

PERFORMANCE OF A REFORMED NATURAL GAS-ACID  
FUEL CELL SYSTEM

Part I. Hydrogen Generator Design

by

John Meek  
B. S. Baker  
A. C. Allen

Institute of Gas Technology  
Chicago, Illinois

INTRODUCTION

Natural gas can be used in low-temperature fuel cell systems in several ways. Although the direct methane cell has proven feasible,<sup>1,2</sup> present and foreseeable technology of such systems will be very costly because of the quantity and type of catalyst required for direct anodic oxidation. The indirect cell, which requires the reforming of methane followed by the utilization of hydrogen, seems more attractive at the present time. With alkaline electrolyte systems it is necessary to use high-purity hydrogen as the fuel. This may be obtained by steam reforming in an external reformer and purifying the gas with a palladium-silver diffuser, or by reforming the gas in situ in a fuel cell that employs a palladium-silver-hydrogen diffusion anode. For application in the gas industry, it is essential that both first costs and operating costs remain low. Accordingly, the program at IGT has focused its attention on development of a hydrogen generator-fuel cell system in which the feed from the reformer is rich in hydrogen but unpurified, the enrichment being achieved by conventional chemical processing techniques. It is realized that this implies the use of a fuel cell of the acid electrolyte type which is presently more expensive than the alkaline electrolyte fuel cell. Nevertheless we believe that the cost of the acid electrolyte cell can be greatly reduced especially where the fuel is hydrogen. The other factors affecting the decision to pursue this system have been outlined in an earlier publication in this series.<sup>3</sup> Design of a natural gas-fueled hydrogen generator is described here in which the product is suitable for use in any acid electrolyte fuel cell.

HYDROGEN-GENERATION PROCESS

The hydrogen generation process used in the IGT system has been described in detail.<sup>3</sup> It consists of three stages: First, natural gas is steam-reformed at 800°C to produce a hydrogen-carbon

monoxide-carbon dioxide mixture. The effluent from this first reactor is then cooled and fed to a carbon monoxide shift reactor operating at about 270°C, wherein the carbon monoxide content of the gas is reduced from a typical 15% (dry basis) to about 2000 parts per million. For certain acid fuel cells operating above 100°C this gas might prove to be an acceptable feed, but, for lower temperature cells it is desirable to further reduce the carbon monoxide content of the feed gas. This is achieved in the last stage by passing the effluent from the carbon monoxide shift reactor through a low-temperature (190°C), selective methanation reactor in which the carbon monoxide content of the gas is reduced to approximately 20 parts per million by reaction with hydrogen to produce methane. Typical gas compositions along with the free energy for these reactions are summarized in Tables 1 and 2. It can be seen that the reforming and carbon monoxide shift reactions proceed, for practical consideration, to equilibrium. The reaction based on the methanation of carbon monoxide alone is a long way from equilibrium and improvement might be expected. The large deviation from equilibrium is probably due to reaction of carbon dioxide with hydrogen to produce carbon monoxide.

#### HYDROGEN GENERATOR DESIGN

For the hydrogen generation process to be attractive it is necessary that an integrated three-stage, gas-fired hydrogen generator that is self-controlling be designed and constructed. Limited data is available on the design of complete hydrogen generation systems with small capacity.<sup>4,5</sup>

In the IGT system an annular reactor design was chosen because it represented a readily packageable easily fabricated unit with low pressure drop.

The first problem encountered in the design was the development of a gas-fired burner that could operate on low-pressure gas (6 in. wc), bring the relatively compact reformer reactor to operating temperature, and maintain the desired heat input to sustain the endothermic steam-reforming reaction. It was found that reactors of the present design, or of almost any design, did not have sufficient heat transfer area to permit heating by a convective process. To obtain a higher effective heat transfer coefficient, both sides of the annular reactor were encased in annular sections filled with the refractory material of high surface area. The sections could be brought to the desired temperature by convection and would in turn heat the reformer by radiation from the refractory. A typical burner design for this system is shown in Fig. 1. The reforming stage of the hydrogen generator, which is capable of reforming 25 cu ft of methane per hour, is shown in Fig. 2.

The second problem encountered in the present system was to find a means for reducing the temperature of the hot flue gases to maintain the second- and third-stage reactors at their proper operating temperatures. The solution was to cool both the flue gas from the natural gas burner and the product gas from the reformer reactor by generating and superheating process steam. The flue gas and the product gases are effectively cooled by this procedure but the achievable temperature control was not accurate

enough for the shift and methanation stages. Obtainment of temperature control in these two reactors within the desired limits, was made possible by jacketing the reactors with constant boiling fluids. Dow-Therm A and Dow-Therm E have been chosen for this purpose. The jacketed shift and methanation units are shown in Figs. 3 and 4. Temperature control of these reactors is achieved by dependence of pressure on temperature of the Dow-Therm liquid-vapor systems. The Dow-Therm vapor pressurizes a Sylphon bellows which transmits a pressure signal to actuate damper valves which control the direction of flow of hot flue gases. Water flow valves which, in turn, control the flow of cooling water to the Dow-Therm vapor condensers also activated by this means.

This mode of control is especially suited to a hydrogen generator that supplies a constant amount of product gas. The Sylphon bellows (Fig. 1) has the further advantage of not requiring any parasitic power from the fuel cell system; it is also relatively inexpensive. The only auxiliary power on the present reformer is that needed for a small air blower which provides combustion air. This blower draws about 40 watts at maximum air demand. The complete system is shown schematically in Fig. 5. The actual unit with a 100 cu ft/hr capacity is shown in Fig. 6.

#### PERFORMANCE CHARACTERISTICS

Hydrogen generators very similar in design to the one described in the present paper have been successfully operated at IGT, producing the gas given in Table 1. The best overall efficiency achieved with this type reformer in 150 hours of intermittent operation has been 41%. This figure was obtained with a unit that has an output of 50 cu ft of hydrogen per hour. The system presently under test has an output capacity of 100 cu ft of hydrogen per hour; overall efficiency figures of about 50% are anticipated.

Gas of the type produced by this hydrogen generator has been extensively tested in low-temperature acid fuel cells. Use of these gases is the subject of Part II of this paper.

#### ACKNOWLEDGMENT

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Table I

Composition, Mole percent

Reactor	H <sub>2</sub>	CO <sub>2</sub>	CH <sub>4</sub>	CO	H <sub>2</sub> O	N <sub>2</sub>	T°C	$\frac{\Delta F}{RT}$
Reformer In	0	0.107	*	0	87.268	0.046	800	-∞
Reformer Out	34.644	3.773	0.045	6.847	54.654	0.036	800	-7.287
Shifter In	34.644	3.773	0.045	6.847	54.654	0.036	270	-5.18211
Shifter Out	41.346	10.475	0.045	0.146	47.952	0.036	270	-2.30975
Methan. In	41.346	10.475	0.045	0.146	47.952	0.036	190	-26.50958
Methan. Out	41.030	10.505	0.190	0.001	48.236	0.036	190	-20.05782

\*See Table II

Table II

Fuel	CH <sub>4</sub>	C <sub>2</sub> H <sub>6</sub>	C <sub>3</sub> H <sub>8</sub>	C <sub>4</sub> H <sub>10</sub>	C <sub>5</sub> H <sub>12</sub>	C <sub>6</sub> H <sub>14</sub>	C <sub>7</sub> H <sub>16</sub>	N <sub>2</sub>	CO <sub>2</sub>
Comp, mole %	12.027	0.394	0.097	0.033	0.009	0.010	0.009	0.046	0.107

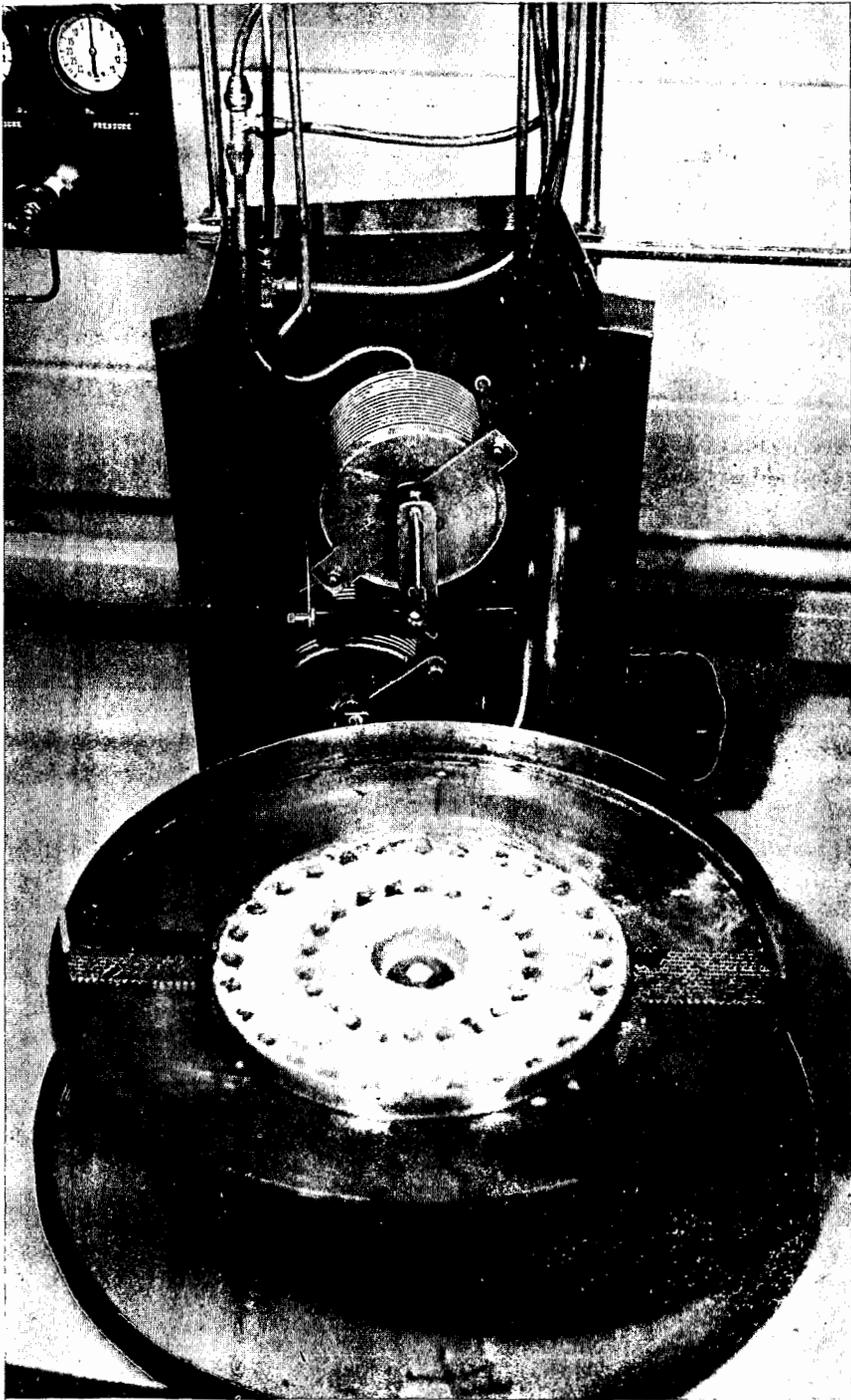


Fig. 1. -NATURAL GAS BURNER USED AS PRIMARY HEAT SOURCE FOR HYDROGEN GENERATOR .

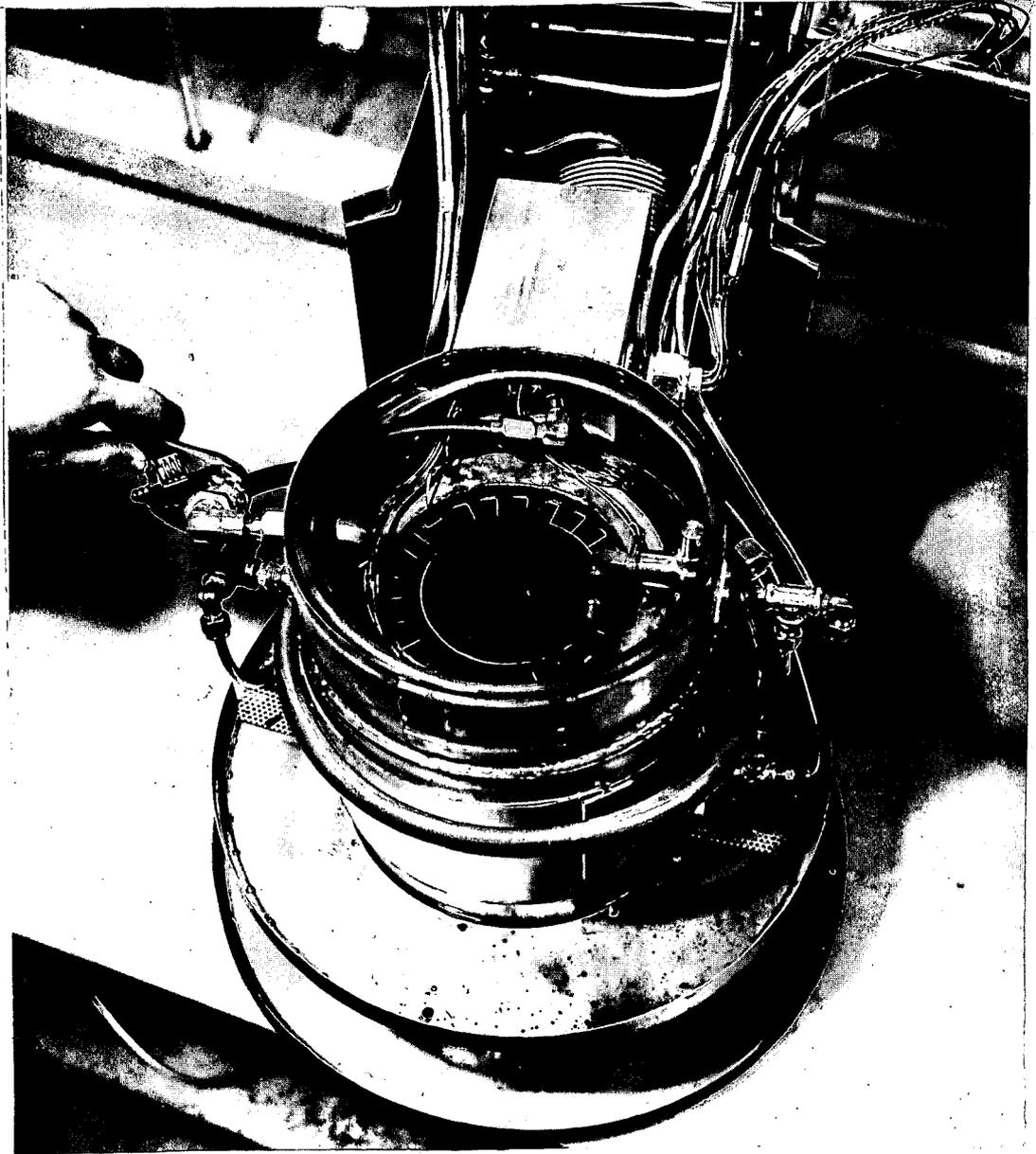


Fig. 2.-REFORMER AND CONTROL SECTION OF THE HYDROGEN GENERATOR

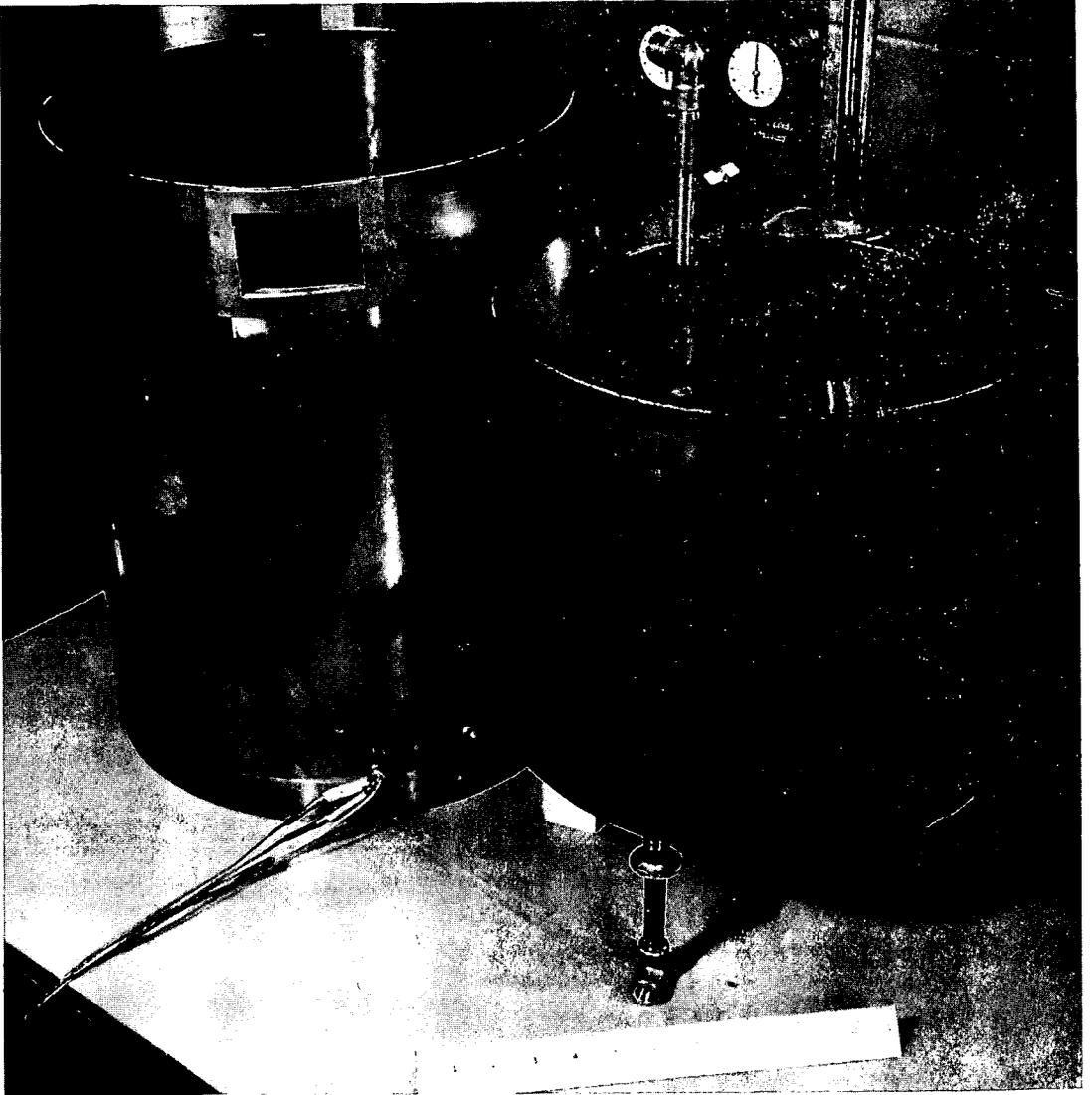
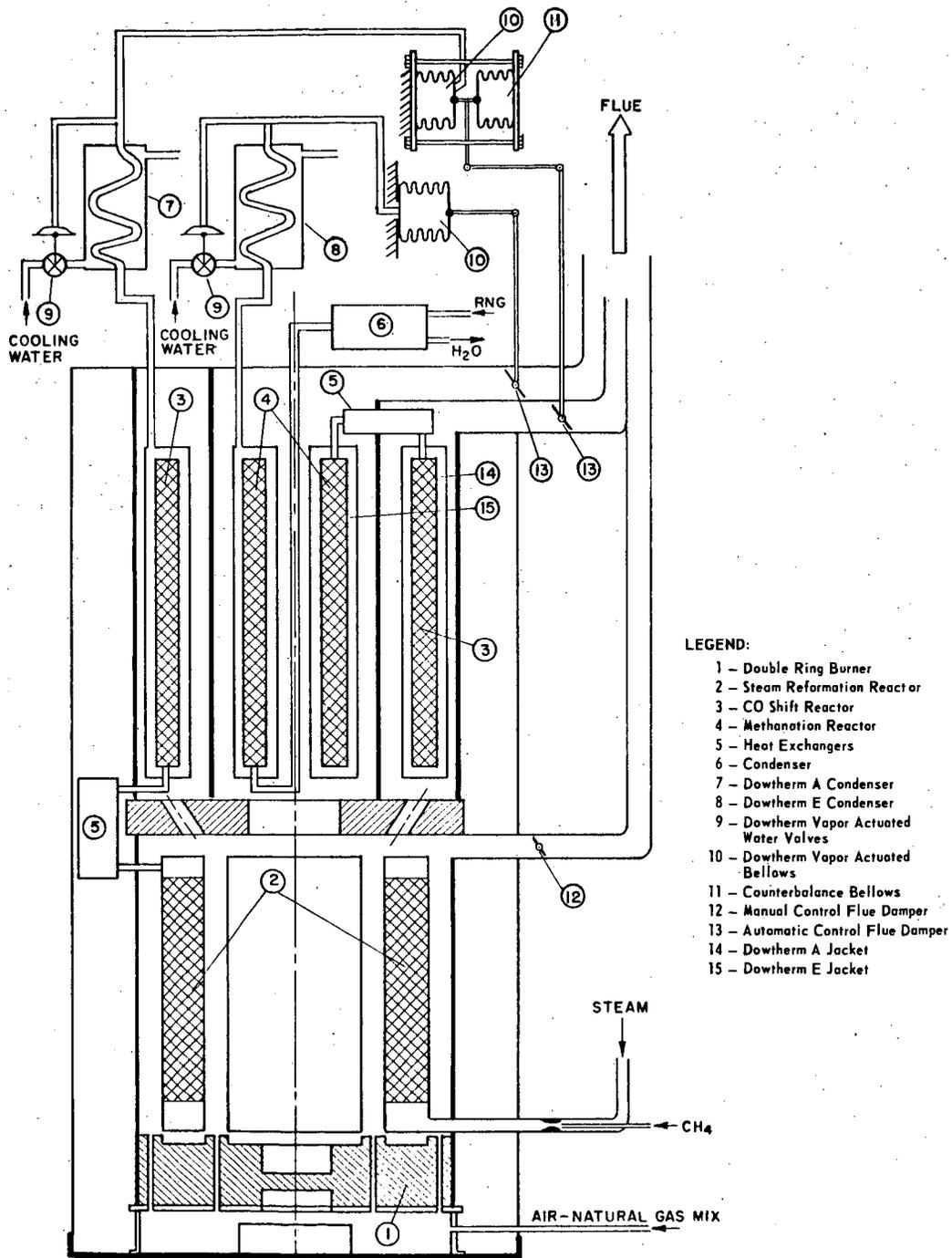


Fig. 3.-JACKETED CARBON MONOXIDE SHIFT REACTOR



Fig. 4.-JACKETED METHANATION REACTOR



LEGEND:

- 1 - Double Ring Burner
- 2 - Steam Reformation Reactor
- 3 - CO Shift Reactor
- 4 - Methanation Reactor
- 5 - Heat Exchangers
- 6 - Condenser
- 7 - Dowtherm A Condenser
- 8 - Dowtherm E Condenser
- 9 - Dowtherm Vapor Actuated Water Valves
- 10 - Dowtherm Vapor Actuated Bellows
- 11 - Counterbalance Bellows
- 12 - Manual Control Flue Damper
- 13 - Automatic Control Flue Damper
- 14 - Dowtherm A Jacket
- 15 - Dowtherm E Jacket

Fig. 5.-SCHEMATIC REPRESENTATION OF IGT HYDROGEN GENERATOR SHOWING REACTORS AND CONTROL LOOPS

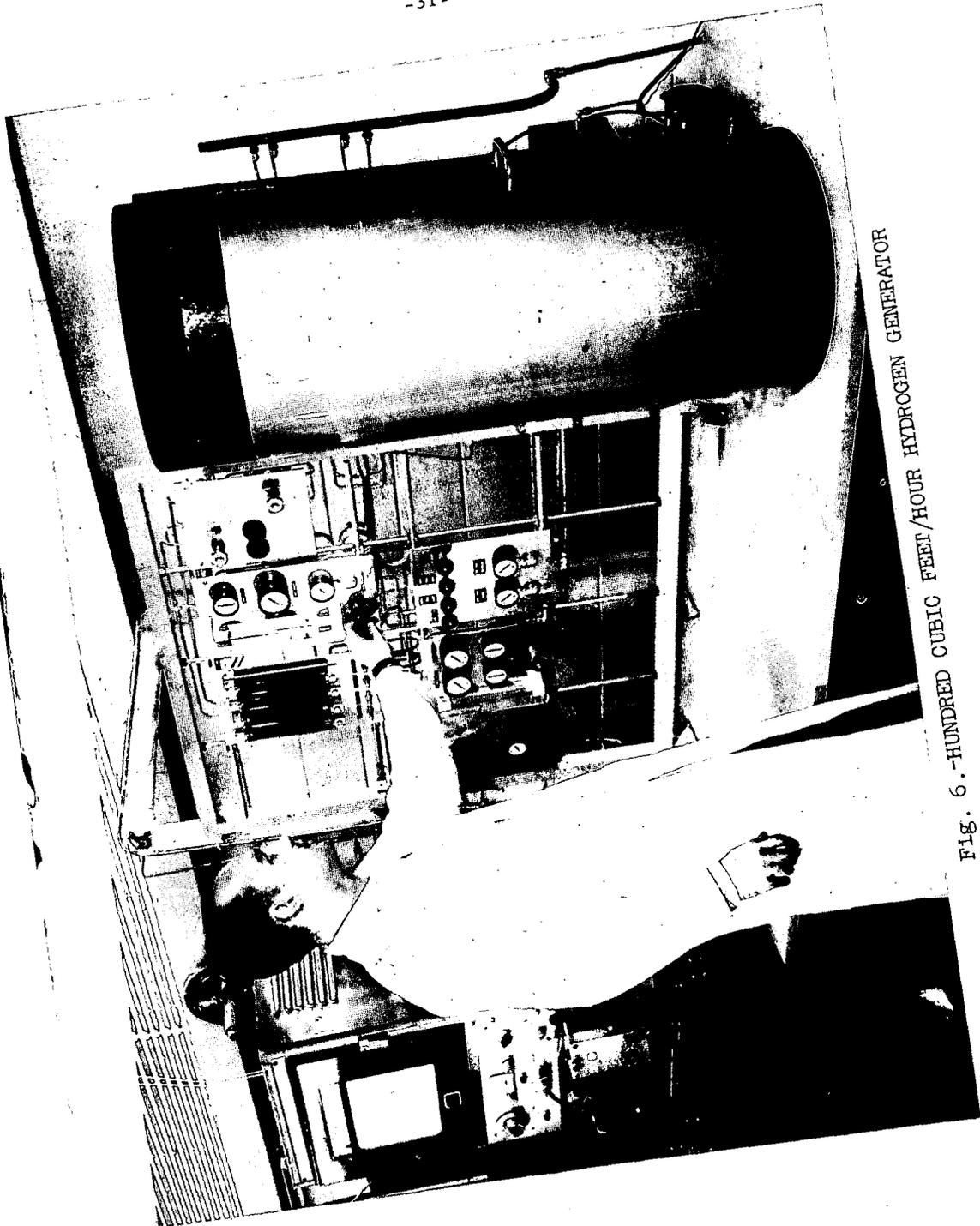


FIG. 6. -HUNDRED CUBIC FEET/HOUR HYDROGEN GENERATOR