quantitative insight into the mechanisms which affect the much used, but little understood heat transfer coefficient in circular tubes --already giving us a relatively simple prediction of this coefficient for liquid metals.

The theoretical work is being continued along the lines indicated above in an attempt to obtain a simplified equation for the velocity distribution in the buffer and laminar regions.

References

A. P. Colburn	Trans AIChE 29 pp 174-210 (1933)
T. von Karman	Trans ASME 61 pp 705-10 (1939)
R. C. Martinelli	Sixth International Congress for Applied Mechanics, Paris, September, 1946
R. C. Martinelli	Trans ASME 69 pp 947-959 (1947)
J. Nikuradse	V.D.I. Forschungheft 356 36 pp (1932)
R. H. Norris & D. D. Stroid	Trans ASME 62 pp 525-533 (1940)
H. Reichardt	Technical Memo No. 1047, NACA (September 1943)
Chi-Teh Wang	Journal of Applied Mechanics A86 pp 85-90 (1946)

NOMENCLATURE

c - Heat capacity

f - Fanning friction factor

g - gravitational constant

h - heat transfer coefficient

k - thermal conductivity

ℓ - mixing length

q - heat per unit time flowing radially toward the center of the pipe

r - distance from center of pipe

t - temperature

u - velocity of fluid

v* - friction velocity = $\sqrt{\frac{\tau_w}{\rho}}$

y - distance from wall

A - Area normal to heat flow

B - Proportionality constant

D - tube diameter

E - eddy conductivity

$\bar{K}$ - effective conductivity = k + E

P - static pressure

S - $\frac{r}{r_w}$

Z - distance along tube

α - $\frac{\epsilon_H}{\epsilon_M}$

ϵ - eddy diffusivity (area/time)

θ - angle displaced around tube axis

μ - viscosity

ν - kinematic viscosity = $\frac{\mu}{\rho}$

γ - density (weight units)

ρ - density (mass units)

τ - shear

Dimensionless Groups

$$u^+ = \frac{u}{v^*} = \frac{u}{u_m}\sqrt{\frac{2}{f}}$$

$$y^+ = \frac{y}{\nu} v^* = \frac{y}{r_w}\,\frac{Re}{2}\sqrt{\frac{f}{2}}$$

$$Nu = \frac{hD}{k} \qquad \text{(Nusselt)}$$

$$Pe = \frac{c\,D\,u_m\,\rho}{k} \qquad \text{(Peclet)}$$

$$Pr = \frac{c\,\mu}{k} \qquad \text{(Prandtl)}$$

$$Re = \frac{D\,u_m}{\mu} \qquad \text{(Reynolds)}$$

$$R = \frac{1}{\frac{1}{\alpha\,Pr} + \frac{\epsilon_m}{\nu}}$$

NOMENCLATURE (Continued)

Subscripts

a, b	respective filaments of fluid
℄	at centerline of tube
m	mean or average
w	at wall
E	eddying
H	heat
M	momentum
1	referring to first integration in Eq. (6)
2	second integration in Eq. (6)
3	third integration in Eq. (6)

Other Activities of the Section

In addition to the handling, experimental and theoretical work on liquid metals, considerable time was spent during the quarter in assisting and attending the NEPA Heat Transfer Symposium in December; the Heat Transfer Symposium held by the Industrial and Engineering Chemistry Division of the ACS in Chicago was attended; and Section IV was assisted in determining the thermal conductivity of various concrete samples.

6.2 Pile Air Monitoring (R. B. Briggs)

During the first week in December considerable difficulty was experienced in removing a swollen slug from one of the channels in the Clinton Pile. Swelling had proceeded to such an extent that the channel was sealed completely. The pile was shut down for several days while that channel was cleared and others were inspected. It was found that several channels were partially blocked either by blistered slugs or by pieces of wood from the broken ends of shield plugs. Because the broken slug was not detected by the monitoring devices in the exit air stream, it was requested by the Operating Department that a method be developed for detecting the swelling of slugs before they become large enough to be difficult to remove from the channels.

Measurement of the air flow in individual channels appears to be the most satisfactory method. It seems feasible to do this by mounting measuring devices on the shield plugs in the front face of the pile with those devices connected to recording instruments and an alarm system. The major difficulty with air flow measurement as a method of detecting swelling is that large changes in the cross-section of a slug result in small changes in the air velocity.

Experimental data have been obtained to show the reduction in air velocity as a function of restriction of a normal channel cross-section. Air velocity measurements were made with a Velometer in a six foot long mock-up of a pile channel loaded with wooden dummy slugs. Lucite pieces were used to simulate various degrees of swelling of a slug. Data obtained for the experimental channel and extrapolated to a 24 foot channel loaded with 15 feet of slugs are reported below:

% Restriction of Normal Cross-Section	% Increase in Slug Cross-Section	% Decrease in Air Velocity
0	0	0
10	22	0
20	45	1
40	89	6
60	130	19
80	180	43
100	220	100

A useful monitoring system will be one capable of detecting significant changes of about 5% in the air velocity. Two types of elements capable of measuring such changes are being investigated; they are a Thermistor and a very small pitot tube. The Thermistor is a thermally sensitive resistor manufactured by Western Electric Company. It operates on the principle of the hot-wire anemometer. The pitot tube is installed in a venturi making it insensitive to small variations in positioning and the assembly is small enough to permit mounting on the pile end of a shield plug. A strain gage, differential pressure transmitter mounted on the other end of the plug will be used for transmitting the velocity pressure to the recorder and alarm system. Both devices are being assembled by the Instrument Department preparatory to testing in the mock-channel.

6.3 Shielding

A summary report was completed and distributed early in this quarter. Progress toward the accurate determination of hydrogen content in magnesium oxychloride cements has reached a point such that consistent data are attainable. Both combustion and Karl Fisher analyses indicate water content of about 40%, which is two or three times that of Portland cement.

Additives such as calgon and pozzolith were tried in an attempt to raise the water content. Increases were of such magnitude that

additives, unless for stablizing reasons, are of little value.

Further x-ray diffraction data by Bredig showed the desirability of a 5:1 $MgO:MgCl_2$ ratio. This corroborates the previous work of Hubbel of Mellon Institute. Upon the basis of this knowledge $MgCl_2$ is added as a saturated solution until the 5:1 ratio is reached. If more wetting appears necessary, water is added as required. This differs from past experiments when only saturated $MgCl_2$ solutions were added to complete the wetting.

In an attempt to fill in the neutron resonance gap between iron and hydrogen, various metal chlorides were used in place of $MgCl_2$. Individual paste samples were made up using lithium, iron, copper, nickel, tin, zinc; lithium-iron and iron-copper were also used. The densities of these pastes were taken; then they were turned over to Bredig for x-ray diffraction studies and water analysis. All samples appeared structurally sound and will be tested at a later date for shielding efficiency and mechanical strength.

Following interest displayed by General Electric in the use of magnesium oxychloride concrete for the new Hanford piles, a large number of the nuclear physical and mechanical properties were determined. The concrete made for these tests is called "MO."

Description

Iron aggregate
Cement: 7.6 $MgO \cdot MgCl_2 \cdot 13\ H_2O$
Maximum density attainable consistent with high strength
No boron

Composition

MgO	7.76%
$MgCl_2$	2.42
H_2O	5.96
Fe punchings 3/4" x 3/4"	47.8
Steel shot 1/8"	21.6
Steel shot 1/20"	14.4
	100.0

In addition to the three 16 inch concrete test blocks, a special colemanite shield was also made. This colemanite shield was made with a front plate of 0.050" aluminum, sides of wood, and back of 1/8" masonite with a 2" thick layer of colemanite power contained therein. The results will be reported by the Physics Division.

A large number of physics tests were run in the west hole of the Clinton Pile. Data compiled show that a shield of two inches of colemanite followed by 48" of MO gives gamma and neutron readings far below tolerance. Relaxation lengths were, for gammas, 8 cm; for neutrons, 6.6 cm. Actually with only 36" of MO behind the two inches of colemanite, the shield is more than equivalent to 48" of W biological shield for gammas, with neutrons still safely below tolerance.

Mechanical tests were run to compare "MO" concrete with Portland. The data show it to be stronger in both compression and tension and twelve times as ductile, as shown below:

Properties	MO	Portland
Compressive Strength		
Standard ASTM 6 x 12 cylinder	4200 psi (2 day) (machine limit, did not break)	3000 psi (28 day)
Modulus of Elasticity		
Standard ASTM 6 x 12 cylinder	300,000 psi	3,500,000 psi
ASTM Beam Test	310,000 psi	3,500,000 psi
Modulus of Rupture		
ASTM Beam Test	1370 psi	410 psi

This concrete was found to have a thermal conductivity about 1/10 that of iron and approximately 50 times that of normal concrete:

"MO"	K - 2.4 BTU/hr ft^2 $^\circ$F
Fe	(212 $^\circ$F) K - 30.4 (Perry's Handbook)
Heat Capacity	
"MO"	.143 cal/g/$^\circ$C
Fe	.104

The cost of materials as of February 1, 1948 was $305/yd^3. From the physics data and mechanical properties, "MO" shielding appears to be a very cheap and efficient substitute for both W thermal and W biological shield. Its behavior over a long term is, however, not known.

6.4 Ra-Be Source Production (R. B. Briggs)

Testing of equipment and process for the production of >100 mg, Ra-Be sources in Building 706-G has been completed. Twenty-seven dummy runs were made with barium bromide as a substitute for radium bromide. No changes in equipment or procedures were required during the last nine runs and a recovery of 100 to 105 percent was demonstrated as based upon

the weights of barium bromide and beryllium powder added to the system in the preparation of each source. The increase in weight appeared to result from the inclusion in the Ba-Be pellet of small bits of glass and graphite and the absorption of moisture during handling prior to weighing.

For one of the runs, ~0.1 curie of radioactive barium bromide was used to obtain information regarding the spreading of contamination. After the run, the only contamination found was on equipment used in handling the radioactive material.

With the completion of the dummy runs, Building 706-G was placed in stand-by condition and responsibility for the building transferred to the Operating Department. No further work will be done on this project until orders are received for large sources.

6.5 Hood Design (A. D. Mackintosh)

A project has been started with the objective of developing a set of standard hood designs for radioactive work of various kinds.

6.6 Small Pump Testing (R. B. Briggs)

Testing of small pumps for use in direct pumping of radioactive liquids to extraction columns was completed, except for minor activity, with the testing of a temporary installation in the Hot Pilot Plant. Results of the testing program show that standard proportioning pumps are satisfactory for transferring liquids but that they should be used in conjunction with flowmeters if the flow rate must be known accurately. Minor activity consists of completing tests on the life of a stainless steel bellows and assisting the Instrument Department in the evaluation of new types of flowmeters.

A Hills-McKenna, double ball check valve in the aqueous feed line to the column was coupled with a liquid piston of methylcyclohexane to a Milton-Roy, micro adjusting, proportioning pump located in the pipe tunnel. Methylcyclohexane fed by a second proportioning pump into a head tank at a level above that of the feed pump and connected into the aqueous feed tank maintained the pumping system at slightly greater than atmospheric pressure. The rate of pumping of aqueous feed was determined by periodic measurement of the rate of addition of methylcyclohexane to the aqueous feed tank. Such an installation would not be considered for plant use but proved to be the simplest, reasonable arrangement of equipment for the Hot Pilot Plant test.

Preliminary tests showed that loss of head on the system and a resulting negative pressure at the pump permitted air to enter the liquid leg through the packing around the piston. This caused the flow rate to decrease from the desired rate to zero within a few hours. The difficulty was overcome by feeding water or methylcyclohexane into the lantern ring in the packing gland to provide a liquid seal on the piston.

The pumping system was used to provide aqueous feed for the second cycle extraction column during one run. At the beginning of the run the pump was adjusted to feed the column at a rate of 7000 ml/hr. During the following 90 hours no changes were made in the pump setting and the flow rate varied between 5960 and 7570 ml/hour. Although the indicated flow rate variation was 23% of the average flow rate, hourly variations were only 1 to 7%. It is probable that much of the variation resulted from errors in measurement of flow. The general performance of the pump was found to be satisfactory and control within ±2% could be achieved easily when operating a properly metered system.

One test has been completed and a second is under way to determine the service life of stainless steel bellows in a bellows pump. The bellows under test are 2 3/4 in. long, 1 in. I.D., 2 1/2 in. O.D., made of 0.010 in., #316 stainless steel and designed for a maximum total stroke of 1/4 in. equally divided in tension and compression. The first bellows failed after 1,700,000 flexures at a rate of 38 flexures of 1/4 in. displacement per minute. Water was pumped at a rate of 5 gal/hr. against 10 ft. of head. The second test is still in progress after pumping water for 159 days; 8,424,000 flexures of 1/8 in. total stroke at a rate of 38 strokes/min. During the 159 day period the variation in flow, at constant ambient temperature and without adjustment of the pump, has been ± 2.4%.

The problem of measuring flow rates of 100 ml/min. of radioactive solutions is being attacked with a flow meter based upon the electrocaloric principle. Heat supplied by an electric heater is introduced into the fluid through the wall of a pipe within which the stream flows. Thermocouples upstream and downstream of the heated section indicate the temperature difference which varies inversely with the flow rate at a constant power input. This method of measuring flow has been found to be accurate within at least ±2%. The Instrument Development Section of the Research Engineering Division is providing for testing an installation that will maintain a constant temperature difference and record the heat input as a measure of instantaneous and integrated flow. It appears that one of these installations will provide a relatively trouble-free method of measuring low flow rates.

7. Administrative

7.1 Personnel

Effort

The Technical Division contained 101 technical and 87 non-technical employees at the close of business February 27, 1948. This is an increase of two and six, respectively, over the figures shown for the last quarter ending November 28, 1947. Personnel changes are as follows:

Hires

5 Chemical Engineers (Davis, Coughlen, Lane, McLain, Nurmi)
1 Chemist (Lanham)
2 Mechanical Engineers (Long, Zapp)

Hires (continued)

1 Research Assistant (Kibby)
11 Non-technical employees (Bamberg, Blalock, Bond, Margaret Day, Delozier, Fowler, Frye, Johnston, Smith, Stradder, Wiggins.)

Terminations

4 Chemical Engineers returned to private industry. (Ward, Bornwasser, Burris, Arehart)
1 Assistant Director transferred to Argonne. (Bigler)
1 Chemist returned to private industry. (DeHaan)
8 Non-technical employees (Allen, Barnes, Beeler, Brown, Caraglin, Frye, Henry, Lively)

Transfers In

1 Administrative Assistant from Service Department (West)
4 Non-technical employees (Barnes, Hall, Leinart, Tuttle)

Transferred Out

2 Chemists to Chemistry Division (Baldwin and Savolainen)
1 Non-technical employee (Margaret Day)

7.2 1000 Project Progress Statistics (W. P. Bigler)

The attached graph summarizes the status of developmental and process design work on the high flux pile.

7.3 Program and Space Planning (C. E. Winters)

Scheduling

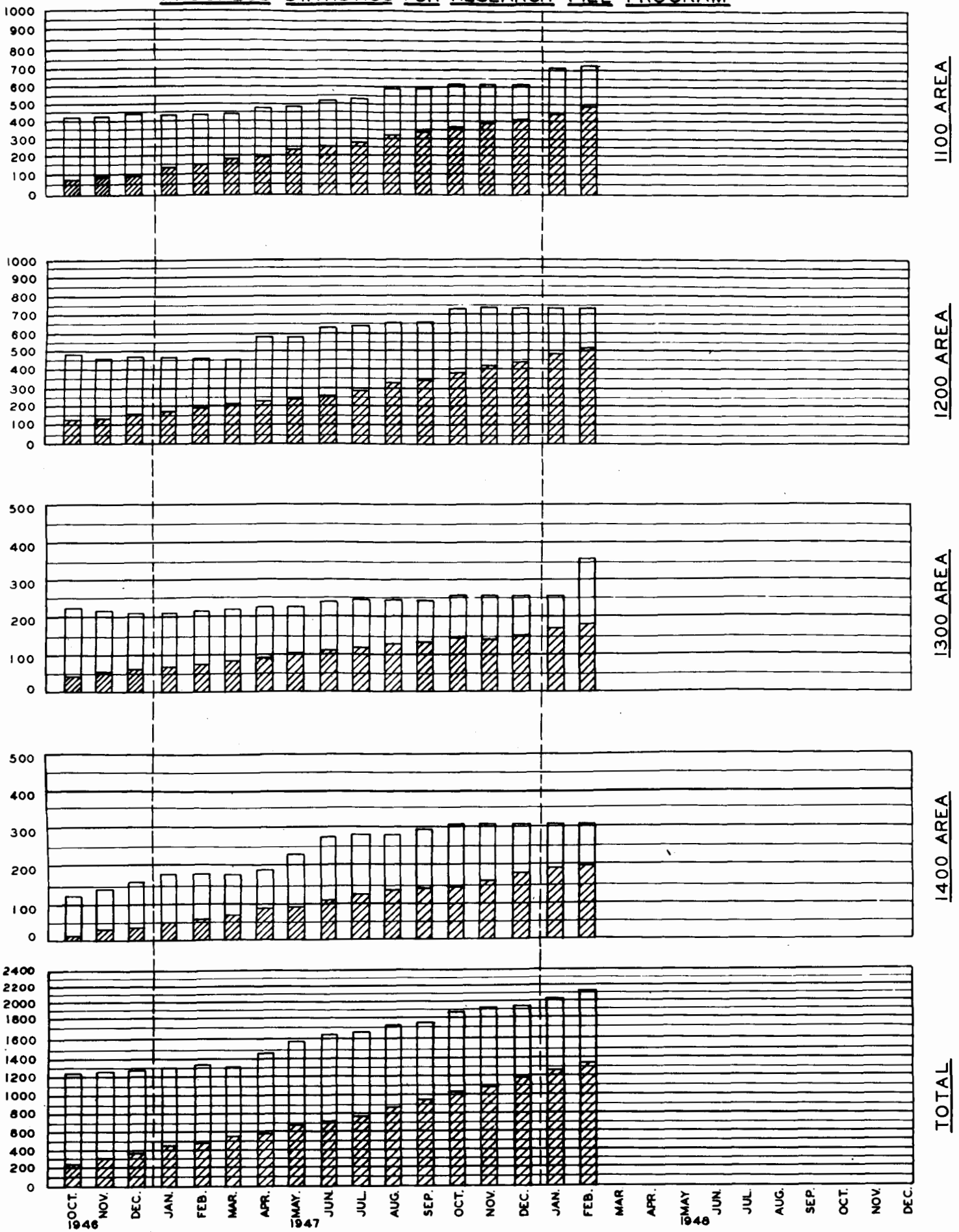
During the month of December, a high flux reactor design schedule was prepared and issued. With the drastic change of plans announced December 30, this schedule is now only of historic value. No further efforts have been expended in this direction, and probably will not be until conditions have stabilized sufficiently to permit scheduling. Efforts to prepare a formal schedule for the work of the balance of the Technical Division were also shelved.

Space Planning

Based upon the assumption that it is desirable to provide the Technical Division with permanent structures to replace the 14 widely scattered buildings it now occupies, and that a group of the order of 100 technical personnel in this group will be required to prosecute adequately an approved program, generalized preliminary plans have been drawn and a typical personnel distribution chart has been prepared.

Drawing # 5420

PROGRESS STATISTICS FOR RESEARCH PILE PROGRAM



MAN MONTHS EXPENDED TO DATE

ESTIMATED REMAINING MAN MONTHS

[redacted]

In this building plan, space is not provided for the Pilot Plant group, since it is assumed that the space in the present and future pilot plants will be adequate. Space is provided for that fraction of the Chemistry Division's analytical group that is normally employed on samples furnished by the applied chemistry and semi-works groups, with some allowance for miscellaneous work for the engineering groups. This preliminary plan shows 63,000 sq. ft. of floor space. A very crude estimate of cost would be $2,000,000. It is not felt that it will be desirable to pursue the plans very much beyond that outlined above until a reasonably firm basis exists for the preparation of a more detailed plan and cost estimate.

Program

During January an extensive list was prepared giving the items which a group similar to the present Technical Division could undertake after the work on the high flux reactor tapers off. To date, efforts to evaluate this list and prepare a realistic, long range program for the division have been unsuccessful. It is planned to continue these efforts until such time as the preparation of a program becomes possible.