DEVELOPMENT AND TEST EVALUATION OF DUPLEX STEAM REFORMER TUBE

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1. BACKGROUND

The steam/methane reformer which produces synthesis gas (hydrogen and carbon monoxide) has been identified in the United States and the Federal Republic of Germany to have a significant potential in the application of high temperature gas reactors (HTR) to synthetic fuels production. The applications include conversion of coal to fluid fuels, oil shale processing, and the closed-loop thermochemical energy pipeline (TCP). In these applications the HTR supplies high temperature (approximately 950°C) helium to heat the steam/methane reformer, where the heat is converted to chemical energy by converting a steam/methane mixture to a hydrogen/carbon monoxide mixture.

As shown in Figure 1, in the TCP application the H_2/CO mixture is piped over long distances (150 to 300 kilometers) to dispersed industrial users where the chemical energy is released in methanator units to produce 600°C heat (e.g., steam) for industrial process heat and daily peaking electricity generation. The product of the methanation reaction (methane and water) is piped back to the HTR plant for recycle through the reformer.

A recent preliminary market study in the U.S. shows that there is a potentially significant economic incentive (possibly as high as 40% cost savings) for the HTR-TCP system to supply heat to one- and two-shift dispersed industrial users and for daily peaking electricity generation. This study indicated potential U.S. markets of 250 GW_t for industrial heat and 150 GW_t in peaking electricity generation by the year 2020, including only new and replacement markets (no retrofitting) and assuming a 2% growth rate. The HTR-TCP system offers the advantage of operating the HTR plant at a unity capacity factor and storing the gases during off-peak hours while supplying one- and two-shift users. The pipeline volume itself provides eight hours of storage (at slightly increased pressure) while additional storage can be provided in underground caverns.

For HTR applications involving a steam reformer (SR) it is uncertain whether an intermediate heat exchanger (IHX) is required.

There are several system configurations (Figure 2) that could be developed for the application of nuclear heat using the steam reformer reaction. They may be characterized as:

SR-PHX — The steam reformer process heat exchanger, constructed with single-wall tubes, is inside the secondary containment and is located directly within the primary coolant path.

SR-IHX — The steam reformer process heat exchanger is constructed with double walled (duplex) tubes providing, relative to the single-wall tube design:
- increased reliability
- capability for leak detection
- greater resistance to hydrogen and tritium diffusion.

Loop - IHX — An intermediate heat exchanger is inserted into the primary loop isolating the primary system from the process heat exchangers via an intermediate loop. The steam reformer process heat exchangers are located outside the reactor containment building.

The considerations (advantages vs. disadvantages) for each of the system configurations are summarized in Figures 3A, B and C. The approach that technically and economically appears to be the most attractive in studies conducted by General Electric, combines the SR process heat exchanger and the IHX in a single component using a duplex tube. A central question concerning the duplex tube concept is whether the design would provide adequate leak monitoring capability and significant reduction in tritium and hydrogen diffusion, while introducing only a small increase in overall temperature difference from the helium to the process gas.

A cooperative GE-KFA effort was undertaken to develop, fabricate, test, and evaluate a duplex steam reformer tube. GE was responsible for the development and fabrication of the tube, and KFA was responsible for testing the tube in the EVA I facility at Jülich. Both GE and KFA are evaluating the thermochemical and metallurgical test data. Actual fabrication of the tube was performed by Foster-Wheeler in accordance with the GE design. This paper reviews the highlights of the fabrication development and preliminary evaluation of the test data.

2. GENERAL DESCRIPTION

A schematic of the duplex steam reformer tube is shown in Figure 4. The H_2O and CH_4 process gas mixture enters the top and flows downward through a bed of catalyst pellets where the reforming reactions take place. After passing through the catalyst bed, the reaction products flow upward through a coiled "pigtail" tube and leave the reformer at the top. As the products pass through the pigtail tube, they are cooled by heat transfer to the reactants. The radial gap between the duplex tubes is filled with a monitoring stream of helium. The stream of helium in the gap annulus provides a technique for sensing a tube leak.

2.1 Design Configuration

The Duplex Tube Assembly consists of a capped tube within another capped tube (Figure 4). The tube material was Incoloy 800H, the nominal tube configuration I.D. was 90.9 mm (3.58 in.), O.D. 127 mm (5.0 in.), and overall length 12 m (472 in.). A radial gap of 0-0.076 mm (0-0.003 in.) was specified between the O.D. of the inner tube and I.D. of the outer tube. Six longitudinal grooves were located on the O.D. of the inner tube to provide a gas flow path between tubes. Nozzle fittings were welded to the outer tube and were used during testing to flow helium through the gap to measure rates of hydrogen permeation from the process gas to the gap. The inlet nozzle was located on the bottom end cap of the outer tube. The outlet nozzle was located at the upper or open end to the outer tube.

2.2 Gap Sizing

Heat is transferred from the helium to the process gas through five thermal resistances consisting of the helium film, the outer tube wall, the gap between tubes, the inner wall, and the process gas film. Calculations were performed to determine the effect of the helium-filled gap between the tubes on the overall heat transfer coefficient and to choose a gap with a resistance that is only a small percentage of the total. Based on these results, a design gap of 0-3 mils was chosen. Figure 5 shows the ΔT's across each of the thermal resistances for typical operating conditions. Note that the process side temperature should be only slightly affected by relatively large changes in the gap width.

2.3 Operating Conditions

Typical design point conditions that were used as a basis for the duplex tube design and for evaluating its performance are shown in Figure 6. The operating conditions were based on results of ongoing process heat studies.

2.4 Permeation Considerations

Some of the hydrogen generated on the process side of the reformer will permeate through the walls of the reformer into the primary helium flow stream. Similarly, a fraction of the tritium generated in the primary circuit will permeate into the process gas stream. The presence of an oxide film on the reformer tubes can result in a greatly reduced permeation rate, particularly at low hydrogen partial pressures. The rate of hydrogen permeation from the reformer tubes (in a 3000 MW_e plant) is estimated to be 28 m^3/hr (STP) with no oxide film. A single oxide film can reduce this to 3 m^3/hr (STP). The duplex tube offers the opportunity of placing four oxide films in the tritium diffusion path, which could significantly reduce the permeation rate of the hydrogen isotopes.

3. MATERIAL SELECTION

Incoloy 800H was chosen as the alloy to be used in the fabrication of the duplex steam reformer tube. The advantages of Incoloy 800H are summarized as follows:

Creep strength camparatively high
Free from cobalt and tantalum (potential radioactivity problem)
Good thermal stability
Hydrogen permeability comparatively low
Should oxidize in steam (iron-base alloy)
Readily available
Good fabricability

4. FABRICATION PROCESS

A survey was made of potential duplex tube fabricators to evaluate possible methods of fabrication and to determine interest in a fabrication program.

The following fabrication methods were evaluated:

- Slip Fit
- Kinetic Expansion
- Hydraulic Expansion
- Drawing
- Swaging
- Intraforming
- Coextrusion

The methods considered to be most satisfactory for fabrication of the test duplex tube assembly were kinetic expansion and hydraulic expansion. Since the results of the survey indicated that the kinetic expansion (explosive deformation) process was the most practical and feasible production method for the test tube, a subcontract was placed with the Foster Wheeler Development Corporation to demonstrate the feasibility of fabricating a duplex tube assembly. Choice of the explosive forming for the test tubes does not imply that such a process would be chosen for production assemblies.

4.1 Fabrication Feasibility Demonstration

The objective of the fabrication feasibility demonstration program was to develop the techniques necessary to fabricate duplex tubes, having controlled dimensions, by explosively expanding an inner tube into an outer tube. Once formed, the radial gap between the two tubes was required to be within 0.076 mm (0.003 in.).

The objectives of the program were successfully met, as demonstrated by the fabrication of a 3.05 m (10-ft.) long stainless steel 304 duplex tube with end caps and with the required tolerances on the gap between the two tubes (Figure 7 - Fabrication Feasibility). However to achieve the small radial gap between the tubes (<0.076 mm), it was found necessary to utilize an annealing process between a "rough sizing shot" and a "final sizing shot" to achieve the required gap between the two tubes. In the simplest terms, this is because elastic "springback" of the inner tube and plastic deformation of the outer tube place limits on the minimum gap that can be attained. No problems were encountered with the end caps or end cap welds.

4.2 Verification Tests

Prior to fabricating the first complete duplex tube assembly for testing at KFA, a verification test program was performed on prototypical duplex assembly materials. The objective of the test program was to establish the explicit charge sizes and the process procedure that was to be used to achieve the desired duplex assembly characteristics. Incoloy 800H tubing material from the production tubes was used for the verification tests.

A total of six duplexing tests were conducted. Figure 7 - Verification Tests summarizes the results of the duplexing tests. After the gap was achieved in the third test, which required three expansion cycles, a fourth test was performed in an attempt to achieve the required interface radial gap of less than 0.003 in. without going through an intermediate annealing step. The required gap was obtained and two more tests were conducted with slightly varying charge sizes to verify that the results obtained were consistent.

5. FABRICATION

The assembly of the tubes was performed in the vertical position by first placing the outer tube into a narrow 40 foot well and then lowering the inner tube. The radial gap before detnaforming is 0.125 inches. The practice assembly operation is shown in Figure 8. Once the inner tube was installed, the tube was positively centered at the top and bottom. A 0.125 inch thick metal strip gage was passed down between the tubes in

four places to ensure that the gap was continuous the full length of the tubes. The detnaforming process was performed in three stages using the parameters established by the verification tests. One of the explosive forming shots is shown in Figure 8. Note that the inner tube is filled with water for the explosive forming.

After final welding and machining of the duplex tube assembly, pressure tests, leak tests, and gap flow tests were performed on the assembly. No direct measurements could be made of the final gap size. The flow test data indicated only the existance of a gap. The completed duplex tube assembly met all the manufacturing and test requirements for testing in the EVA facility at KFA.

6. TEST PLAN

The test plan is shown in Figure 9. The test conditions and test runs shown in the plan were chosen to allow verification of the thermodynamic behavior of the DSR tube under various temperatures, steam to methane ratios, and flowrates. The total test duration was 3 weeks, with about 1 week at the highest helium temperature, 950°C.

7. ASSEMBLY AND INSTALLATION IN EVA

A sketch of the DSR tube installed in EVA is shown in Figure 10. Temperatures, pressures, and process gas compositions were obtained at each of the four measurement planes shown. Measurement plane 1 is located at the top of the catalyst bed. The catalyst consisted of highly porous alumina substrate Raschig rings coated with nickel.

The DSR tube was surrounded by a shroud which left an annulus 15 mm wide between the outside of the duplex tube assembly and the inside of the shroud. This resulted in a large helium velocity along the length of the tube and, therefore, a large heat transfer coefficient between the helium and the steam reformer tube.

Temperatures were measured using two thermocouples at each measurement point. Pairs of thermocouples were located in the high temperature helium and process gas streams at each of the four measurement planes. Figures 11 12 show the instrumentation installed at plane 4 on the wall of the DSR tube and at the bottom of the pigtail subassembly, respectively.

Process gas compositions were determined by gas chromatograph (G.C.). Small diameter tubes attached to the outside of the pigtail were used to obtain samples of process gas at the four measurement planes. Process gas pressure at each measurement plane was measured by comparing the static pressure in each of the G.C. sampling tubes to the process gas inlet pressure. Chart recorders at the EVA facility provided continuous monitoring of temperatures, flowrates, and gas compositions.

8. TEST DATA

Results of the DSR tests in EVA are presented in the summary of Figure 13. Power stored in the chemical reaction was calculated based on compositional changes of the process gas from the reformer inlet to outlet and was referenced to the process gas inlet temperature. The total power transferred included sensible heat effects along with compositional changes due to the chemical reaction. In these tests, the sensible heat component ranged from 15 to 20 percent of the total power transferred.

8.1 Energy Balance Uncertainty

The energy balance was calculated by dividing the helium enthalpy change by the process gas enthalpy change. Thus, for a test rig with no heat losses the energy balance should be 100%. Any heat transfer from the helium stream other than to the process gas would increase the value of the energy balance to greater than 100%. Heat losses from the process gas would also result in an energy balance of greater than 100%. However, due to the arrangement of the EVA test rig, such losses are highly unlikely.

Uncertainty in the energy balance measurement was determined based on uncertainties in the measurement of flowrates, temperatures, and process gas compositions. The maximum deviations observed in the energy balances for the measurement points were +2.2% and -10%, which were well within the ±15% total uncertainty in energy balance resulting from measurement uncertainties.

9. PERFORMANCE EVALUATION

Performance of the duplex tube was measured by two parameters: (1) the amount of energy stored in the chemical reaction per mole of methane feed, and (2) the percentage of the methane feed which reacted. For both of these performance indicators, the moles of methane feed included methane formed from the pyrolysis of any higher hydrocarbons present in the natural gas.

The thermochemical pipeline (TCP) concept provides a method of transporting process heat long distances by storing energy in a chemical reaction. The first performance indicator is a measure of the energy which can be transported without loss normalized to the methane feed rate and is, therefore, an important parameter if this reformer is to be used in conjunction with a TCP. The second parameter simply measures how completely the methane feed was consumed in the chemical reaction.

The effect of process gas temperature on the performance of the duplex tube reformer is shown in Figure 14 for methane ratios of approximately 1.9, 2.8, 3.55, and 3.75. The effect of steam to methane ratio on reformer performance is shown in Figure 15 for plane 4 process gas temperatures of approximately 825°C. In all of these cases, reformer performance was found to improve as temperatures and steam to methane ratios increased. Uncertainties were calculated to be 3% for the energy transferred to the reforming reaction and 5% for methane conversion based on 2% uncertainty associated with the gas analyses.

The measurement uncertainties are not large enough to change the order in which the data points appear on any of the performance plots. However, changes of at most 20% in the slopes of the lines connecting the data points are possible.

The results of the test measurement points were used to verify the accuracy of the DSR analytical computer code. As a result, several changes were incorporated into this code in order to obtain better agreement with the experimental data. The results of fitting the experimental data using the revised code (DSR 1) are presented in Figures 15A, 15B and 15C for measurement point 476. For these figures the experimental data are plotted along with the DSR 1 computer code results to facilitate comparisons between the two. Figure 15A shows temperature profiles, Figure 15B plots the methane, steam and hydrogen compositions, and Figure 15C presents carbon monoxide, carbon dioxide, and nitrogen compositions. In general, the temperature profiles of measurement points having an energy balance approaching 100% were fit very well by the DSR 1 computer code. The discontinuity in the process gas temperature predicted by the DSR 1 code near the top of the reformer is the result of adding all of the energy released in the pyrolysis of hydrocarbons to methane into the first axial node of the computer model.

The data fits of composition agree to within two mole percent (absolute) of the experimental values. For the hydrogen and steam compositions, this amounts to a 5% relative error. A larger relative error is attributed to the CO_2, CO, and CH_4 data since the compositions of this constituents were in the range of 3 to 12 percent.

The DSR 1 code calculates the composition of CO_2 assuming that the water shift reaction is in equilibrium. In every case, the experimentally determined CO_2 concentration was greater than that calculated by DSR 1. The experimental data provided gas compositions at plane 4 that indicated an equilibrium constant greater than theoretically possible. Therefore, it is suspected that some systematic error may have been present in the determination of CO_2 composition which resulted in somewhat greater concentrations being reported than were actually present.

10. MATERIAL EXAMINATIONS

A preliminary evaluation has been made of some aspects of the post-test condition of the duplex tubing material. At this time, results from visual inspection, chemical analysis, and hardness measurements indicate that, as expected, no significant changes took place during the testing of the duplex tube. No analysis has yet been made of the surface layers or the nature of the precipitate structure.

Bow and diameter measurements along the tube have been made and indicate changes from the pre-test measurements.

Interfacial gap measurements have been obtained and indicate a maximum radial gap of 0.074 inches at an axial position of 20 feet from the top weld of the adapter section. The extent of the wide open gap over portions of the cross section can be seen in Figure 16. Over some of the remaining portions of the circumference the gap is within the specified 0 to 3 mils. The gap was closed over most of the circumference on cross sections that were taken from the ends of the tube. Only the middle section showed such large radial gaps. Figure 17 is a cross section taken at the bottom of the duplex tube. The large gap shown around the end cap is typical of the sections made during the fabrication feasibility phase.

Tensile test results have been obtained and a preliminary analysis indicates the following results which agree with expectation from the process and test conditions:

1. The inner tubes of the EVA tested sections have approximately a 60% higher yield strength (.2% offset) than the outer tubes of the corresponding sections. The detnaforming procedure has substantially cold worked the inner tube of the duplex assembly.
2. The inner and outer tubes of the pre-detna formed archive section have essentially (to within 9% at room temperature) comparable yield strengths.
3. There is no degradation (as regards yield strength) of the tube or adaptor material as a result of the EVA test.
4. There is a reduction in the ductility of the EVA tested material, particularly the inner tubes, when compared with the pre-detna formed material.
5. The EVA test resulted in no significant yield strength, tensile strength, or ductility variation with circumferential position.
6. Inner tube, outer tube, and adaptor material in the pre-EVA test and post-EVA test condition, tested at room temperature, are within ASME SB-407 (408) yield strength, tensile strength and elongation specifications.

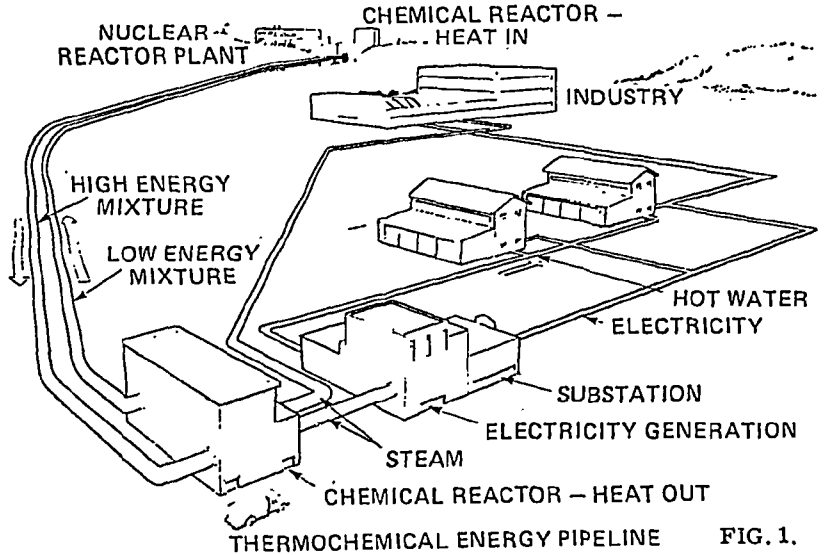


FIG. 1.

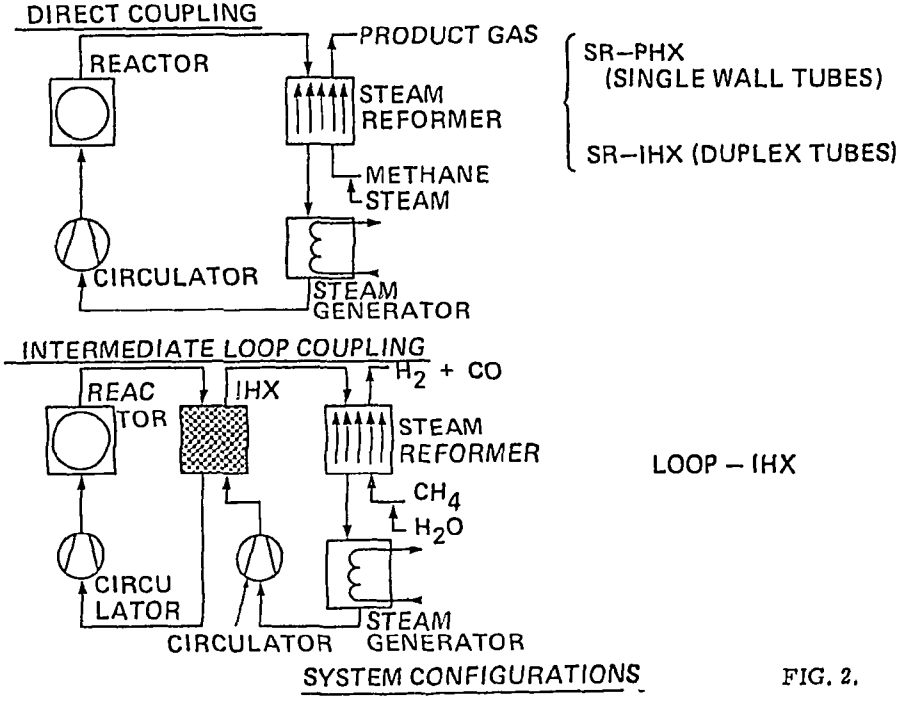


FIG. 2.

CONFIGURATION	ADVANTAGES	DISADVANTAGES
SR-PHX (SINGLE TUBE)	- LOWEST COST SYSTEM - MAXIMUM USE OF TEMPERATURE POTENTIAL - SIMPLE DESIGN	- HYDROGEN AND TRITIUM DIFFUSION MAY BE A PROBLEM - RELIABILITY DUE TO LEAKS OR RUPTURE MAY BE A PROBLEM - MAY REQUIRE INERT CONTAINMENT - MAY NOT BE LICENSABLE

FIG. 3. A COMPARISON OF COUPLING ARRANGEMENTS

CONFIGURATION	ADVANTAGES	DISADVANTAGES
SR-IHX (DUPLEX TUBE)	- LESS EFFICIENT THAN SR-PHX BUT MORE EFFICIENT THAN LOOP-IHX - PROBABLY LESS COSTLY THAN LOOP-IHX CONFIGURATION - INCREASED RELIABILITY - MINIMUM DOWN TIME - LEAK DETECTION CAPABILITY - IMPROVED RUPTURE CHARACTERISTICS - MINIMUM DESIGN PERTURBATION FROM SR-PHX - SAME MATERIAL DEVELOPMENT PROGRAM AS FOR SR-PHX - HYDROGEN AND TRITIUM DIFFUSION BARRIER	- MORE COSTLY THAN SR-PHX - MAY REQUIRE INERT CONTAINMENT

FIG. 3. B COMPARISON OF COUPLING ARRANGEMENTS (Cont.)

CONFIGURATION	ADVANTAGES	DISADVANTAGES
LOOP-IHX	- APPLICABLE AT HIGH TEMPERATURES - WITH NEW MATERIALS CAN OFFER INCREASED RELIABILITY - HYDROGEN AND TRITIUM DIFFUSION BARRIER - WOULD NOT REQUIRE INERT CONTAINMENT	- PROBABLY MOST COSTLY CONFIGURATION - SIGNIFICANT ADDITIONAL MATERIALS AND COMPONENT DEVELOPMENT - MATERIALS MAY NOT BE AVAILABLE - INCREASED OPERATING COSTS

FIG. 3. C COMPARISON OF COUPLING ARRANGEMENTS (Cont.)

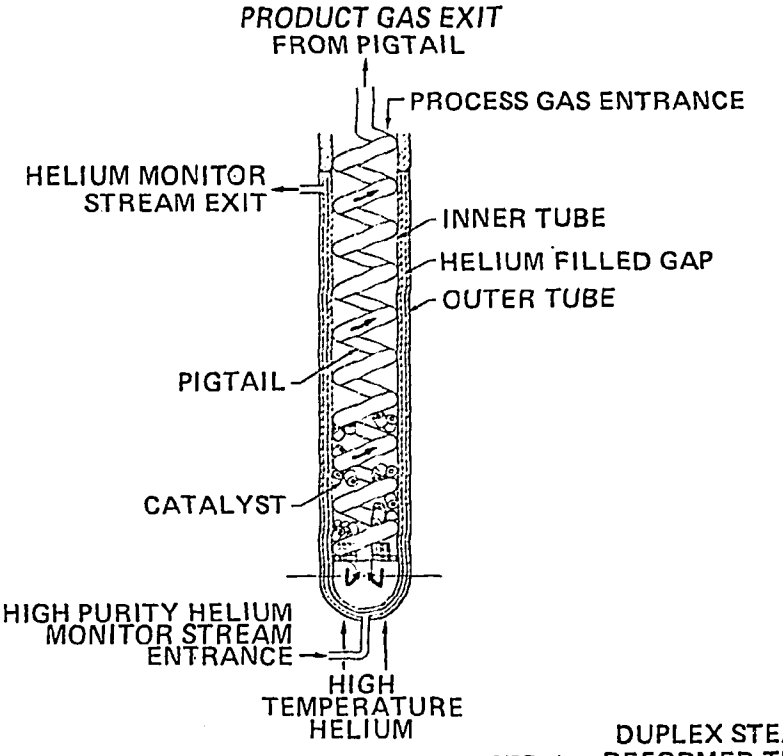


FIG. 4. DUPLEX STEAM REFORMER TUBE

TEMPERATURE DIFFERENCE ACROSS DUPLEX TUBE WALLS

THERMAL RESISTANCE	ΔT, °F	% OF TOTAL ΔT
HELIUM FILM	70	31
OUTER WALL	25	11
GAP	11	5
INNER WALL	38	17
PROCESS FILM	61	36
	205	100

FIG. 5.

DETNAFORMING PROCESS DEMONSTRATION

PHASE	MATERIAL	PROCEDURE	GAP SIZE (mils)
FABRICATION FEASIBILITY (10 TESTS)	304 S.S.	TWO EXPANSIONS, WITH INTERMEDIATE ANNEALING	1.0 TO 3.0
VERIFICATION TESTS (6 TESTS)	INCOLOY 800H (PRODUCTION TUBES)	THREE EXPANSIONS, WITH AND WITHOUT INTERMEDIATE ANNEALING	1.3 TO 2.0
PRODUCTION (2 TUBE SETS)	INCOLOY 800H	THREE EXPANSIONS	1.5 TO 3.0

FIG. 7.

DESIGN AND OPERATING CONDITIONS

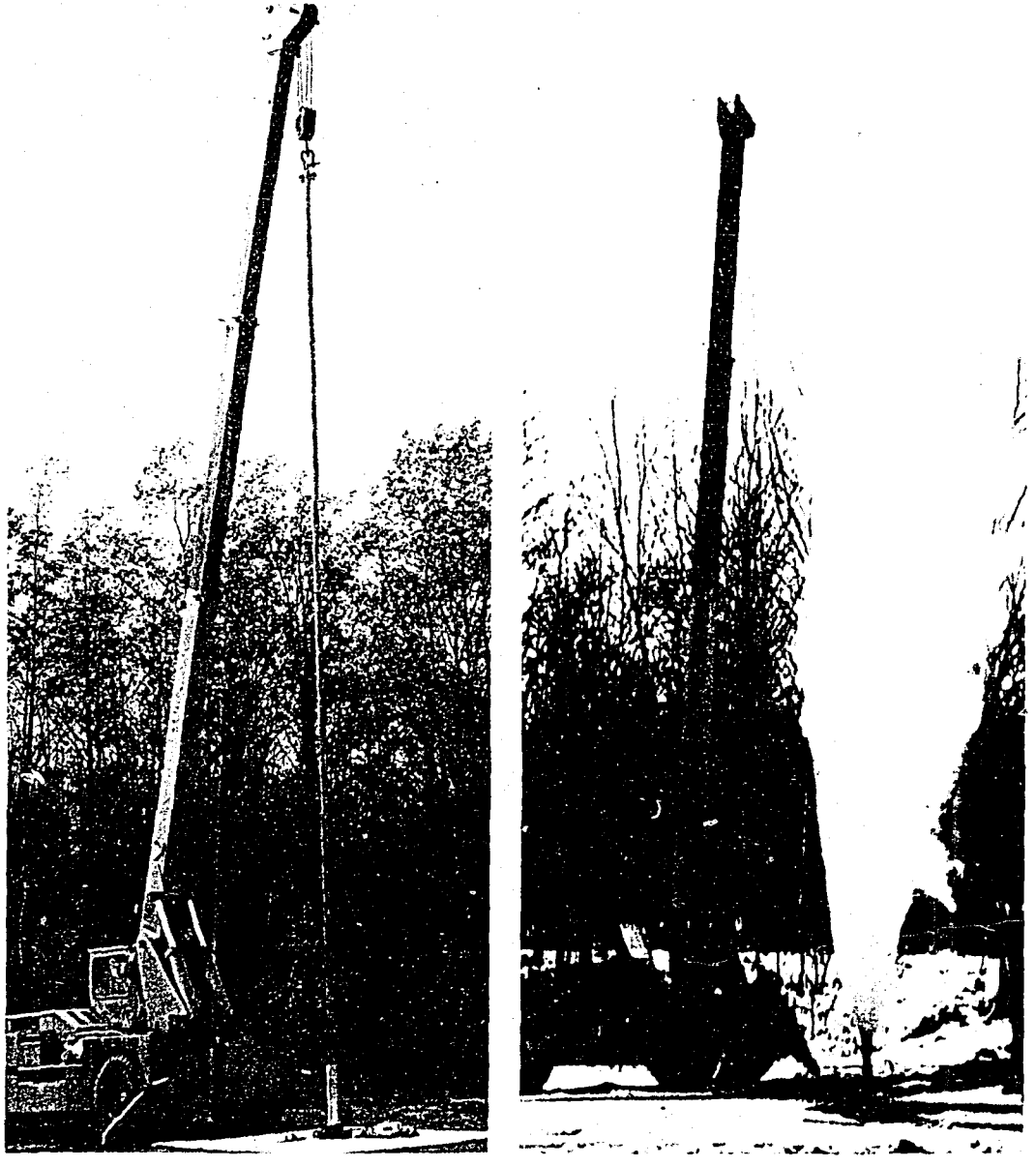
HELIUM SIDE	METRIC	ENGLISH
INLET TEMPERATURE	950°C	1742°F
OUTLET TEMPERATURE	700°C	1292°F
INLET PRESSURE	39.4 b	571 psia
MAX. PRESSURE DROP	0.24 b	3.5 psi
MASS FLOW RATE	0.202 kg/sec	0.466 lbm/sec

PROCESS SIDE		
INLET TEMPERATURE	450°C	842°F
REFORMING TEMPERATURE	825°C	1517°F
OUTLET TEMPERATURE	600°C	1112°F
OUTLET PRESSURE	40 b	580 psia
MAX. PRESSURE DROP	6 b	87 psi
METHANE INLET FLOW RATE	98 kg/h	216 lbm/h
STEAM INLET FLOW RATE	321 kg/h	709 lbm/h
H_2O/CH_4 INLET MOLE RATIO	3	

DUPLEX REFORMER TUBE DATA		
MIN. TUBE I.D.	90 mm	3.54 inch
TUBE LENGTH	12 m	39 feet

FIG. 6.

DUPLEX TUBE ASSEMBLY



INSTALLATION

DETNAFORMING

FIG. 8.

TEST PLAN

HELIUM SIDE

INLET TEMPERATURE . . . . . 950°C
INLET PRESSURE . . . . . 40 BAR
MASS FLOW RATE . . . . . 0.1 TO 0.46kg/sec

PROCESS SIDE

INLET TEMPERATURE . . . . . 500°C
INLET PRESSURE . . . . . 35 BAR

	TEST RUN								
	1	2	3	4	5	6	7	8	9
REFORMING TEMPERATURE °C	⩽810	810	810	810	810	810	825	825	825
METHANE INLET FLOW RATE Nm^3/h	150	125	100	75	125	125	125	125	125
STEAM INLET FLOW RATE kg/h	410	342	273	205	256	171	342	256	171
H_2O/CH_4 INLET MOLE RATIO	4:1	4:1	4:1	4:1	3:1	2:1	4:1	3:1	2:1

FIG. 9.

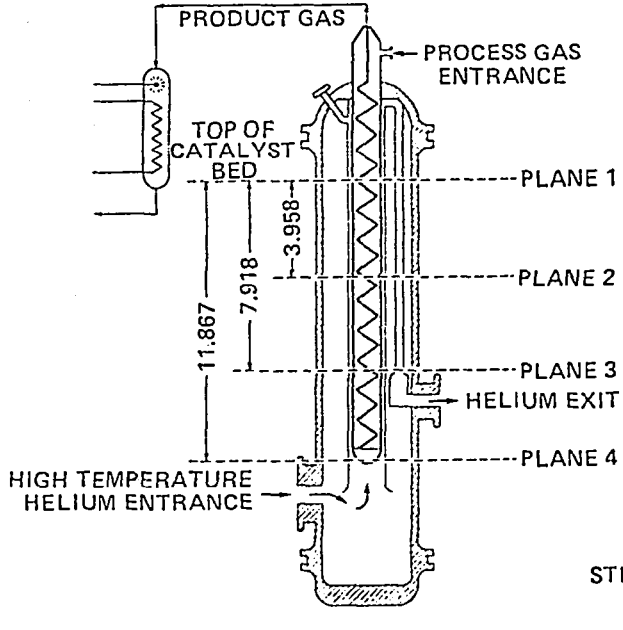
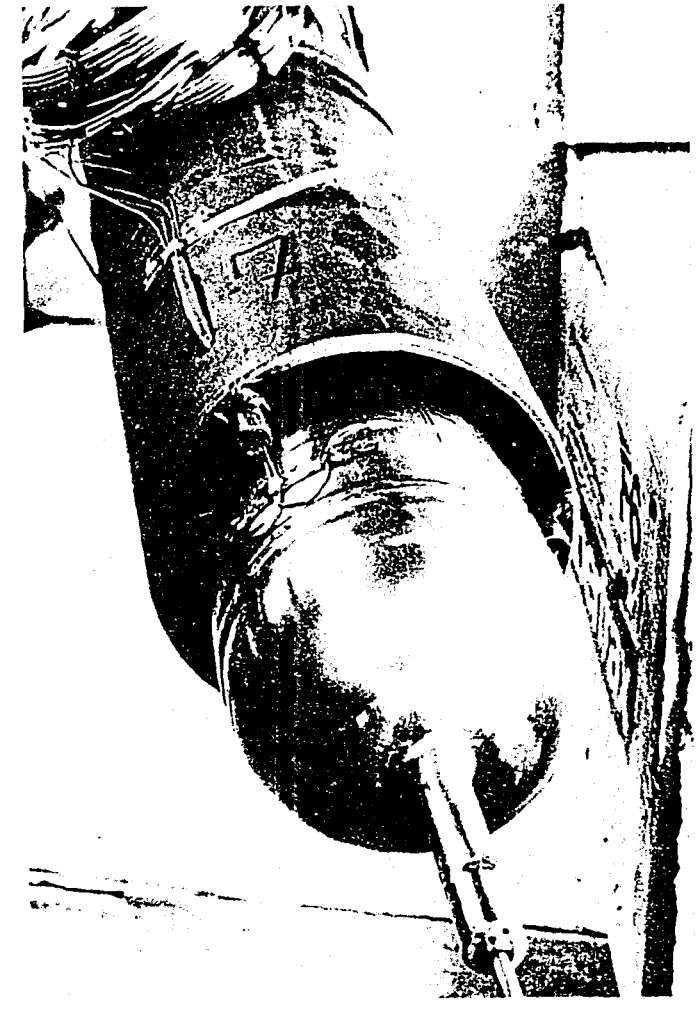


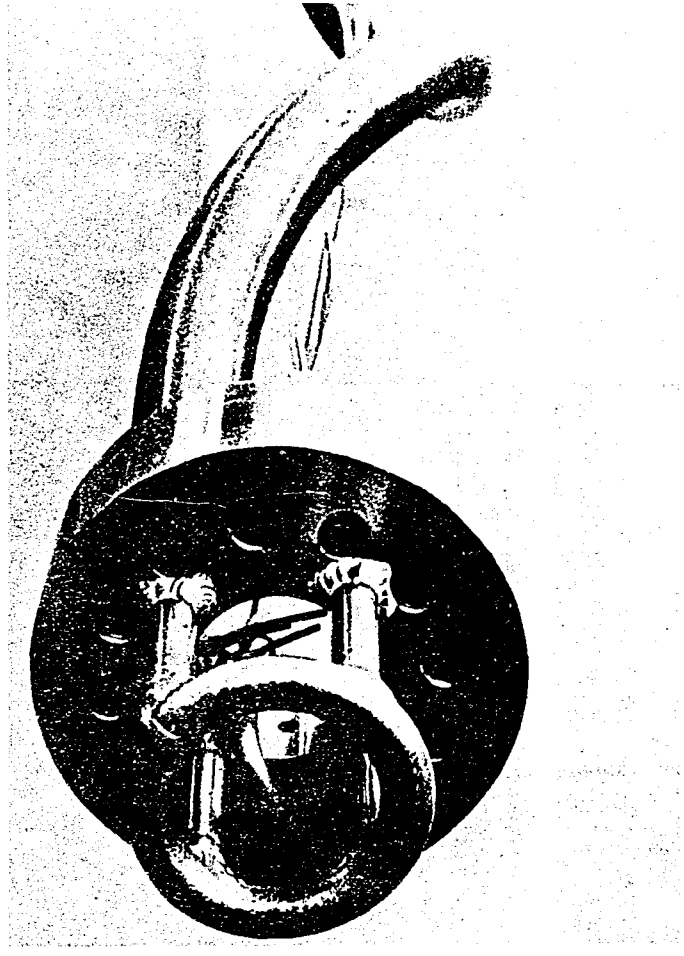
FIG. 10.

STEAM REFORMER TUBE IN EVA



INSTRUMENTED DUPLEX TUBE TEST SECTION

FIG. 11.



PIGTAIL AND
INSTRUMENTATION
SUBASSEMBLY

FIG. 12.

RESULTS OF DUPLEX STEAM REFORMER TESTS IN EVA

H_2O/CH_4	1.87	2.82	3.56	3.81
MEASUREMENT POINT	475	477	478	470
POWER TRANSFERRED (Kw)				
TOTAL	197	231	222	253
STORED IN CHEMICAL REACTION	166	185	180	206
ENERGY BALANCE (%) (HELIUM ΔH/PROCESS ΔH)	97.3	93.1	90.0	93.6
FLOWRATES (kg/sec)				
HELIUM	0.2380	0.4291	0.4308	0.3325
STEAM	0.04679	0.07025	0.07444	0.09274
NATURAL GAS	0.02806	0.02791	0.02346	0.02730
TEMPERATURES (°C)				
HELIUM				
PLANE 1	796.8	829.5	817.0	817.7
PLANE 4	947.7	921.3	901.7	949.6
PROCESS GAS				
PLANE 1	509.5	596.6	502.9	515.8
PLANE 4	825.9	826.2	823.1	825.8
PIGTAIL				
PLANE 1	649.3	662.4	660.5	658.6
PLANE 4	825.9	826.2	823.1	825.8
METHANE CONVERTED (%)	58.9	66.7	73.9	76.3

FIG. 13.

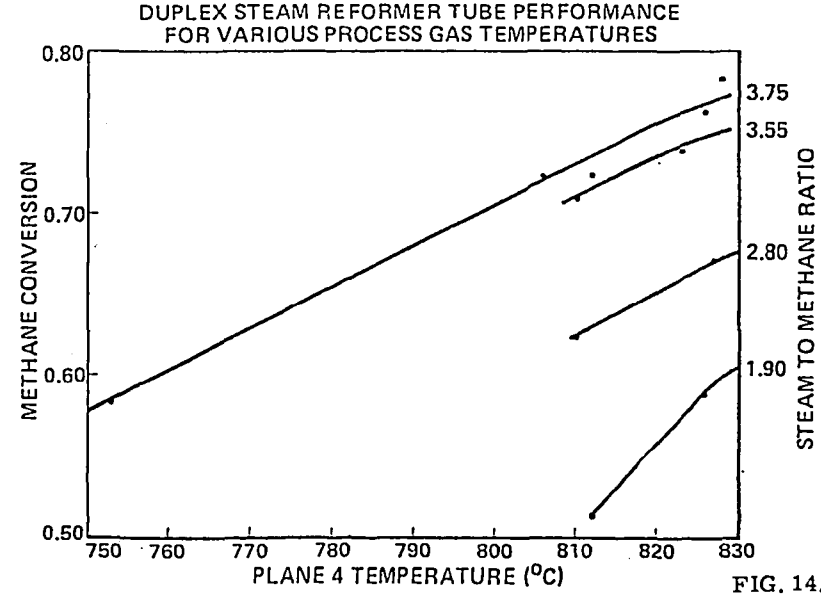


FIG. 14.

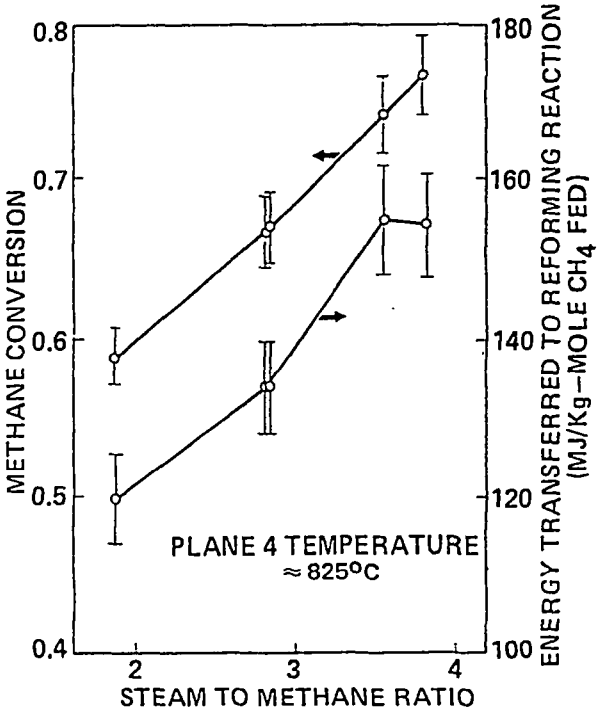


FIG. 15.

DUPLEX STEAM REFORMER TUBE PERFORMANCE FOR METHANE CONVERSION

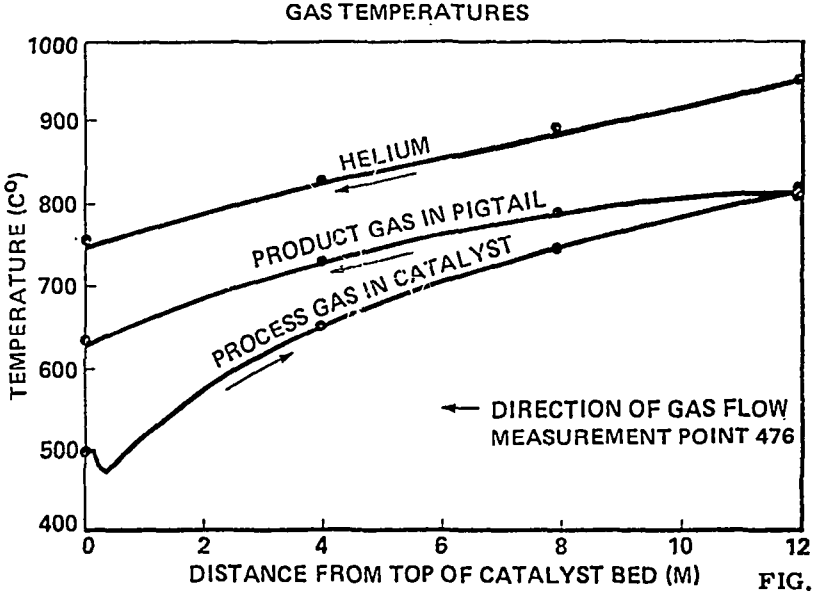


FIG. 15. A

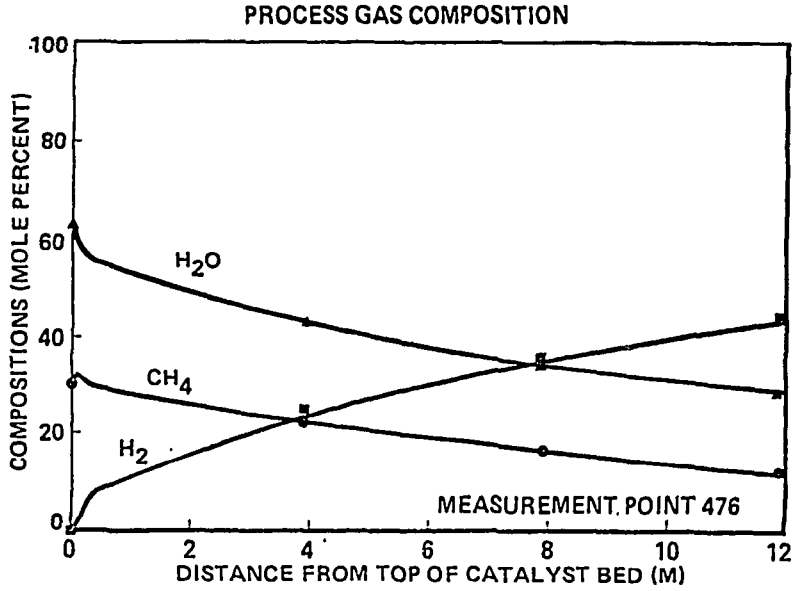


FIG. 15. B

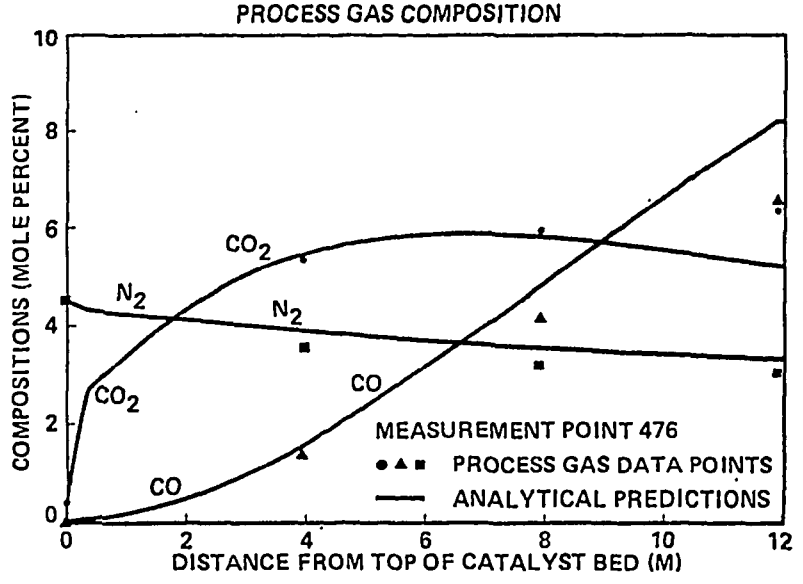
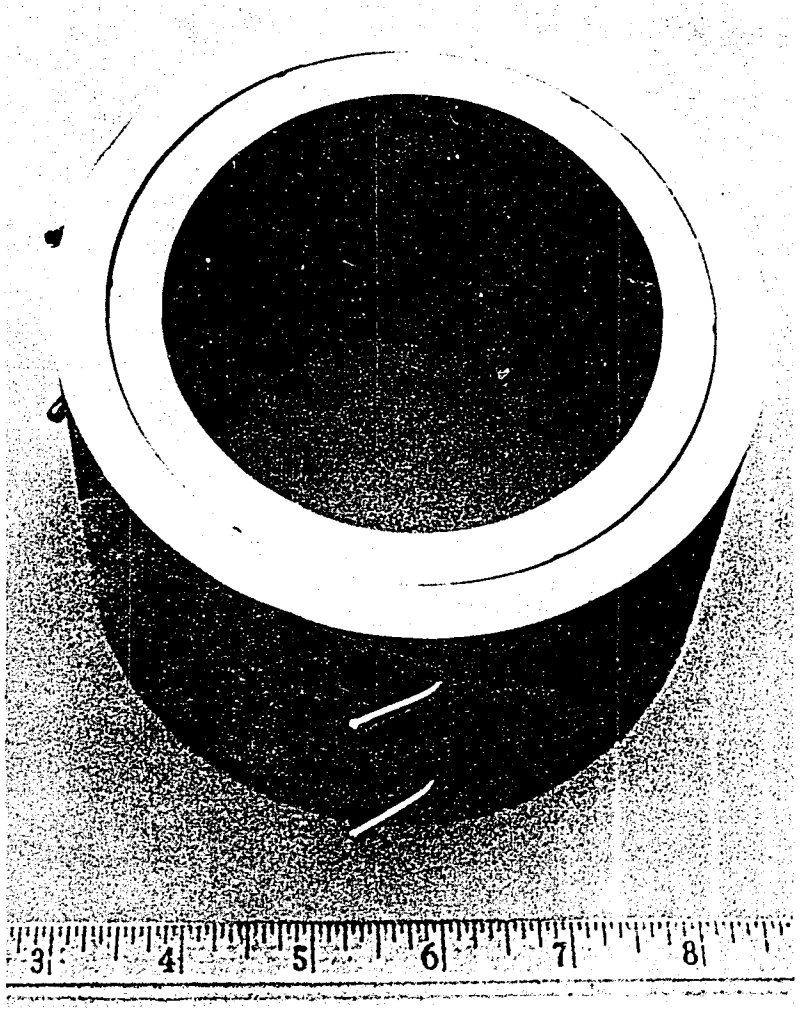
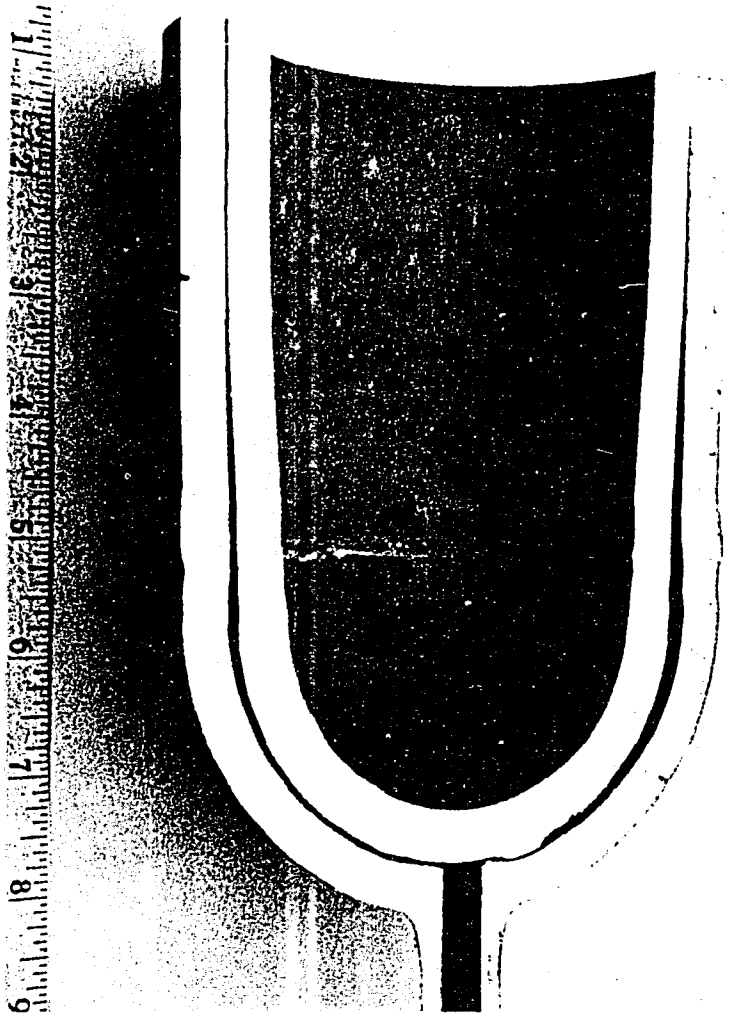


FIG. 15. C



SECTION THROUGH
MIDDLE OF
DUPLEX TUBE

FIG. 16.



BOTTOM SECTION
WITH END CAPS
AND NOZZLE

FIG. 17.