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FINAL REPORT

AN INVESTIGATION OF THE REDUCTION OF CARBON DIOXIDE IN A SILENT ELECTRIC DISCHARGE

Prepared Under Contract NAS 2-9551

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
AMES RESEARCH CENTER
Moffett Field, California 94035

Biotechnology
LOCKHEED MISSILES & SPACE COMPANY, INC.
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Acknowledgements

The experimenter and author wishes to thank Barbara Greenough for her advice and encouragement during the laboratory study and for her assistance in reviewing and editing this Final Report.

Barbara Greenough is the contributor of the Feasibility Study which has concluded our investigation.

The contract's Technical Monitor was P. D. Quattrone, Chief, Advanced Life Support Project Office, NASA Ames Research Center, Moffett Field, California.

CONTENTS

Summary

An investigation of the reduction of CO_2 to O_2 and CO in a Silent Electric Discharge was undertaken with the intent of correctly understanding the process and operating it at optimum efficiency. The study showed that current alone (in the ionized plasma induced by the silent electric discharge) was reponsible for the CO_2 reduction process. Voltage and frequency were important only in so far as they induced current in the plasma. Pressure and temperature are minimmally influential in the process. Flow rate is important only in that gas residence time in the plasma is proportionally related to CO_2 reduction.

The large power consumption in the process was recognized as resulting from the low power factor of the reactor vessel which electrically behaves like a capacitor. The power factor was subsequently improved by adding an inductive element to make the reactor vessel capacitance part of a resonant circuit. It was found that the CO_2 reduction process was most efficient in terms of power vs reduction rate when a voltage was employed that was only slightly higher than that needed to induce the plasma. The study concluded with a demonstration that with this apparatus one liter of CO_2 could be reduced with an expenditure of less than 360 kilojoules (100 watt hours) of energy.

Purpose of the Investigation

This study of carbon dioxide reduction in a silent electric discharge was carried out under NASA contract NAS 2-7551. This contract was awarded to the Biotechnology Department of Lockheed Missiles and Space Company in response to Lockheed's proposal LMSC/D554881, in which a method of studying the subject was outlined.

It was the ultimate purpose of the investigation to determine whether or not the reduction of CO_2 in a silent electric discharge is efficient enough in terms of power consumed per volume of CO_2 reduced to encourage development of a practical equipment which could be used in manned spacecraft to recover O_2 from respired CO_2.

The initial inspiration for the concept derives from studies reported in Russian literature[(1)(2)]. In those studies CO_2 gas was passed through a double-walled, concentrically constructed glass vessel with metallic electrodes plated on the exterior walls of the vessel. A high voltage alternating current was applied to the electrodes, causing the phenomenon of a "silent electric discharge" in the region through which the gas was passing. A percentage of the gas, (up to 30%), was reduced to O_2 and CO, however at the penalty of extremely high power consumption for the amount of O_2 recovered. Despite this, the process looked interesting because of the low temperatures and low pressures required.

A Lockheed-supported preliminary investigation of the method was carried out in 1972. A glass reactor vessel of the general type described in Russian literature was constructed and an experiment closely conforming to the Russian method was carried out. In this early study efficiencies were lower than those reported by the Russian workers, power consumption being higher and CO_2 reduction rates being lower.

The present study was undertaken with the intent of accurately monitoring the parameters of the experiment and attempting to improve the efficiency of the process through proper understanding and management of the electrical forces involved.

The report which follows will show that once the electrical events mediating the process were correctly understood, an encouragingly high efficiency could be established in the reduction of CO_2 in a silent electric discharge.

The Silent Electric Discharge Reactor Vessel

The reactor vessel used in this experiment was constructed in 1972 in conformance with a type variously reported in Russian technical literature and described as an "ozonator" or "ozonizer". Figure 1 clearly shows the construction of the vessel. It consists basically of two concentric glass cylinders having walls 2 mm thick sealed together at the ends, and provided with inflow and outflow tubes at the extreme ends. Silver electrodes are plated on the outside diameter of the exterior cylinder and on the inside diameter of the interior cylinder. An air gap of approximately 1.3 millimeters exists between the cylinders, and it is in this gap that the "silent electric discharge" occurs.

Experience with this specific reactor vessel shows that at 40.0 kN/m^2 (300 mmHg) CO_2 pressure the phenomenon of the silent electric discharge does not occur until the applied AC voltage reaches a level of about 3200 volts, regardless of the frequency. Above that level the carbon dioxide gas becomes partially ionized and conductive. This ionized gas is referred to as the "plasma" in this report. Figure 2 is a photograph of the voltage and current curves occurring during typical operation with CO_2 gas flowing through the reactor vessel. The voltage trace is the sharp trace, while the current trace is seen to lead by almost 90 degrees, and has a "fuzzy" high frequency component riding on the extreme excursions. This "fuzz", when expanded on the oscilloscope, is seen to consist of a dense sequence of sharp spikes, and evidently represents gas conduction occurring as a series of almost instantaneous, very frequent and closely spaced events during each extreme swing of the current waveform. This general phenomenon has been studied and reported in the literature (3), and further analysis will not be attempted in this report.

It should be noted that the reactor vessel is in fact an electrical capacitor, and as such, a current lead of 90° is to be expected in the absence of any resistive component. In cases where the oscilloscope shows less than a 90° current lead (as in Figure 2), it is assumed that a resistive component caused

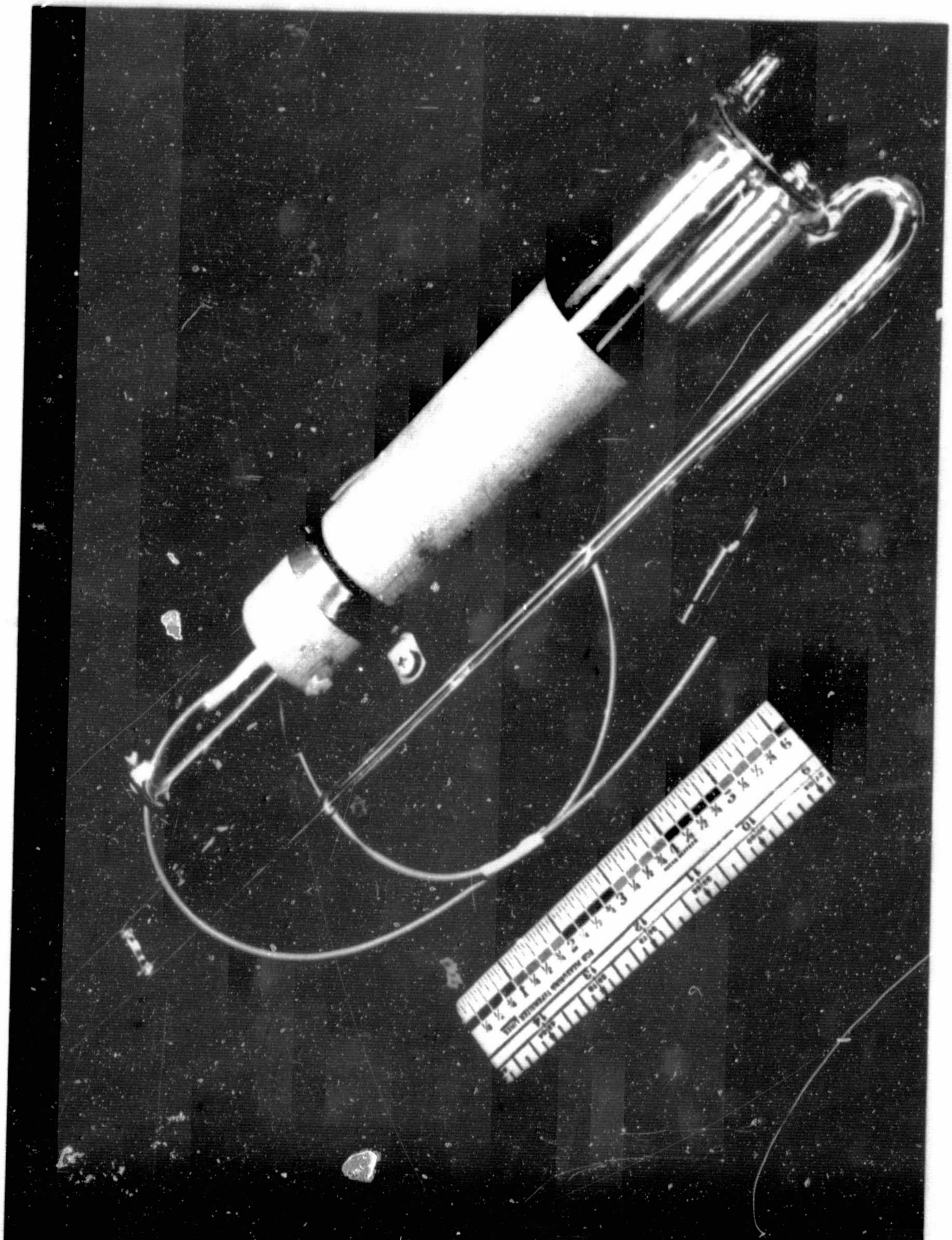


Figure 1 The Reactor Vessel

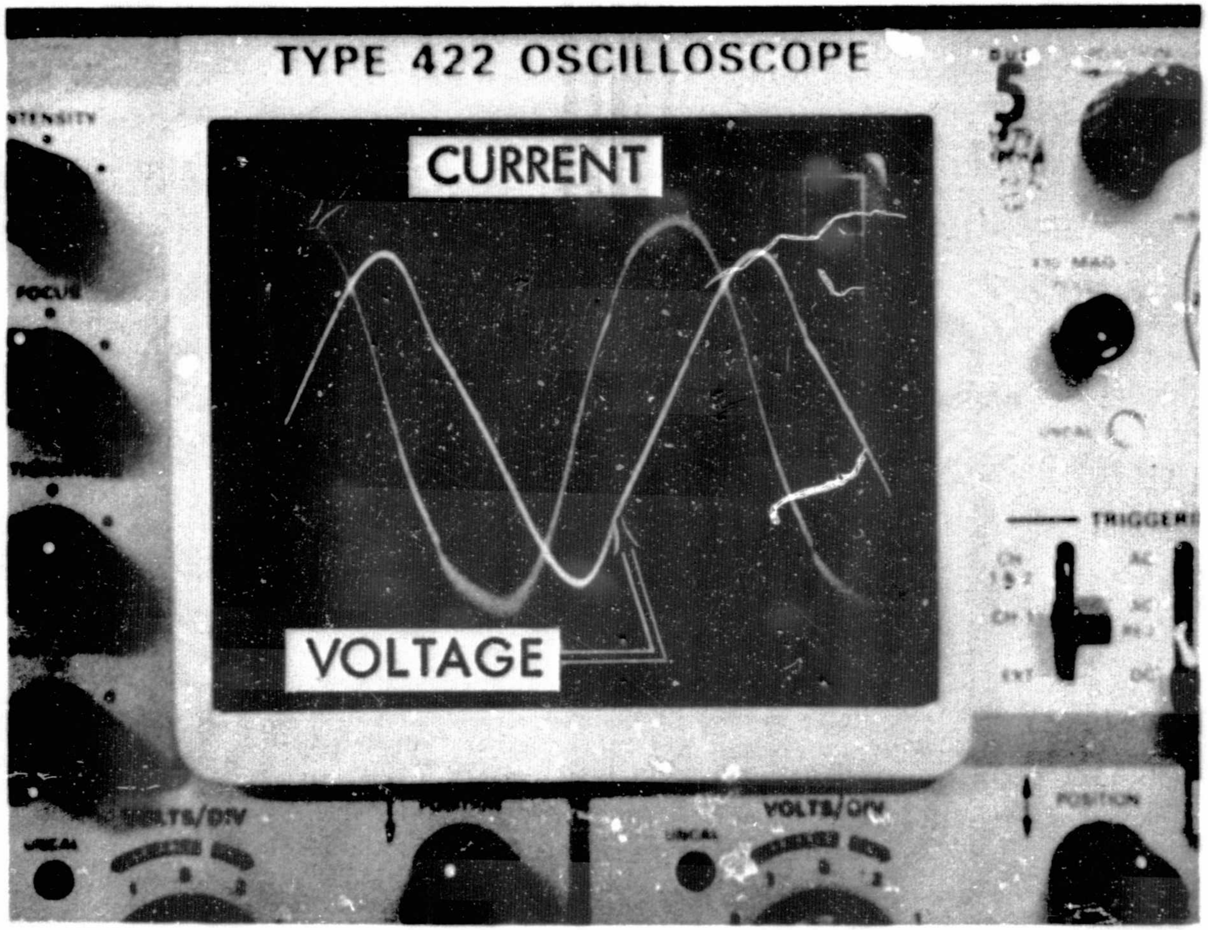


Figure 2 Voltage and Current in the Reactor Vessel

by the work of reducing CO_2 to O_2 and CO enters in, and that the true power required by this CO_2 reduction process may be computed by multiplying AC volts times AC amperes times power factor, where power factor equals the cosine of the phase difference in degrees as follows:

$$\text{True power} = \text{power factor} \cdot V_{ac} \cdot A_{ac}$$

$$\text{Power factor} = \text{cosine } \phi$$

Experience with the collected data showed this assumption to be correct.

Since the reactor vessel is a capacitor, it was thought advisable to measure the capacitance. Additionally, since ionization of the gas was assumed to partially or completely short-circuit the air gap, it was also desirable to fill the reactor with a conductive fluid (salt water) to simulate that condition. An Electro Scientific Industries 250 DE Impedance Bridge was used, and the following values were measured.

Minimum capacitance = 105 picofarads
Maximum capacitance (shorted) = 246 picofarads

The partial short-circuiting of the air gap did occur, as is shown by the calculated values of capacitance shown in tables of experimental data, and this shorting was greater with higher electrical currents. Capacitance was calculated as follows:

$$\text{Capacitance in picofarads} = \frac{A_{ac} \cdot 10^{12}}{2\pi \cdot \text{Frequency} \cdot V_{ac}}$$

The Experimental Equipment

The experimental equipment consisted of two groups of apparatus; one group to control and measure the gas through the reactor vessel, and the second group to control and monitor the electrical energy applied to the reactor vessel. This description covers the apparatus in its final and very satisfactory form, since various improvements were made as the experiment proceeded.

Gas Control Apparatus:

Figure 3 diagrams the gas control apparatus. Pure CO_2 gas was supplied by a conventional gas bottle with shutoff valve and a pressure reducing regulator. The gas was then supplied at about 137.9 kN/m^2 (20 PSIA) to a Fairchild model 16 vacuum regulator, used to set the required pressures in the reactor vessel. Gas flow rate was measured by two Brooks flowmeters of the type utilizing a glass tube with a tapered bore and two floating balls of different densities. Brooks model R-2-15-AAA covered a range of 5 ml/minute to 145 ml/minute, while the model R-2-15-AA covered a range of 10 ml/minute to 265 ml/minute, both referred to 101.3 kN/m^2 (760 mmHg) pressure. Since the flow rates were measured inside the vacuum line, it was necessary to correct the readings for the various pressures in accordance with the formula supplied by the manufacturer. Pressure was measured with a Wallace and Tiernan model FA160 absolute pressure gauge, 0 to 30 PSIA. Gas flow was regulated by a Hoke type 1335G4Y micrometer valve. Finally, a Sargent-Welch "Sarvac" model 8804 two-stage vacuum pump was employed to obtain the vacuum and pull the gas through the system. A small fan was used to keep the pump cool, although this was merely a precautionary measure. The gas pumped out of the system was filtered through a Tygon tube packed with fiberglas, to remove oil droplets. The sensing head of the Beckman model 777 oxygen analyzer was fitted snugly into the outlet of the Tygon tube, and a piece of #20 metal wire was run along side of the sensor to make a small space for the gas to escape. A cylindrical plastic container was used to hold the Dupont Freon TF solvent in which the reactor vessel was immersed. This maintained the temperature of the vessel between room temperature and 47.6°C, the boiling point of the solvent, in all experiment runs.

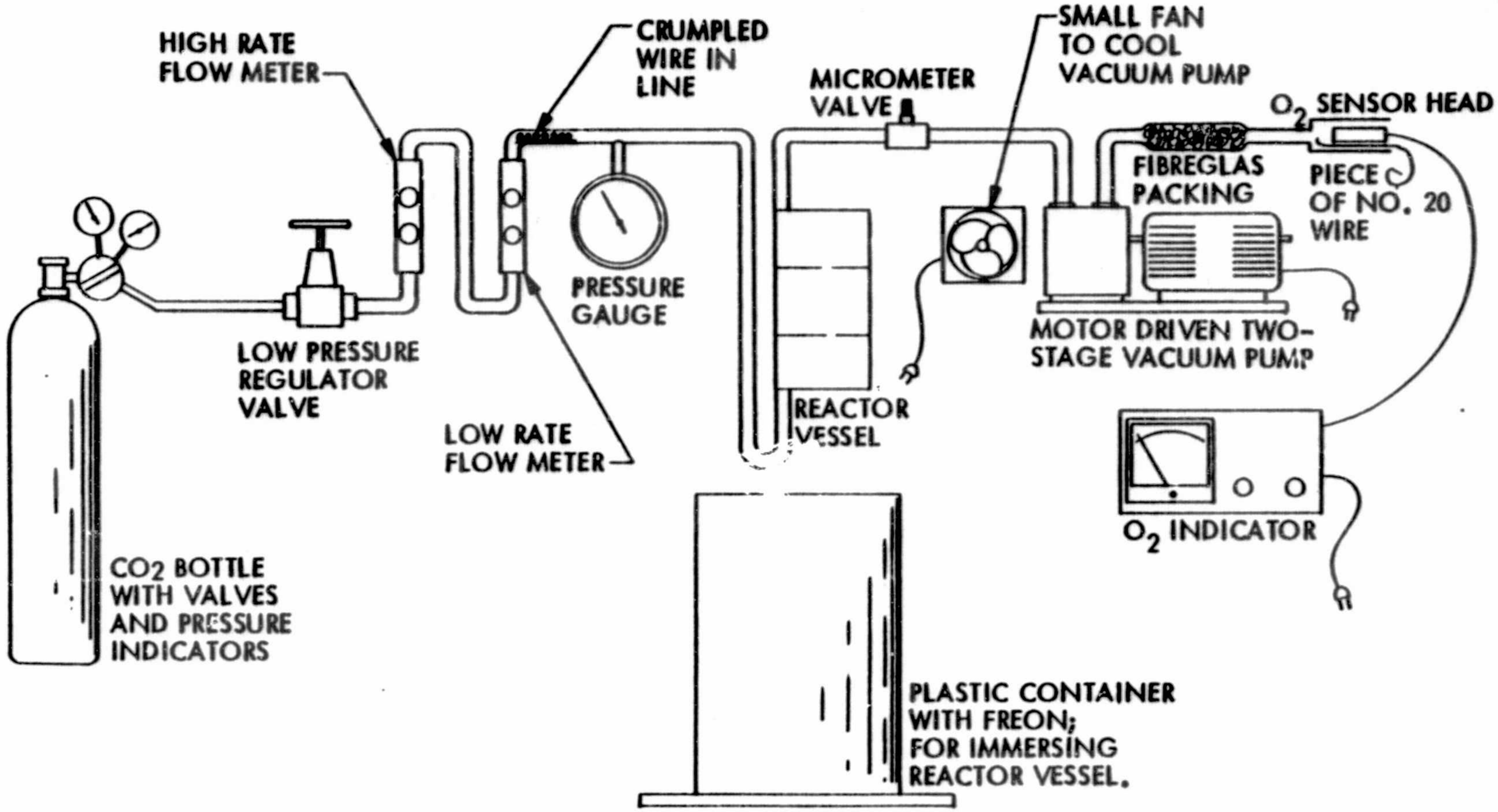


FIGURE 3 GAS CONTROL APPARATUS

Most of the "plumbing" was done with aluminum "Swagelok" ¼" fittings and Imperial Eastman "polyflo" ¼" plastic tubing. Some Tygon tubing of various sizes was used at several points in the test seup, and simply force-fit over the mating element.

Electrical Instrumentation:

The electrical instrumentation is shown in Figure 4. A Hewlett Packard model 200 CD sine wave oscillator served as input to the McIntosh model MI-200 AB 200 watt output audio amplifier. The 4 ohm output of the amplifier supplied the primary winding of the high voltage transformer. A Weston model 433 AC ammeter with 0-2.5 and 0-5 ampere ranges was connected into the primary circuit. A Simpson model 260 multimeter was connected across the transformer primary winding, after the ammeter, to measure applied primary voltage.

Two transformers, Figure 5, were used in the experiment. The large transformer (Elma Engineering 60 Hz, 50 KV) was used for the 60, 150, and 400 Hz frequencies, while the smaller transformer (Electro type E14593B) was used for the 400, 1000, 1700, and 3000 Hz. It was this second transformer that proved most useful, in fact ideal, for this investigation.

The high voltage from the transformer secondary winding was measured by means of a resistor divider made of 24 one-megaohm 2 watt resistor in series, and a pair of 47 kilohm resistors in parallel as the voltage sensing resistance. This appears in Figure 5. A Fluke model 8000 A digital AC voltmeter was used to measure the voltage which had been divided by 1000 in the resistor chain. A 1000 ohm wire-wound 20 watt resistor was used for current sensing, and another Fluke model 8000A digital AC voltmeter measured the voltage drop across the resistor in millivolts, which converted directly to milliamperes. The Fluke meter could have been used to read AC milliamperes directly, however this was not considered safe in the event that the meter failed and presented an open circuit to the reactor vessel wiring.

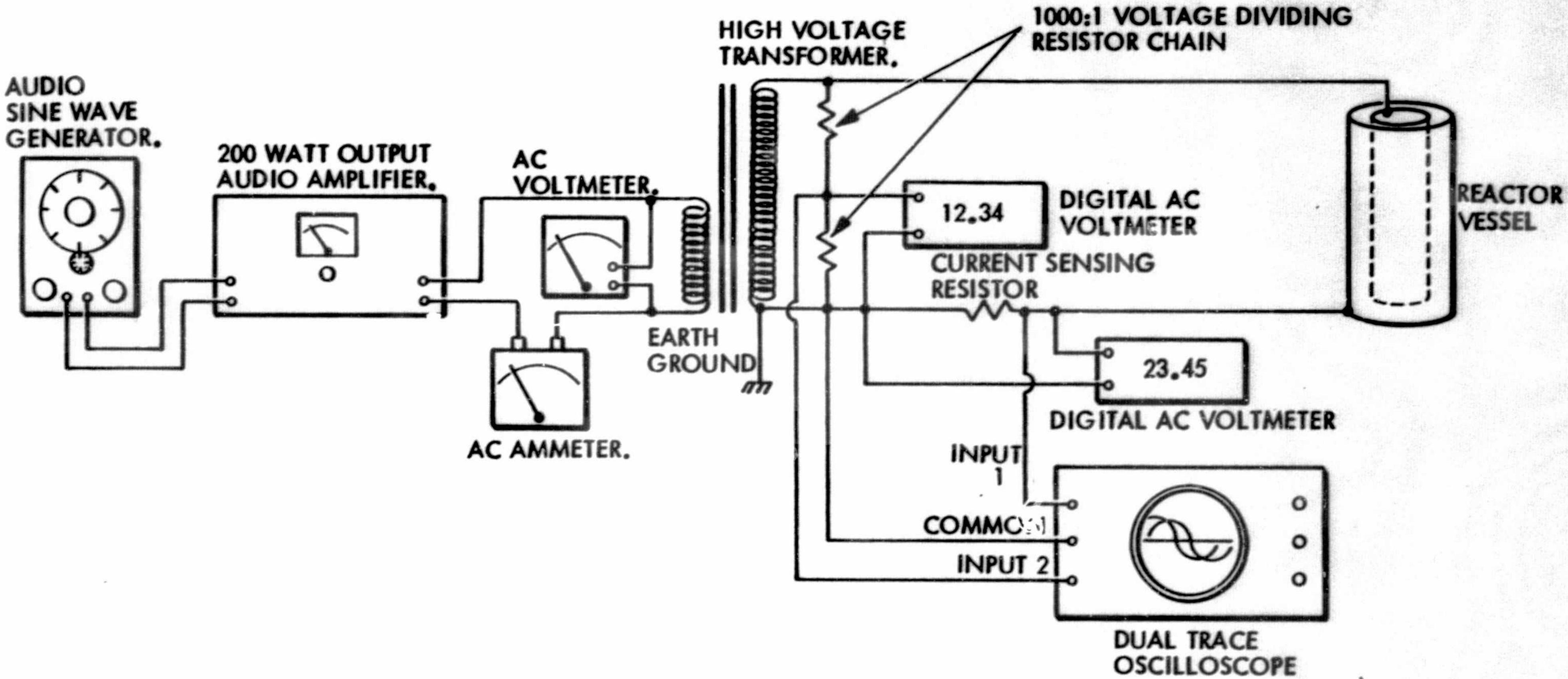


FIGURE 4 ELECTRICAL INSTRUMENTATION

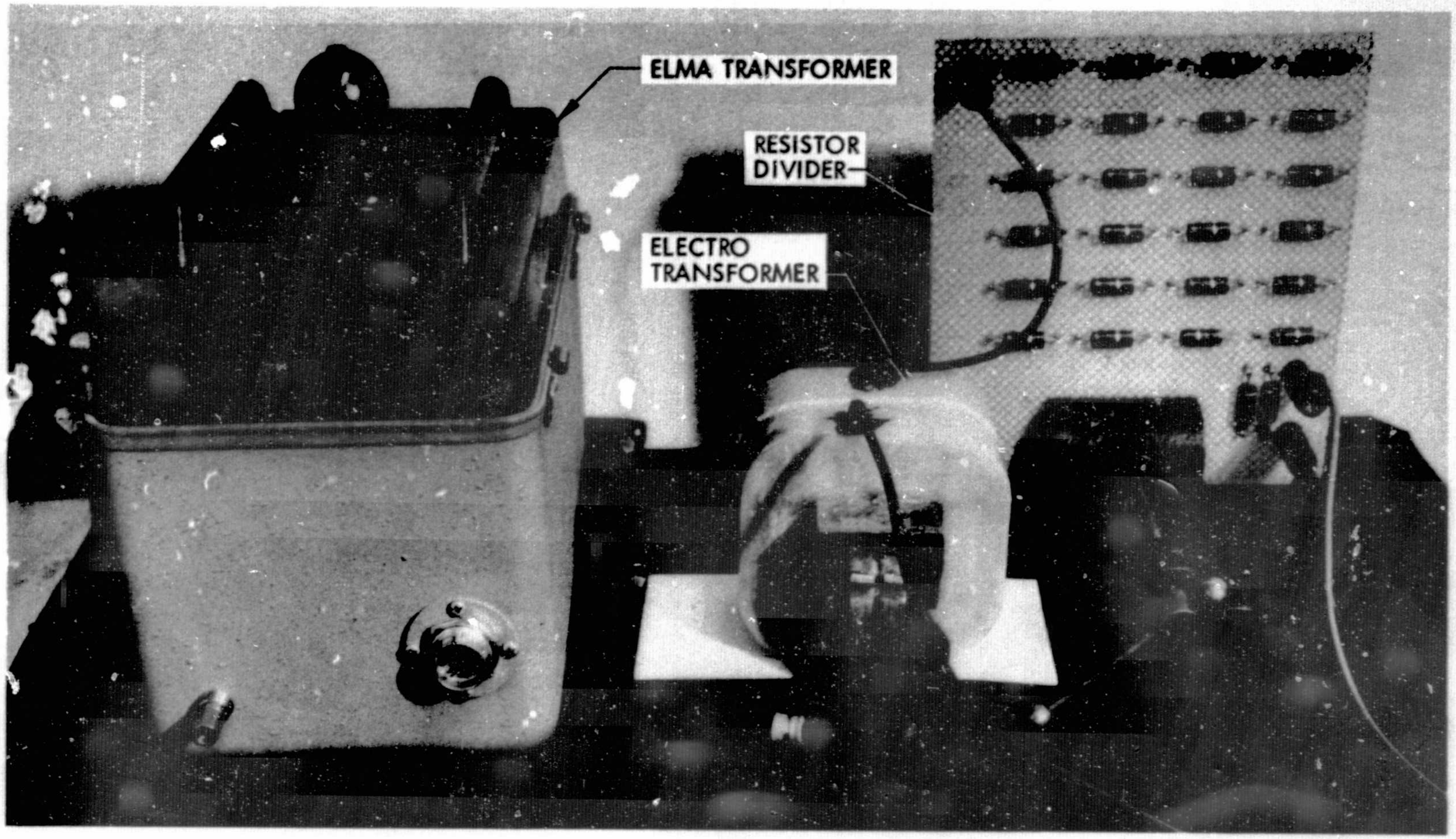


Figure 5 The High Voltage Transformers and the Resistor Voltage Divider

A Tektronix model 422 oscilloscope was connected across the voltage sensing resistor and current sensing resistor to show the voltage and current waveforms in their phase relationship. Careful reading of the oscilloscope face permitted a reasonably close measure of phase shift.

Finally, high voltage insulated wire was used for the line from the transformer to the resistor chain and the internal electrode of the reactor vessel, while the low end of the high voltage winding was attached to earth ground and the external electrode of the reactor. The whole high voltage apparatus was set on a thick rubber mat. In lab work, a protective screen cage is advisable, and in any case extreme care must be used to avoid personnel contact with the high voltage.

The entire experimental setup is shown in Figure 6. This is a convenient apparatus and permits rapid and reliable acquisition of data and easy control and recording of the experimental conditions.

The Beckman model 777 Oxygen Analyzer was used rather than a gas chromatograph because it permitted rapid, continuous and "real-time" monitoring of the gas composition coming from the reactor vessel. It was this versatility that permitted collecting the large and diversified body of data utilized in this report. The Beckman analyzer was calibrated periodically to atmospheric oxygen, assumed to be an ideal 21%. The sensing head was refurbished once before and once during the experiment. It was assumed that a given percentage of O_2 in the processed gas represented twice that value of CO_2 reduced, based on the reaction formula:

$$2\ CO_2 \longrightarrow O_2 + 2\ CO$$

Because of the minute leaks in the system there was always some atmospheric O_2 in the base reading, and this was always subtracted from the experimental reading, prior to calculating the CO_2 reduced.

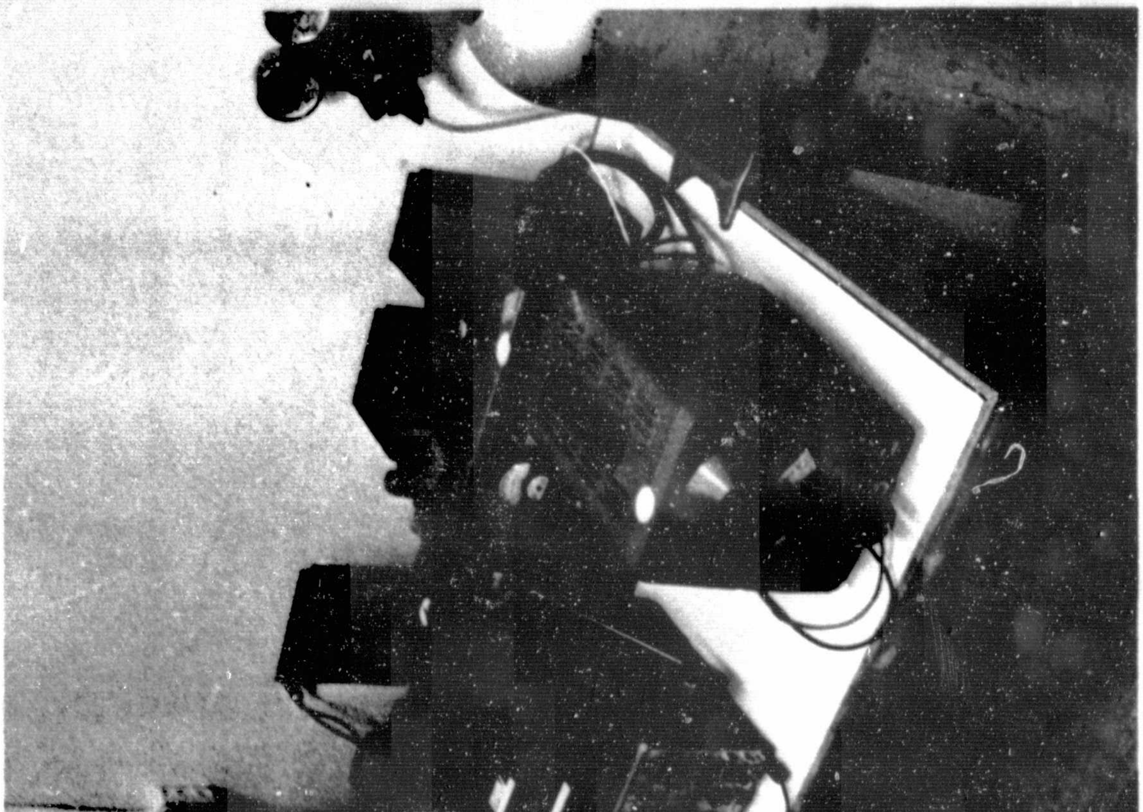


Figure 6 The Complete Experimental Setup

Allowing for accumulated errors in the various instruments it is considered that all data points are reliable to within ±10% of the final "CO_2 reduced" value.

The Experiment; Procedures and Data

Effects of Voltage and Frequency:

The experiment was conceived of as deriving from those described by the Russian researchers Andreev, Semiokhin, Panchenkov, and Baraev[1][2]. It was expected that by getting accurate data on the CO_2 reduction phenomenon under various carefully controlled conditions, the physics of the process could be correctly understood and as a result the efficiency of the process improved to a point encouraging practical application. Experimental variables were seen as follows:

- Frequency
- Voltage
- Flow rate
- Pressure
- Temperature

After the apparatus had been assembled and debugged by a series of trial runs, it was decided to run a formal test in which voltage and frequency were the variables, while flow rate, pressure and temperature were held constant.

The results of the first run are contained in table 1. This table shows the values established for the five variable parameters as well as the percentage of CO_2 reduced to O_2 and CO. The final column shows calculated electrical capacitance of the reactor vessel, since this turned out to be a variable dynamic characteristic and was of considerable interest as the test series proceeded.

Using the information in this table several graphs were developed. Graph 1 shows the relationship between voltage, frequency and current in the reactor vessel. The information shown by the dotted portion of the lines was calculated by assuming an ideal reactor capacitance of 105 pF, which is the case at potentials below about 3200 AC volts, before the gas inside the reactor has begun conducting and increased the apparent electrical capacitance of the vessel. This graph is of interest mainly in that it demonstrates the dynamic electrical impedance of the reactor vessel.

Graph 2 was constructed so that data could be interpolated for the making of graph 3, however this graph shows clearly that no CO_2 reduction takes place until apotential of about 3200 AC volts has been reached, at which voltage the gas starts to ionize.

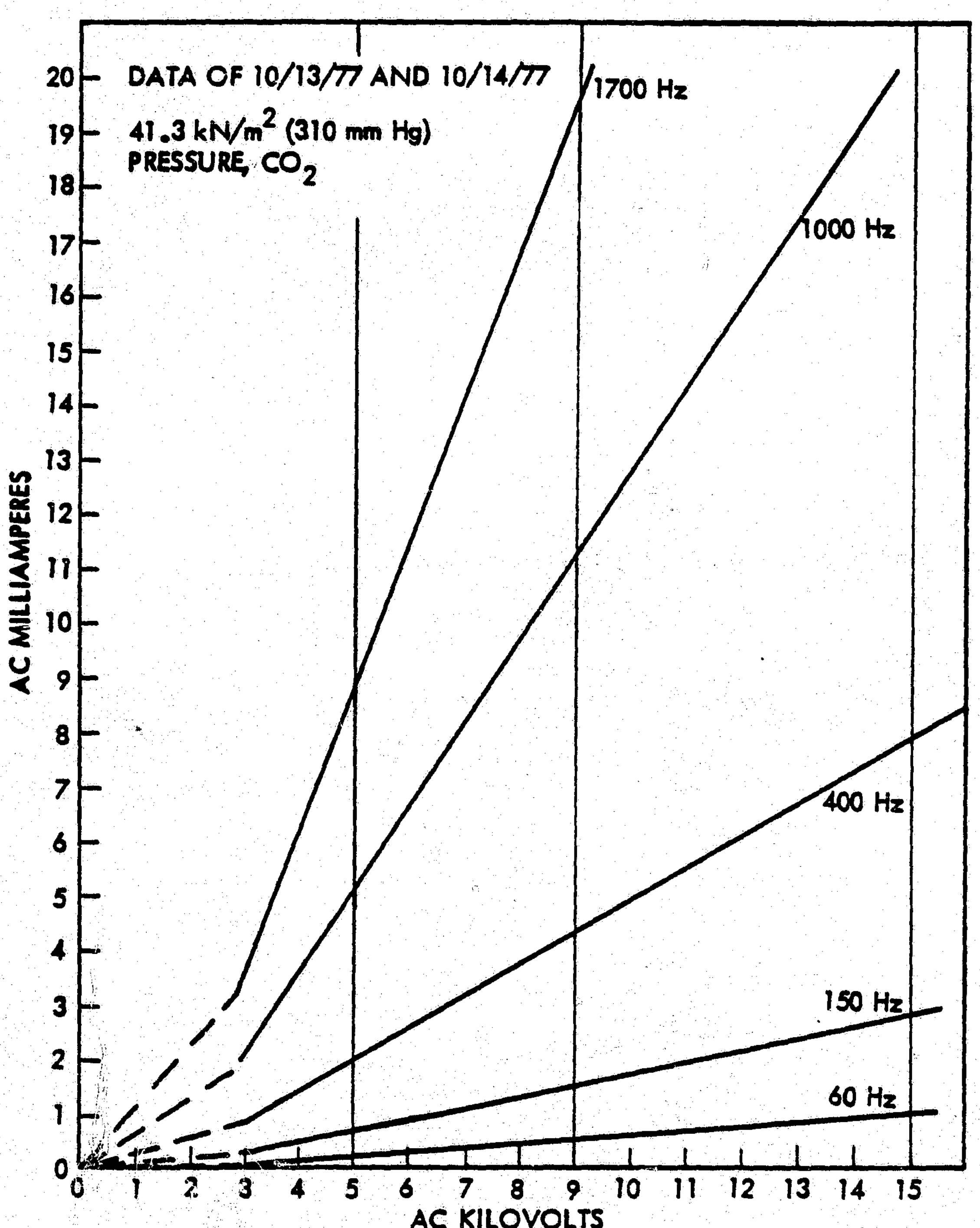
Graph 3 shows the relationship of CO_2 reduction to voltage and frequency. The CO_2 levels were derived by interpolation from graph 2. Graph 3 clearly shows that increasing voltage or frequency, or both, increases the amount of CO_2 reduced. At this point in the experiment it seemed that it would be necessary to generate and use much higher frequencies to produce dramatic results, since the voltages in use were already about as high as practical. It will be noted that doubling voltage or doubling frequency tends to double CO_2 reduction, however there is a diminishing effect due to back reaction as voltage or frequency is increased, and inspection of the data suggest that a conversion rate of 30% may represent the limit of this process.

Data from 10/13/77 and 10/14/77

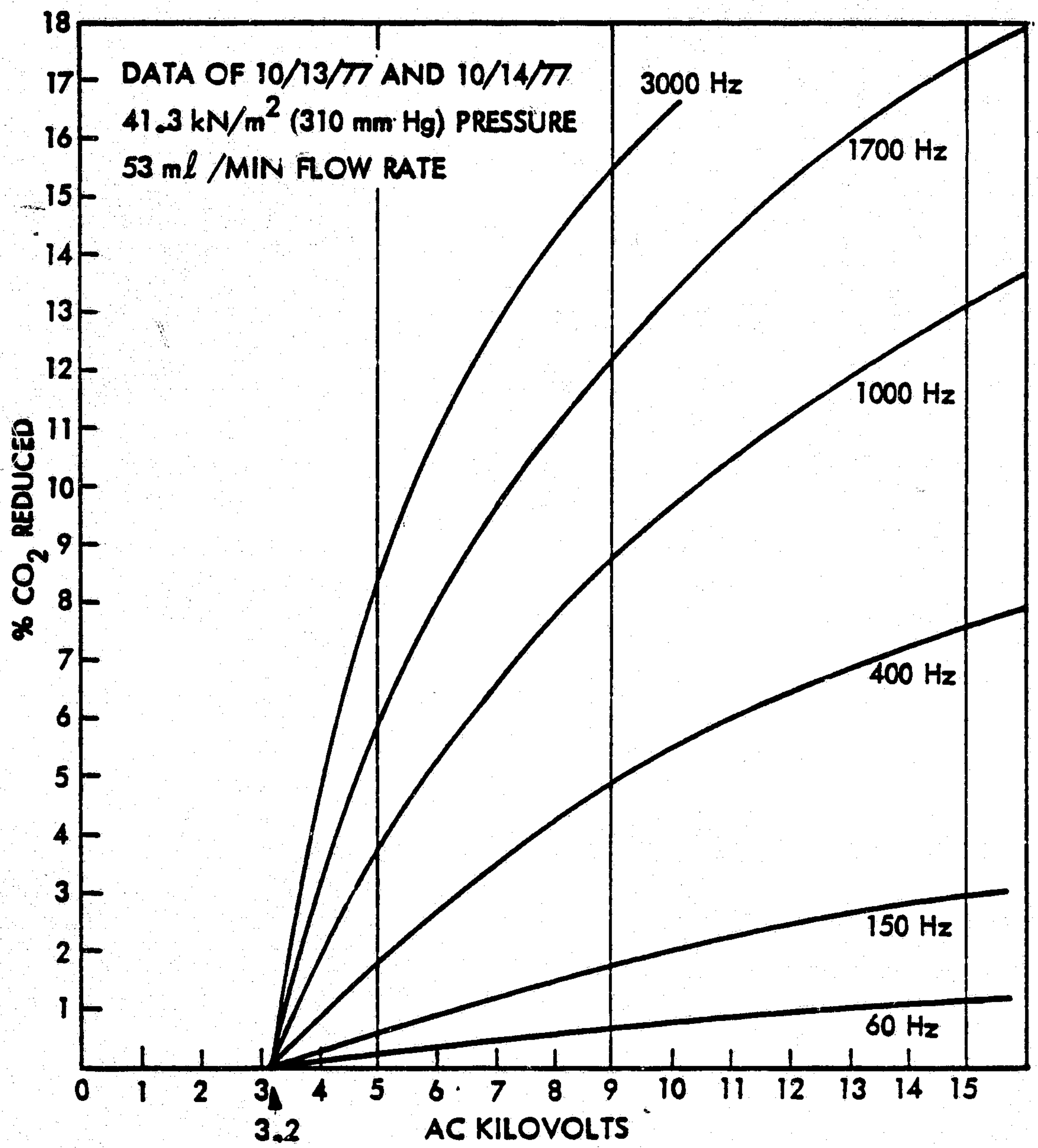
Frequency Hz	AC Kilo-volts	AC Milli-amperes	Pressure kN/cm^2	Pressure mmHg	Flow Rate, ml/minute	% CO_2 Reduced	Calculated Reactor Capacitance, Picofarads
60	5.0	0.23	41.3	310	53	0.18	122
60	9.0	0.56				0.60	165
60	15.0	1.05				1.24	186
150	5.0	0.7				0.60	149
150	9.0	1.55				1.65	183
150	15.0	2.75				2.96	195
400	5.0	2.1				1.76	167
400	9.0	4.5				4.24	199
400	15.0	7.75				6.88	206
1000	5.0	5.1				3.80	162
1000	8.0	9.65				7.72	192
1000	9.0	11.7				9.00	207
1000	14.0	18.5				12.56	210
1700	5.0	8.9				6.04	167
1700	9.0	19.5				12.40	203
1700	15.0	33.5				17.36	209
3000	5.5	17.7	41.3	310	53	10.00	171

Table 1 Relation of Frequency and Voltage to CO_2 Reduction

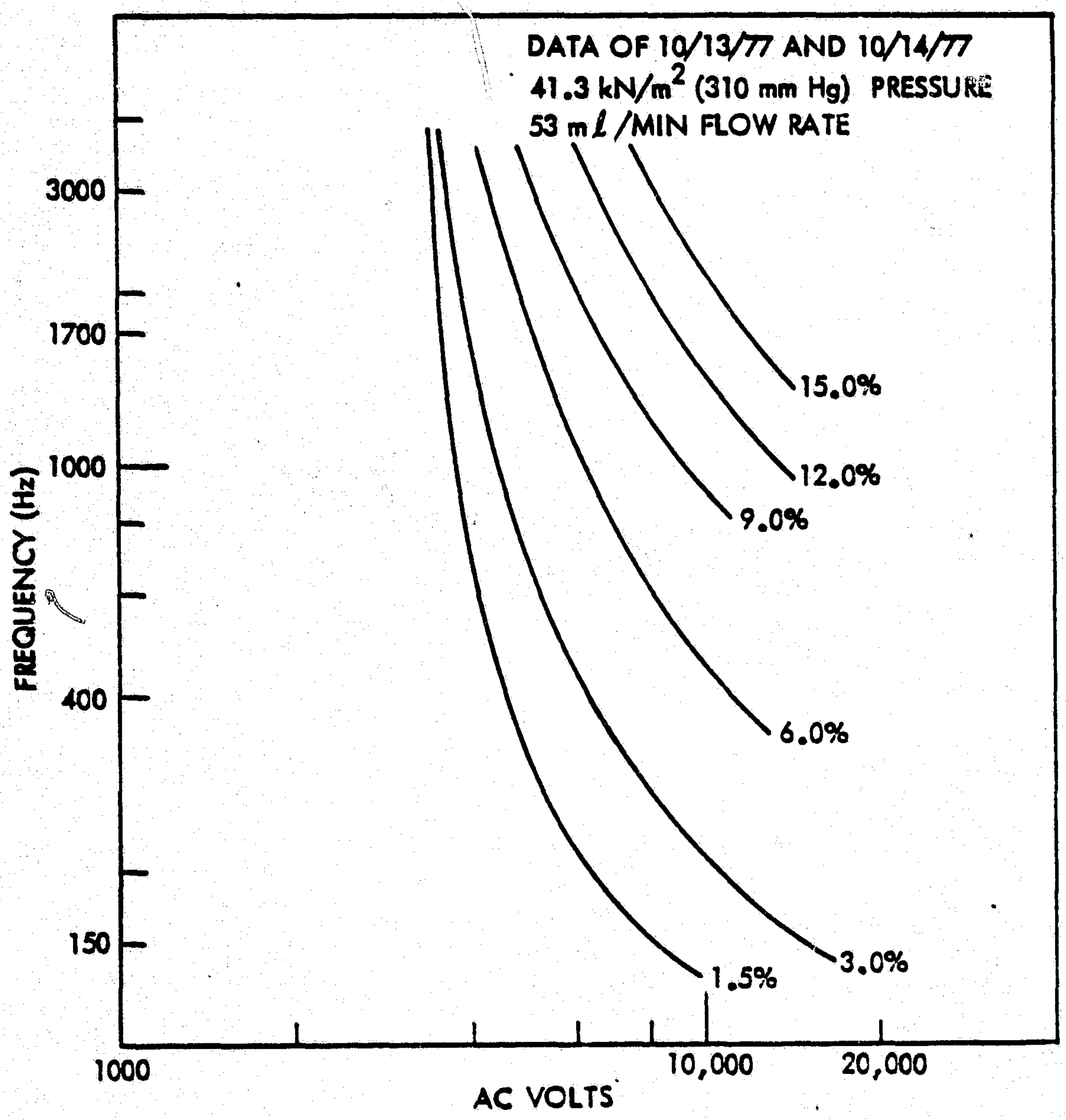
GRAPH 1 REACTOR CURRENT, VOLTAGE AND FREQUENCY



GRAPH 2 CO_2 REDUCTION RELATED TO VOLTAGE AND FREQUENCY



GRAPH 3 CO_2 REDUCTION; EFFECT OF VOLTAGE VS FREQUENCY



Optimum Pressure:

A test run was next performed for the purpose of determining the pressure at which the CO_2 reduction process proceeded most efficiently. The Russian literature indicated 40.0 kN/m^2 (300 mmHg) as the most efficient point, so it decided to use that pressure as one point, with pressure higher and lower as the other data points. Table 2 complies the results of this test. An attempt has been made to adjust the values for CO_2 reduction based on the slightly different flow rates at each pressure level, and these adjusted values were used to make up graph 4. This graph suggests that 46.7 to 53.3 kN/m^2 (350 to 400 mmHg) may be the most efficient operating pressure. Only two frequencies and two voltages are used, as it appeared from the first data run that this would produce high CO_2 reduction rates and more reliable data.

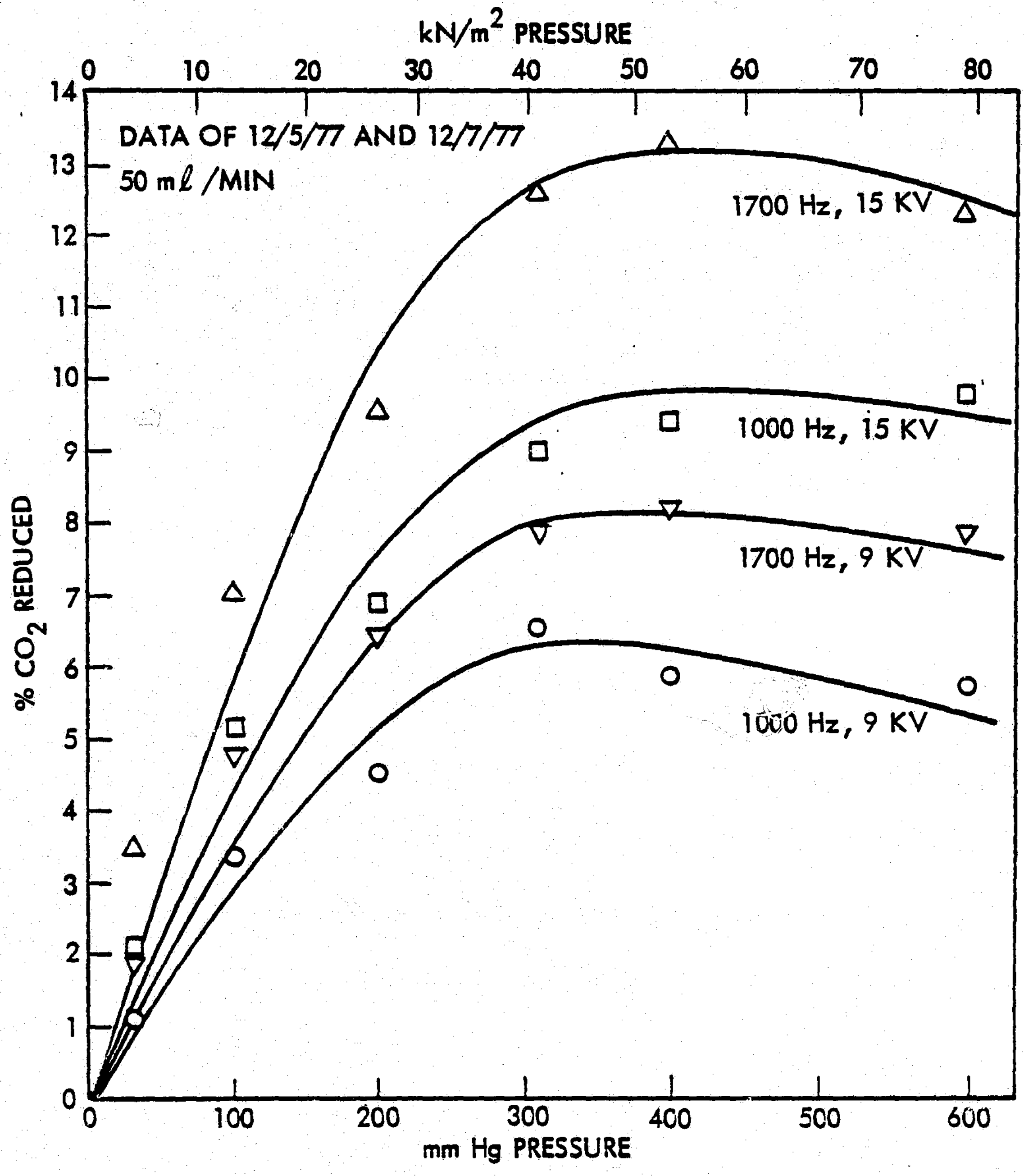
Somewhat further along in the experiment this test was repeated, this time with an effort to achieve more accurate results. Table 3 contains these data, and graph 5 shows the curve. Graph 5 supports a conclusion that 40.0 to 53.3 kN/m^2 (300 to 400 mmHg) is the most efficient pressure region.

Data from 12/5/77 and 12/7/77

Frequency Hz	AC Kilovolts	AC Milliamperes	Pressure kN/m^2	Pressure mmHg	Flow Rate ml/min	% CO_2 Reduced	Calculated Reactor Capacitance Picofarads	% CO_2 Normalized for 50ml/min
1000	9.0	13.2	4.0	30	46	1.18	233	1.09
1000	15.0	21.0	4.0	30	42	2.50	223	2.10
1700	9.0	21.5	4.0	30	46	2.06	224	1.90
1700	15.0	34.5	4.0	30	42	4.11	215	3.45
1000	9.0	13.0	13.3	100	56	3.00	230	3.36
1000	15.0	21.0	13.3	100	57	4.50	223	5.13
1700	9.0	21.5	13.3	100	56	4.26	224	4.77
1700	15.0	34.5	13.3	100	57	6.12	215	6.98
1000	9.0	12.5	26.7	200	56	4.04	221	4.52
1000	15.0	21.0	26.7	200	56	6.16	223	6.90
1700	9.0	20.5	26.7	200	56	5.76	213	6.45
1700	15.0	34.0	26.7	200	56	8.54	212	9.56
1000	9.0	12.5	41.3	310	52	6.22	221	6.47
1000	15.0	21.0	41.3	310	52	8.64	223	8.99
1700	9.0	20.0	41.3	310	52	7.58	208	7.88
1700	15.0	34.0	41.3	310	52	12.12	212	12.60
1000	9.0	12.0	53.3	400	59	4.96	212	5.85
1000	15.00	20.5	53.3	400	59	7.96	218	9.39
1700	9.0	19.0	53.3	400	59	6.94	198	8.19
1700	15.0	33.5	53.3	400	59	11.20	209	13.22
1000	9.0	10.5	80.0	600	50	5.76	186	5.76
1000	15.0	19.5	80.0	600	50	9.80	207	9.80
1700	9.0	17.0	80.0	600	50	7.82	177	7.82
1700	15.0	31.5	80.0	600	50	12.14	197	12.14

Table 2 CO_2 Reduction vs Pressure

GRAPH 4 CO_2 REDUCTION VS PRESSURE

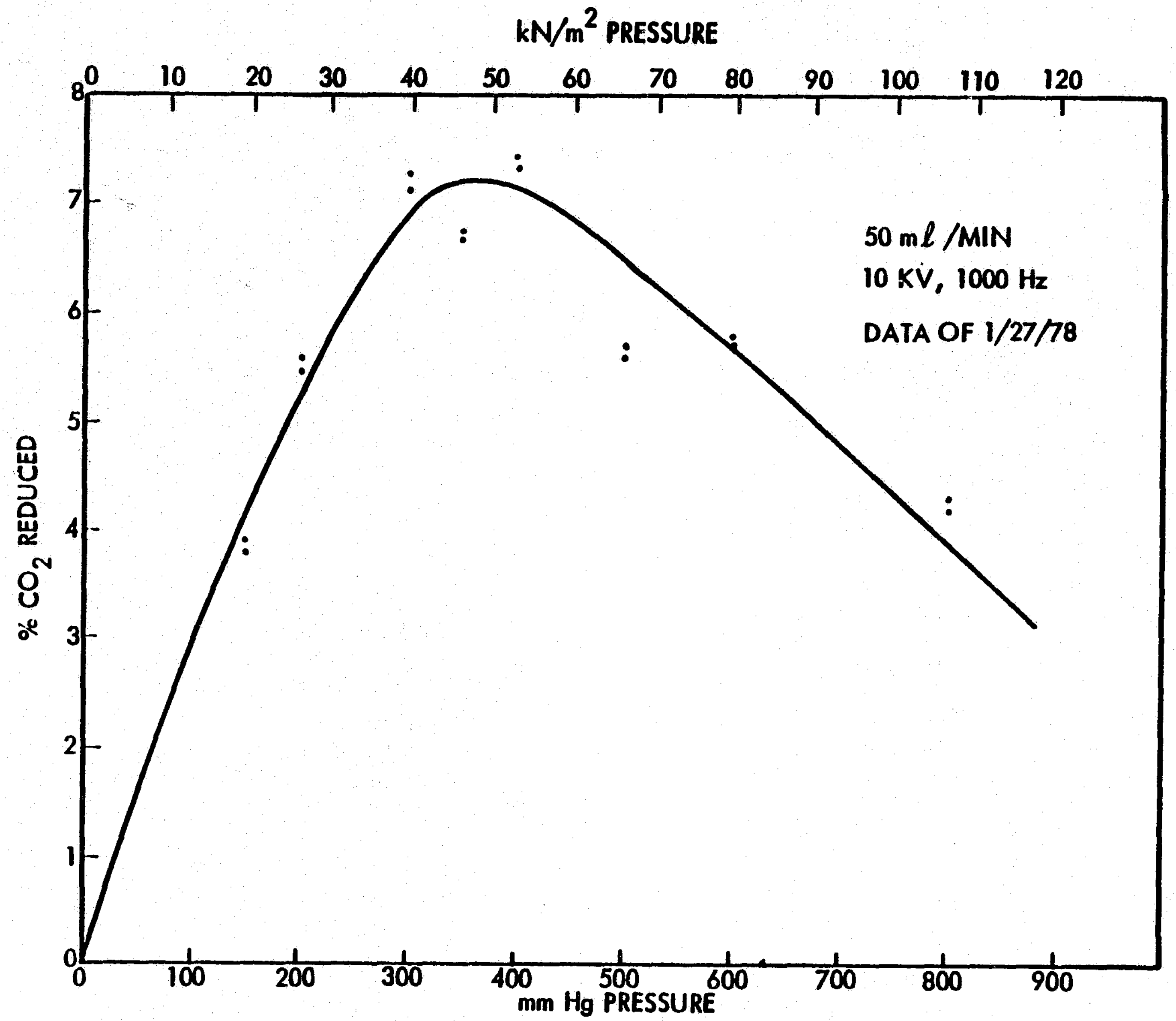


Data from 1/27/78

Frequency Hz	AC Kilovolts	Pressure kN/m^2	Pressure mmHg	Flow Rate m l/min	% CO_2 Reduced
1000	10.0	20.0	150	50	3.86
		20.0	150		3.86
		26.7	200		5.56
		26.7	200		5.46
		40.0	300		7.12
		40.0	300		7.26
		46.7	350		6.72
		46.7	350		6.72
		53.3	400		7.34
		53.3	400		7.30
		66.7	500		5.56
		66.7	500		5.60
		80.0	600		5.72
		80.0	600		5.70
		106.7	800		4.22
1000	10.0	106.7	800	50	4.30

Table 3 Test for Optimum Pressure

GRAPH 5 TEST FOR OPTIMUM PRESSURE



Effect of Flow Rate:

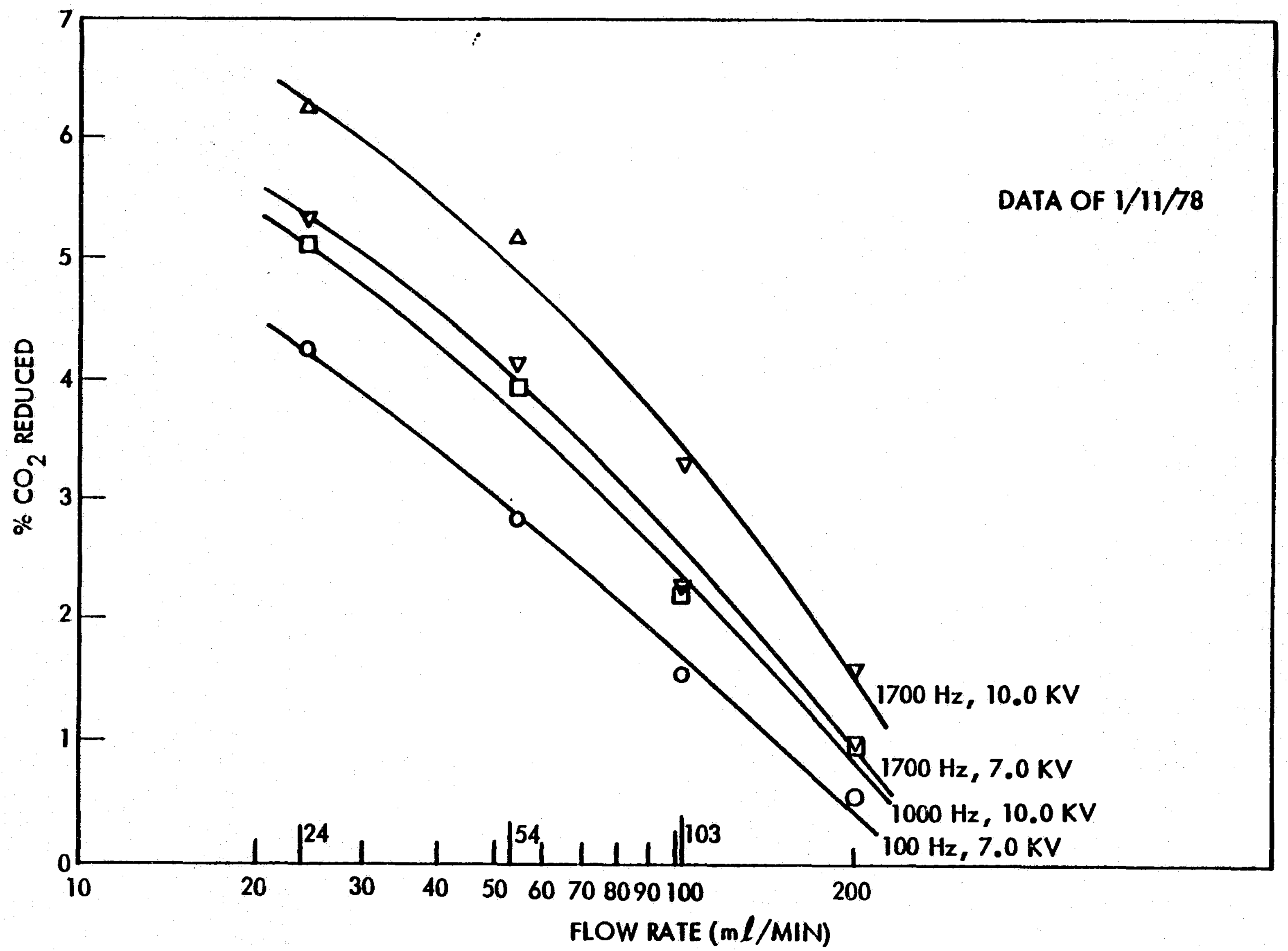
The next run was devised to test the effect of flow rate on CO_2 reduction. Tables 4A, 4B and 4C compile the data from this test. The pressures of 26.7, 40.0 and 53.3 kN/m^2 (200, 300 and 400 mmHg) were chosen since they bracketed the optimum pressure range determined by the previous test. Graphs 6A, 6B and 6C present these data visually. As was anticipated, the rate of CO_2 reduction is directly related to flow rate; cutting the flow rate in half tends to double the CO_2 reduction and vice versa. Again, there is a limit to this process, due to back reaction, possibly reaching a limit in the vicinity of 30% CO_2 reduced.

Data from 1/11/78

Frequency Hz	AC Kilovolts	AC Milliamperes	Pressure kN/m^2	Pressure mmHg	Flow Rate ml/min	% CO_2 Reduced	Calculated Reactor Capacitance Picofarads
1000	7.0	9.0	26.7	200	24	4.24	205
1000	10.0	13.5			24	5.12	215
1700	7.0	15.0			24	5.34	201
1700	10.0	23.0			24	6.28	215
1000	7.0	9.0			54	2.84	205
1000	10.0	13.5			54	3.92	215
1700	7.0	15.5			54	4.12	207
1700	10.0	23.0			54	5.18	215
1000	7.0	9.0			103	1.56	205
1000	10.0	14.5			103	2.20	231
1700	7.0	15.0			103	2.28	201
1700	10.0	23.0			103	3.30	215
1000	7.0	9.0			200	0.58	205
1000	10.0	13.5			200	0.96	215
1700	7.0	14.5			200	0.98	194
1700	10.0	22.5	26.7	200	200	1.60	211

Table 4A Effect of Flow Rate on CO_2 Reduction; 26.7 kN/m^2 (200 mmHg) Pressure

GRAPH 6A FLOW RATE VS CO_2 REDUCTION AT 26.7 kN/m^2 (200 mmHg) PRESSURE

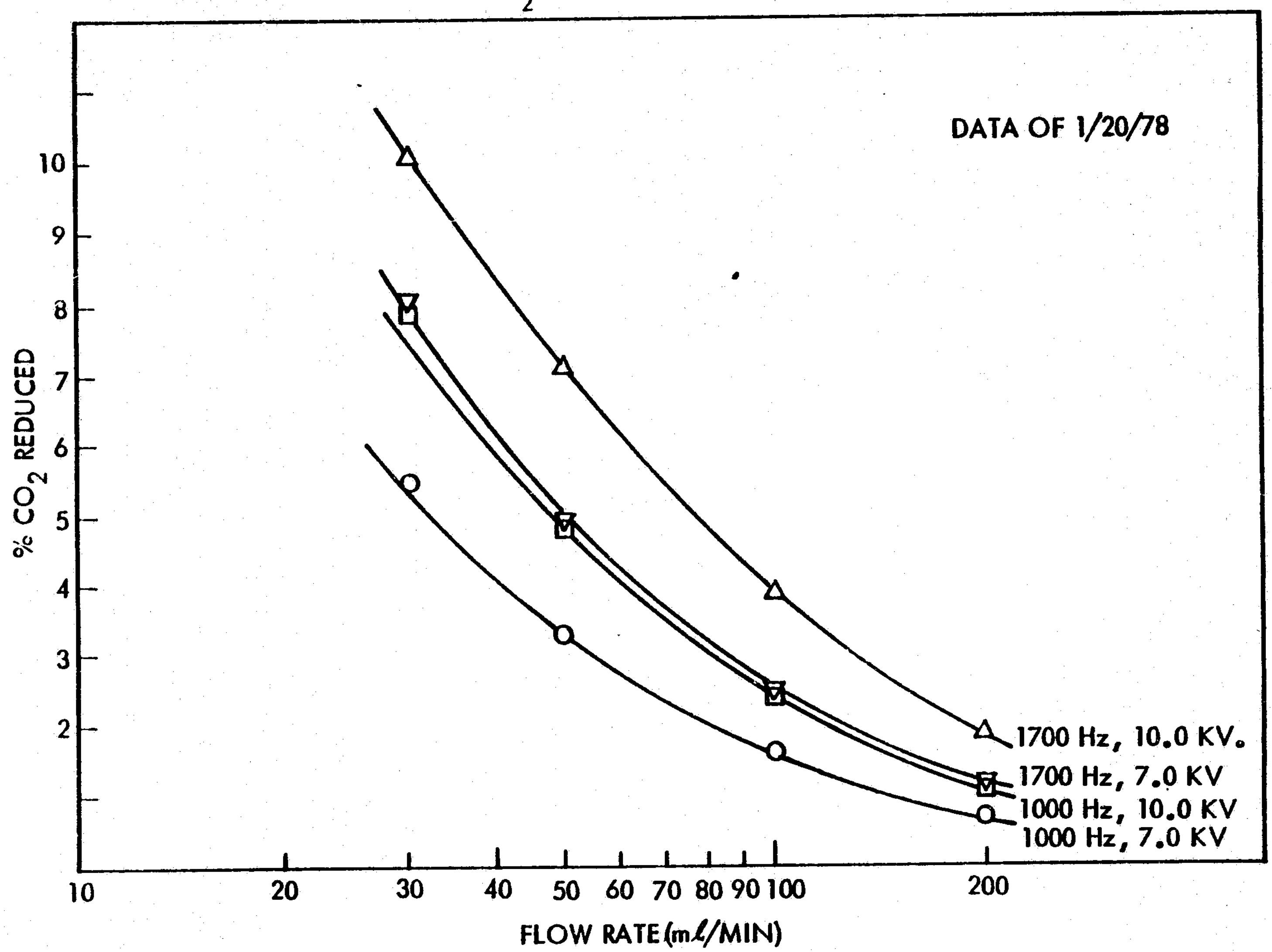


Data from 1/20/78

Frequency Hz	AC Kilovolts	AC Milli-amperes	Pressure kN/m^2	Pressure mmHg	Flow Rate ml/min	% CO_2 Reduced	Calculated Reactor Capacitance Picofarads
1000	7.0	8.5	40.0	300	30	5.46	193
1000	10.0	13.5			30	7.84	215
1700	7.0	15.0			30	8.02	201
1700	10.0	23.0			30	10.08	215
1000	7.0	8.5			50	3.26	193
1000	10.0	13.5			50	4.80	215
1700	7.0	15.0			50	4.96	201
1700	10.0	23.5			50	7.12	220
1000	7.0	8.3			100	1.60	193
1000	10.0	13.5			100	2.38	215
1700	7.0	14.5			100	2.46	194
1700	10.0	23.5			100	3.86	220
1000	7.0	8.5			201	0.72	193
1000	10.0	13.5			201	1.04	215
1700	7.0	14.5			201	1.06	194
1700	10.0	23.0	40.0	300	201	1.94	215

Table 4B. Effect of Flow Rate on CO_2 Reduction; 40.0 kN/m^2 (300 mmHg) Pressure

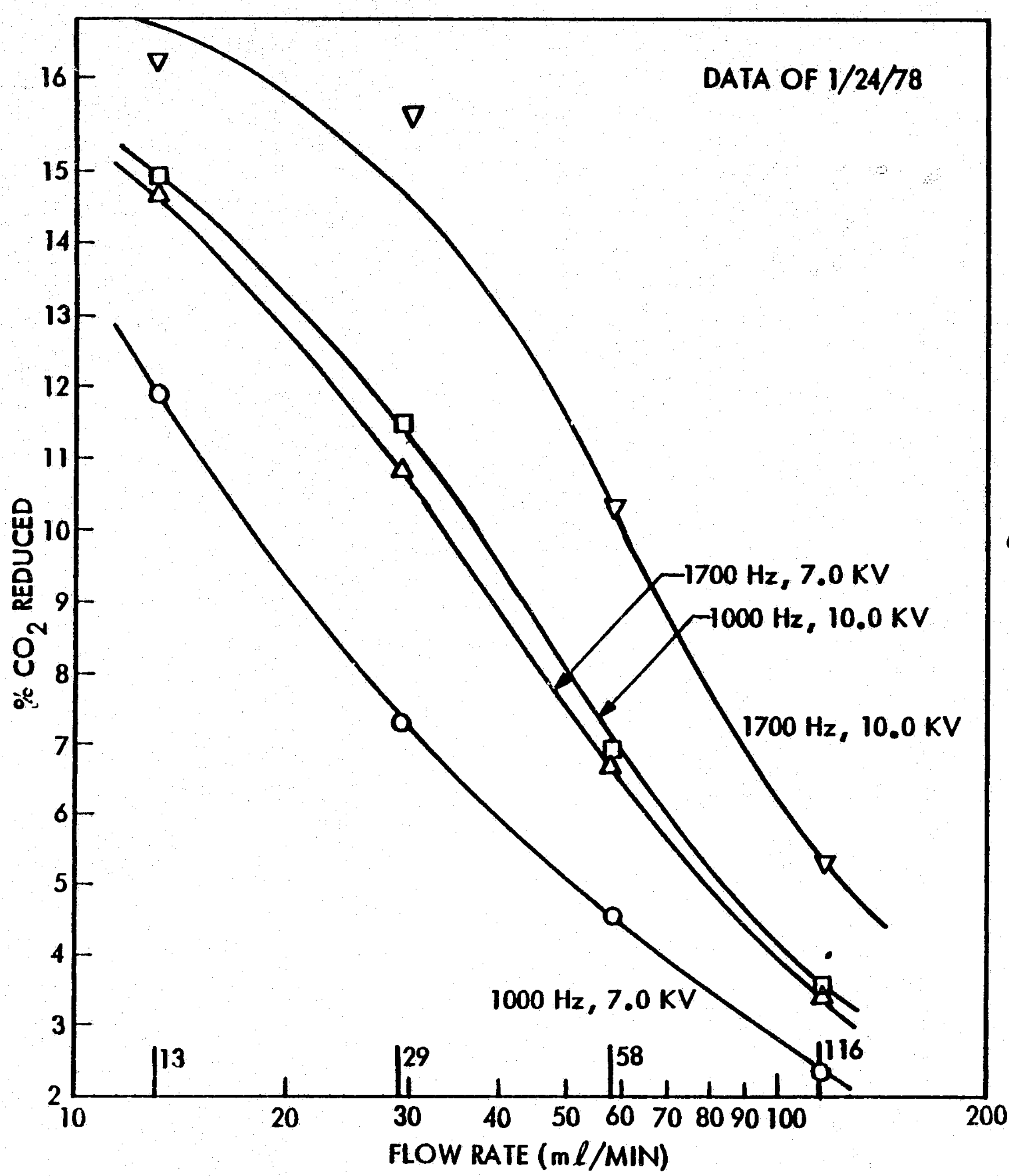
GRAPH 6B FLOW RATE VS CO_2 REDUCTION AT 40.0 kN/m^2 (300 mmHg) PRESSURE



Data from 1/24/78

Frequency Hz	AC Kilovolts	AC Milliamperes	Pressure kN/m^2	Pressure mmHg	Flow Rate ml/min	% CO_2 Reduced	Calculated Reactor Capacitance Picofarads
1000	7.0	8.0	53.3	400	13	11.86	182
1000	10.0	12.5	↑	↑	13	14.96	199
1700	7.0	13.0			13	14.64	174
1700	10.0	21.0			13	16.56	197
1000	7.0	7.5			29	7.30	171
1000	10.0	12.5			29	11.50	199
1700	7.0	13.0			29	10.82	174
1700	10.0	21.5			29	15.74	201
1000	7.0	8.0			58	4.60	182
1000	10.0	13.0			58	6.94	207
1700	7.0	13.5			58	6.74	181
1700	10.0	21.5			58	10.16	201
1000	7.0	8.0			116	2.38	182
1000	10.0	13.0			116	3.60	207
1700	7.0	13.5	↓	↓	116	3.46	181
1700	10.0	21.5	53.3	400	116	5.36	201

Table 4C. Effect of Flow Rate on CO_2 Reduction; 53.3 kN/m^2 (400 mmHg) Pressure.



GRAPH 6C FLOW RATE VS CO_2 REDUCTION AT 53.3 kN/m^2 (460 mmHg) PRESSURE

Effect of Current:

The preceeding tests succeeded in describing the effects of voltage, frequency, pressure and flow rate on the CO_2 reduction, as originally intended, however experience with the data suggested that a test should be run in which current through the reactor was the prime variable. Three frequencies were chosen, and the currents were carefully set to the values shown in the table by adjusting the voltages applied to the reactor. Table 5 shows the data which is plotted on graph 7. The surprising feature of this graph is the way in which the three frequency curves are displaced from one another along the current axis. These displacements result because at higher frequencies the reactor vessel presents a lower electrical impedance to the applied current. If the current displacements are subtracted, the data points will tend to fall along a single line.

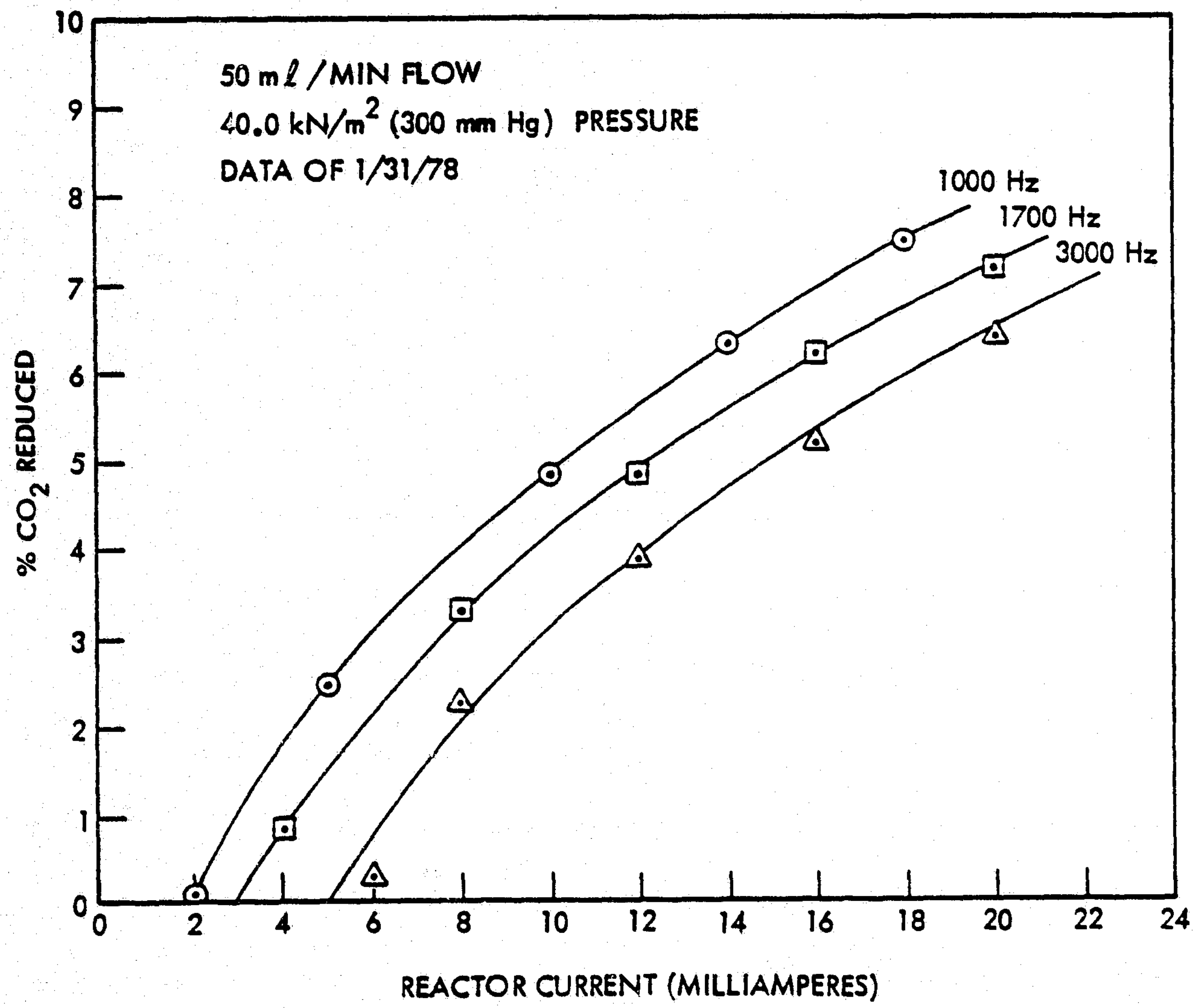
This was an important test in the series because it brought out the fact that current is the mediator of the CO_2 reduction process, and that frequency and voltage are of interest only in that they determine the current "through" the reactor vessel in conformance with the electrical formulas applying to capacitive reactance.

Data from 1/31/78

Frequency Hz	AC Kilovolts	AC Milliamperes	Pressure kN/m^2	Pressure mmHg	Flow Rate ml/min	% CO_2 Reduced	Calculated Reactor Capacitance Picofarads
1000	1.65	1.0	40.0	300	50	0.00	96
	2.6	2.0				0.14	122
	4.7	5.0				2.42	169
	7.8	10.0				4.84	204
	10.5	14.0				6.30	212
1000	13.2	18.0	40.0	300	50	7.44	217
1700	1.0	1.0	40.0	300	50	0.00	94
	1.9	2.0				0.00	99
	3.4	4.0				0.86	110
	4.8	8.0				3.30	156
	6.0	12.0				4.84	187
	7.4	16.0				6.20	202
1700	8.9	20.0	40.0	300	50	7.18	210
3000	1.0	2.0	40.0	300	50	0.00	106
	2.0	4.0				0.00	106
	3.0	6.0				0.28	106
	3.4	8.0				2.16	125
	4.2	12.0				3.88	152
	4.9	16.0				5.22	173
3000	5.6	20.0	40.0	300	50	6.40	189

Table 5. Effect of Current and Frequency on CO_2 Reduction

GRAPH 7 EFFECT OF CURRENT AND FREQUENCY ON CO_2 REDUCTION



Current vs. Pressure:

Once the role of current was recognized it was thought that a test should be run in which two values of current were held constant across a wide range of pressure. Table 6 contains the data from this test, and graph 8 presents it in visual form. These data again support the belief that 40.0 to 53.3 kN/m^2 (300 to 400 mmHg) is the optimum pressure for the process.

The two different slopes from 53.3 to 160.0 kN/m^2 (400 to 1200 mmHg) were a puzzle until it was realized that:

1. Ionization potential might vary with pressure.
2. Current through the reactor that could be accounted for by the minimum capacitance of 105 picofarads did not enter into the CO_2 reduction process.

In short, it was hypothesized that the reduction process was mediated solely by current in the plasma, and this value was equal to the total current minus the minimum capacitance current, thus:

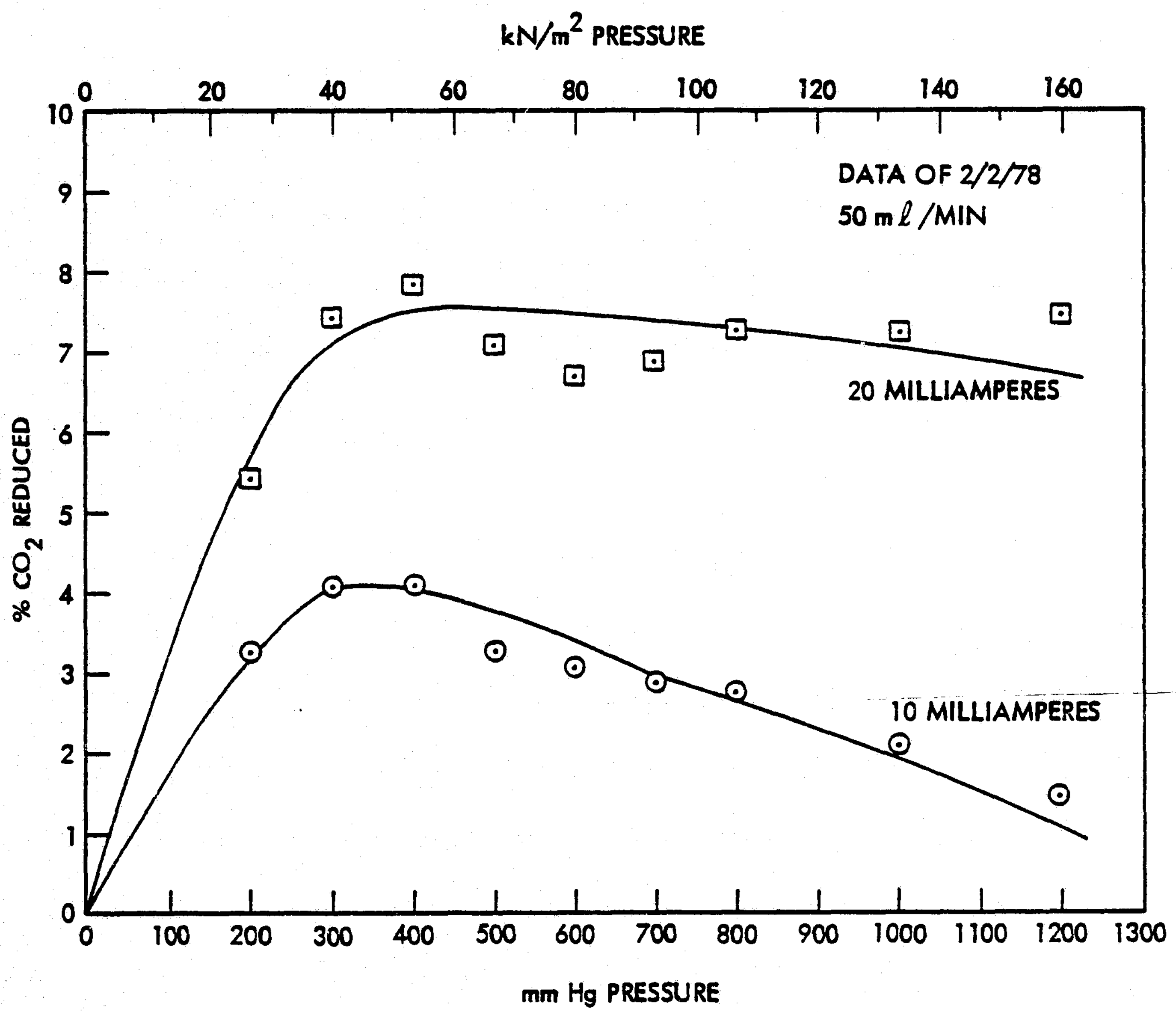
$$A_{plasma} = A_{total} - A_{minimum\ capacitance}$$

Data from 2/2/78

Frequency Hz	AC Kilovolts	AC Milliamperes	Pressure kN/m^2	Pressure mmHg	Flow Rate ml/min	% CO_2 Reduced	Calculated Reactor Capacitance Picofarads
1700	5.0	10.0	26.7	200	50	3.30	187
	8.8	20.0	26.7	200		5.42	213
	5.4	10.0	40.0	300		4.08	173
	9.1	20.0	40.0	300		7.44	206
	5.9	10.0	53.3	400		4.10	159
	9.4	20.0	53.3	400		7.82	199
	6.3	10.0	66.7	500		3.32	149
	9.9	20.0	66.7	500		7.14	189
	6.8	10.0	80.0	600		2.88	138
	10.2	20.0	80.0	600		6.74	184
	7.2	10.0	93.3	700		2.86	130
	10.7	20.0	93.3	700		6.92	175
	7.6	10.0	106.7	800		2.80	123
	11.2	20.0	106.7	800		7.30	167
	8.4	10.0	133.3	1000		2.10	111
	12.0	20.0	133.3	1000		7.24	156
	9.1	10.0	160.0	1200		1.46	103
1700	13.1	20.0	160.0	1200	50	7.50	143

Table 6. Pressure vs CO_2 Reduced; Constant Currents

GRAPH 8 PRESSURE VS CO_2 REDUCED, CURRENTS HELD CONSTANT



Effect of Plasma Current:

Having postulated that current in the plasma was the sole agent of the reduction process, tests were planned to check this supposition.

First a test was run to determine the effect of pressure on the ionization potential. Three frequencies were used, and pressures were varied over a wide range. The problem of determining the onset or extinction of ionization was solved by using a small AM radio to pick up the abrupt roar of broad band radio-frequency energy that was present whenever the plasma was present. The plasma onset voltage was slightly higher than the extinction voltage, and it was decided to record the extinction voltages since this seems to more correctly represent the situation, with all data points being collected in the ionized state. It will be noted in Table 7 that frequency had almost no effect, at least in the range studied. Graph 9 was prepared from the data. It is probable that this graph explains the peaking of the CO_2 reduction process at 40.0 to 53.3 kN/m^2 (300 to 400 mmHg) as an artifact, since for a constant voltage across the reactor vessel there would be first a rising rate of CO_2 reduction as the pressure was increased from the lowest value, then a peaking prior to the decline caused by the ever rising ionization potential as the pressure increase was continued.

A test was then run using one frequency only in which three reactor currents were spaced by 5 milliamperes and 10 milliamperes at a wide range of pressures. The minimum capacitance currents were computed as follows:

$$A_{min\ cap.} = 2\pi \cdot \text{Frequency} \cdot \text{Capacitance} \cdot \text{AC volts}$$

True plasma current was computed as follows:

$$A_{plasma} = A_{total} - A_{min\ cap}$$

These data are compiled in Table 8 while graph 10 was prepared to show the percent CO_2 reduced vs. the true plasma current. Notice that the curves for 26.7 and 33.3 kN/m^2 (200 and 250 mmHg) reflect the phenomenon seen in the earlier graphs where a reduced process efficiency exists below 40.0 kN/m^2 (300 mmHg).

Graph 10 seems to confirm that for a given flow rate, referred to 101.3 kN/m^2 (760 mmHg), current in the plasma is the sole mediator of CO_2 reduction. Voltage and frequency are involved only in that they establish the current. Pressure above 40.0 kN/m^2 (300 mmHg) seems to have no effect other than causing an increase in the ionization potential. Graph 10 also shows the very important fact that the CO_2 reduction process is most efficient at low current levels, a point that is developed further in the following pages.

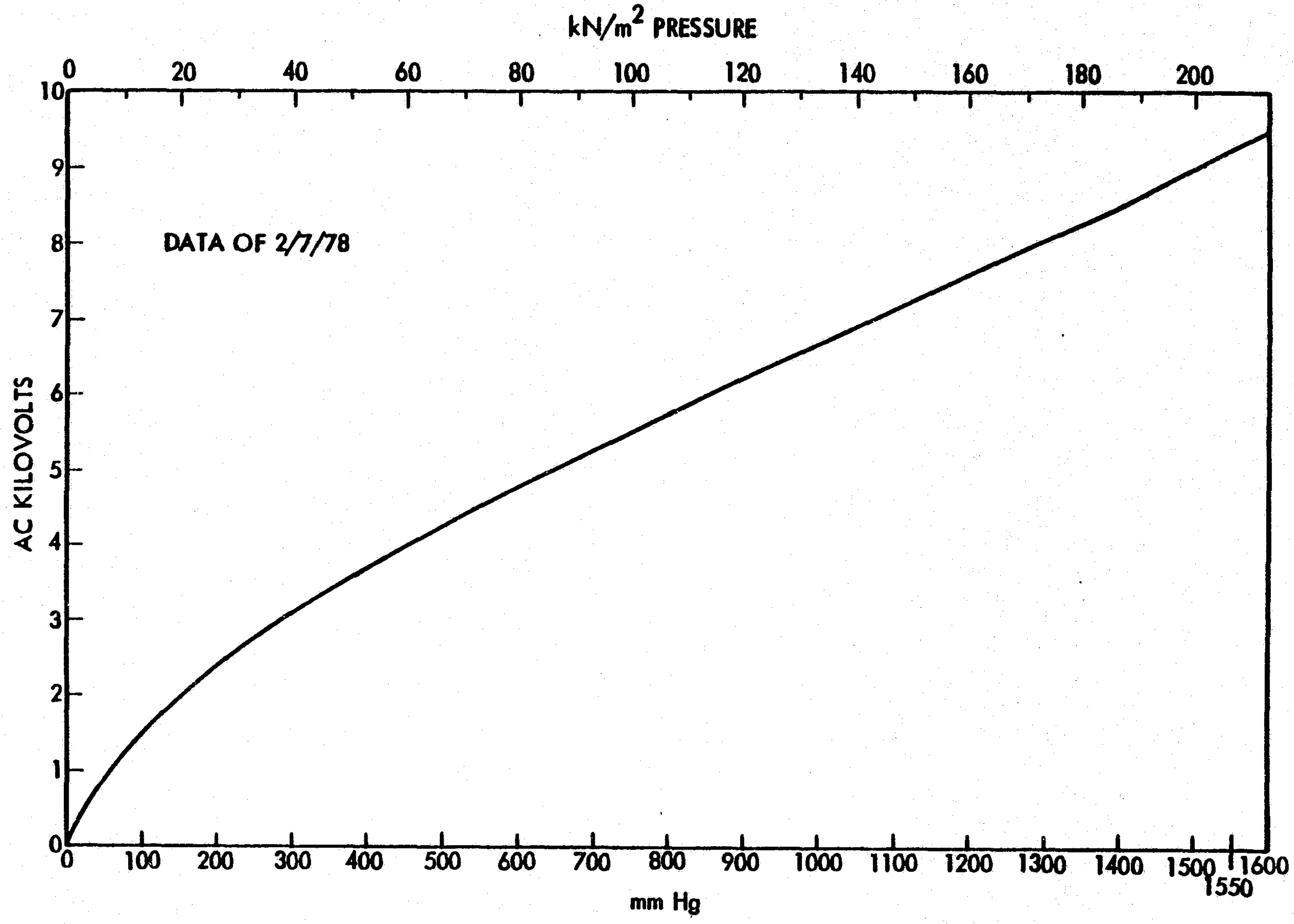
Data from 2/7/78

Table 7: Ionization Extinction Potentials for the Specific Reactor Vessel Used in the Experiment.

Pressure kN/m^2	Pressure mmHg	Extinction Potential, AC Kilovolts 1000 Hz	1900 Hz	3000 Hz
13.3	100	1.45	1.45	1.45
26.7	200	2.40	2.40	2.36
40.0	300	3.25	3.20	3.10
53.3	400	3.70	3.70	3.60
80.0	600	4.80	4.75	4.75
106.7	800	5.80	5.80	5.75
133.3	1000	6.70	6.75	6.70
160.0	1200	7.70	7.70	7.65
186.7	1400	8.60	8.50	8.50
206.6	1550	9.40	9.35	9.30

Voltages underlined were used to construct the graph.

GRAPH 9 MINIMUM AC VOLTAGE, REGARDLESS OF FREQUENCY, AT WHICH CO_2 IONIZATION IS MAINTAINED

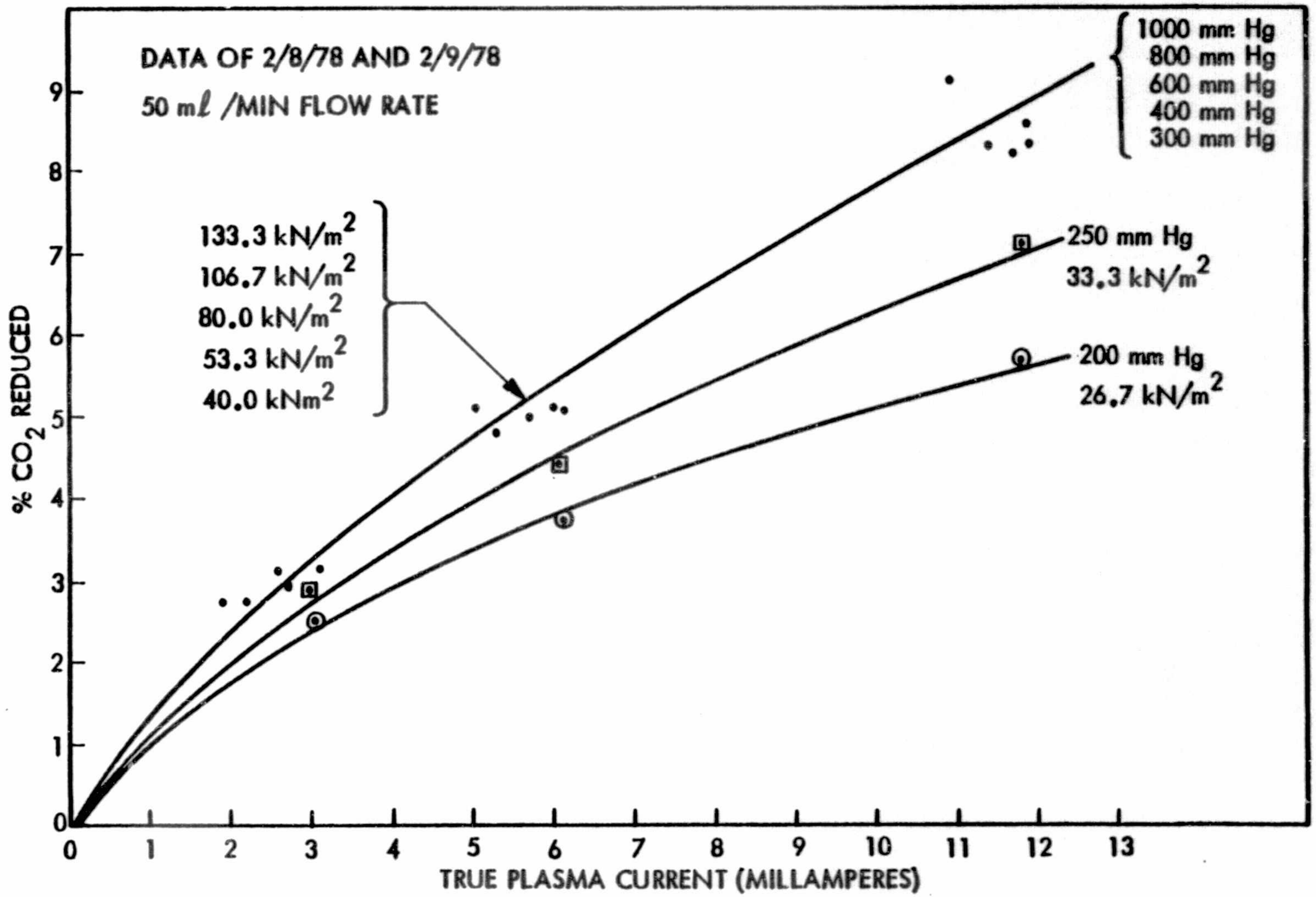


Data from 2/8/78 and 2/9/78

Frequency Hz	AC Killovolts	AC Milliamperes	Pressure kN/m^2	Pressure mmHg	Flow Rate ml/min	% CO_2 Reduced	Calculated Reactor Capacitance Picofarads	True Plasma Current, Milliamperes
1700	4.12	7.66	26.7	200	50	2.50	174	3.04
	4.83	12.66	26.7	200		3.76	203	6.12
	9.70	22.66	26.7	200		5.68	219	11.78
	4.54	8.03	33.3	250		2.86	166	2.94
	6.21	13.03	33.3	250		4.40	196	6.07
	10.0	23.03	33.3	250		7.14	216	11.81
	4.77	8.44	40.0	300		3.14	166	3.09
	6.50	13.44	40.0	300		5.12	194	6.15
	10.3	23.44	40.0	300		8.16	213	11.89
	5.80	8.98	53.3	400		3.08	145	2.48
	7.13	13.98	53.3	400		5.10	184	5.98
	10.8	23.98	53.3	400		8.34	208	11.87
	6.61	10.08	80.0	600		2.94	143	2.67
	8.36	15.08	80.0	600		4.84	169	5.70
	11.9	25.08	80.0	600		8.10	197	11.73
	7.95	11.14	106.7	800		2.76	131	2.22
	9.67	16.14	106.7	800		4.78	156	5.29
	13.15	26.14	106.7	800		8.34	186	11.39
	9.08	12.07	133.3	1000		2.76	124	1.89
	10.9	17.07	133.3	1000		5.14	147	4.85
1700	14.4	27.07	133.3	1000	50	8.90	176	10.92

Table 8. CO_2 Reduced Relative to Pressure and True Plasma Current

GRAPH 10 CO_2 REDUCTION VS PLASMA CURRENT



Effect of Temperature:

No thorough check of the influence of temperature was possible due to the limitation of time and equipment to do this, however a rough check was made by inserting a 12 inch length of 1/8" gas line leading into the reactor vessel, and removing the reactor vessel from the Freon bath. Checks were run on CO_2 reduction rates while heating the tube with a 2000 watt heat gun. There was no significant or repeatable difference in reduction rates with the heat gun applied to or removed from the tube, so it is cautiously concluded that the temperature of the gas, in the range tested, is not significantly able to influence the reaction.

Improving the Power Factor:

Examination of the tables of data will show that very high levels of AC voltage and AC current were measured at the reactor vessel, implying power levels up to 500 watts. It would be expected that the boiling of the Freon would be considerable, however a very slight boiling actually occurred at these high power levels, and no boiling or detectable heating at the lower powers used for most of the tests. This is explained by assuming that most of the power was actually "wattless power" caused by the electrical reactance of the reactor vessel which in fact is an electrical capacitor. The reactor vessel, filled with CO_2 gas is a dynamic device, with the capacitance varying between 105 picofarads and 246 picofarads depending on the degree of ionization of the CO_2 gas.

During the experiment the frequency of 1700 Hertz was used most since it seemed to be the one that "worked best", and permitted the highest voltages to be obtained from the Electro transformer. The suspicion arose that the inductance of the transformer was of such a value that combined with the capacitance of the reactor vessel, a resonance was occurring at about 1700 Hz. This would be a very desirable situation, since it would permit making a resonant system in which the AC current lost "through" the capacitance of the reactor vessel could be stored in the inductance of the transformer, reducing the power input to the resonant circuit to just that needed to replace losses due to pure resistance.

The transformer was removed from the test setup and it's primary winding shorted, to simulate a low impedance driving source, then the inductance of the secondary was measured on the ESI 250 DE impedance bridge giving a value of 34.3 Henrys. The DC resistance was almost exactly 10,000 ohms. The reactor vessel had been checked on the same ESI instrument earlier, with a value of 105 picofarads filled with dry air, and 246 picofarads filled with salt water to simulate a completely shorted gap. Using these values, the following resonant frequency limits would be expected:

34.3H, 105 pF	=	2652 Hz
34.3H, 246 pF	=	1733 Hz

In practice the transformer winding would be expected to have some capacitance which would add to the capacitance of the reactor vessel, thus somewhat lowering the frequency derived above.

It appeared that the transformer/reactor vessel circuit was in fact resonant, and had been used at a resonant frequency of 1700 Hz all along.

In order to investigate this, the Electro transformer was reinstalled in the test setup, and an AC voltmeter and an AC ammeter were connected to the transformer primary circuit. The inputs of a dual trace oscilloscope were connected to the high voltage divider circuit and to the reactor current sensing resistor so that the voltage/current phase relationship of the power applied to the reactor vessel could be observed. The locations of these instruments are shown in Figure 4.

A preliminary run showed that resonance did occur in close conformance to the above prediction. At low reactor voltages, resonance occurred at around 2390 Hz, while at a high reactor voltages, resonance occurred at about 1670 Hz. Resonance was determined by noting the dip in current in the transformer primary circuit, coupled with a slight rise in primary circuit voltage.

A careful series of measurements was then made, and the results compiled in Table 9. The current lead over voltage in the secondary, or reactor, circuit was measured as closely as possible by observing the two waveforms on the face of the oscilloscope. The table shows a number of very interesting things, as follows:

1. Resonant frequency decreases as the voltage across the reactor increases and the ionization of the gas increasingly "shorts out" the gap in the reactor vessel, causing reactor vessel capacitance to increase.
2. When secondary circuit V·A is multiplied by the power factor (p.f.) a true power figure only somewhat smaller than the primary circuit V·A figure is obtained. This suggests that primary circuit power is a reliable indicator of the power actually consumed by pure resistances in the resonant system.

3. The power factor is at its highest value when the voltage across the reactor vessel is not too far above the ionization potential of the gas in the reactor. This suggests that the CO_2 reduction process may proceed most efficiently in this region.

4. The very high V·A figure observed for the secondary circuit results from low power factor introduced by the capacitance, and may be effectively reduced by the use of a compensating inductance to form a resonant circuit.

Note: The formula used to compute resonant frequency is as follows:

$$\text{Resonant Frequency in Hertz} = \frac{1}{2\pi\sqrt{\text{Inductance} \cdot \text{Capacitance}}}$$

Inductance in henrys

Capacitance in farads

Data from 2/16/78

AC Kilovolts Applied to Reactor	Resonant Frequency Hz	Primary Circuit V · A	Secondary Circuit V · A	Ø, or Secondary Circuit Current Phase Shift Measured Degrees	Cos Ø, or Secondary Circuit Power Factor	$p.f. \cdot V_{sec} \cdot A_{sec}$	Calculated Reactor Capacitance Picofarads
1.0	2390	0.19	1.57	89.28	0.013	0.02	105
2.0	2390	0.74	6.18	87.48	0.044	0.14	103
3.0	2380	1.68	13.83	86.40	0.063	0.87	103
4.0	2070	10.15	29.44	72.00	0.309	9.10	141
6.0	1800	29.05	76.20	71.28	0.321	24.46	187
8.0	1740	49.00	143.84	76.68	0.230	33.08	206
10.0	1710	66.56	229.00	79.20	0.187	42.82	213
12.0	1700	93.48	336.60	80.28	0.169	56.38	217
14.0	1670	119.19	443.80	81.00	0.156	69.23	216

Power factor (p.f.) = cos Ø

Pressure: 41.3 kN/m^2 (310 mmHg)

Flow Rate: 50 ml/min, CO_2

Table 9 Resonance Characteristics

Optimizing the Efficiency of CO_2 Reduction:

As noted above, the power factor was best when the voltage across the reactor was only slightly higher than the ionization potential. This led to the thought that the CO_2 reduction process might proceed most efficiently at that same voltage, therefore a run was planned to test this possibility. Table 10 shows the results. The value of 3200 AC volts is the point where, at 41.3 kN/m^2 (310 mmHg) pressure, ionization is just maintained, so this became the starting point for a series of small voltage increments. As noted in the previous series of tests, the primary circuit V·A value was shown to be a reliable indicator of true power input to the system. The column showing "kilojoules/watt hrs per liter of CO_2 reduced" uses this as the basis for the computation. As is seen from this column, the most efficient reduction of CO_2 occurred at a potential about 20% higher than that required for ioniation, and at a very small primary circuit input power. The value of less than 360 kilojoules (100 watt hours) of energy per liter of CO_2 reduced represents the best level of performance that was achieved and could be consistently repeated.

Data from 2/17/78

					Primary Circuit		
Secondary AC Kilovolts	Resonant Frequency Hz	Primary V · A	Secondary V · A	% CO_2 Reduced	Kilojoules per Liter of CO_2 Reduced	Watt Hours per Liter of CO_2 Reduced	Calculated Reactor Capacitance Picofarads
3.20	2340	2.5	16.00	0.38	789.5	219.30	106
				0.34	882.4	245.10	
3.33	2310	3.71	18.32	1.08	412.2	114.51	114
				1.14	390.5	108.48	
3.48	2270	5.21	20.88	1.74	359.3	99.81	121
				1.70	367.8	102.16	
3.85	2120	8.10	26.95	2.84	342.3	95.07	137
				2.84	342.3	95.07	
4.24	2010	11.93	33.93	3.60	397.7	110.46	149
				3.64	393.3	109.25	
4.66	1950	16.19	43.80	4.36	445.6	123.78	165
				4.58	424.2	117.83	
8.58	1770	48.50	171.60	9.20	632.6	175.72	210
				9.16	635.4	176.49	

Pressure: 41.3 kN/m^2 (310 mmHg)

Flow Rate: 50 ml/min, CO_2

Table 10 Determination of the Most Efficient Condition for CO_2 Reduction

Summary of the Experimental Findings:

A concise overview of the experimental results is presented in the following capsule statements:

1. Increasing voltage or frequency applied to the reactor vessel increases the rate of CO_2 reduction with a limit of 30% conversion suggested by inspection of the data. Since the reactor vessel is an electrical capacitor, current "through" the vessel is in accordance with the laws for capacitive reactance.

2. With voltage and frequency applied to the reactor vessel at constant values, peak CO_2 reduction efficiency occurs between 40.0 and 53.3 kN/m^2 (300 and 400 mmHg) pressure.

3. CO_2 reduction is directly related to flow rate through the reactor vessel, with back reaction setting a limit of diminishing returns at very low flow rates.

4. Current is the mediator of the CO_2 reduction process, with voltage and frequency of interest only as inducers of current flow.

5. Gas ionization potential increases linearly as pressure increases, and this coupled with low CO_2 reduction rates at low pressures and a fixed voltage and frequency across the reactor vessel results in the "artifact" of peak process efficiency at 40.0 to 53.3 kN/m^2 (300 to 400 mmHg) pressure.

6. The electrical capacitance of the reactor vessel varies from a fixed base value at sub-ionization potentials to a higher, variable value caused by potentials in the ionization range. Current accounted for by the base value of capacitive reactance does not result in CO_2 reduction, while current accounted for by the difference in capacitive reactance in the ionized state is the sole mediator of the reduction reaction.

7. Temperature has little or no influence on the CO_2 reduction process.

8. The high currents through the reactor due to the base, or minimum, capacitance result in a low power factor. By establishing a resonant circuit with the transformer high voltage winding as the inductance power factor was improved and the power input to the resonant circuit was much lower than to the reactor vessel alone.

9. By operating the reactor vessel as part of a resonant circuit, and applying a voltage that was about 20% higher than the ionization potential, the overall efficiency was high enough to permit the repeatable reduction of one liter of CO_2 with an energy input of 342 to 360 kilojoules (95 to 100 watt hours).

Feasibility Study

A feasibility study was conducted to determine whether, based on our preliminary test data on the plasma CO_2 reduction process, we could recommend to NASA that this process has sufficient potential to warrant further development.

A brief review of the U.S. literature revealed that no other new CO_2 reduction technology has been introduced in the past eight to ten years. Furthermore the only other advanced technology currently being pursued is the solid electrolyte process[4] which was the selected CO_2 reduction process in the AILSS study[5]. The use of the well-developed Sabatier process is planned for the Regenerative Life Support Experiment (RLSE) preprototype test bed at NASA-JSC, and there are, to our knowledge, no active plans to pursue the Bosch process.

In comparing the plasma with the solid electrolyte process, we only considered power because both processes, and in particular the plasma process, are in early phases of development and hardware weight estimates could not be considered valid at this point in time.

Tests conducted in our laboratory plasma reactor consistently produced results indicating that, for this particular reactor configuration, power requirements in the range of 95 to 100 watt hrs or 342 to 360 kilojoules per liter of CO_2 reduced were obtained. It is significant that this performance was achieved at ambient temperature with rudimentary electronics and an arbitrary reactor configuration.

For the solid electrolyte process, which operates at in excess of 900°C, we used literature data[6] and estimated the power requirement to be in the range of 56 watt hours or 202 kilojoules per liter of CO_2 reduced. This estimate is based on data in Reference 6, and the calculation was made as follows. For a given electrolyzer furnace skin temperature of 120°F, a 1562°F operating temperature, an external area of 48.4 ft. and eight inches of Johns-Manville Micro Quartz insulation ($k = 0.6$);

$$Q = KA\Delta t/X = (0.6)(48.4)(1442)/8 = 5234 \text{ Btu/hr}$$
$$= 1536 \text{ watt/hr}$$
$$= 5.53 \text{ megajoules}$$

If we take into account a typical efficiency of 50% for penetrations the heater power becomes Q = 1536/0150 = 3072 watts. From this, we subtracted 50% of the electrolyzer power (153 watts) which is attributed to heating due to resistive losses.

The total power for the one man electrolyzer and its furnace heating is then (3072-153) + 306 = 3225 watts. This corresponds to 112 watt hrs or 403 kilojoules per liter of O_2 generated or 56 watt hrs or 202 kilojoules per liter of CO_2 reduced.

Simple block diagrams of these two processes are shown in Figure 7 to illustrate their similarities and differences. Both require a high temperature CO disproportionator and a gas separator. In the case of the plasma system, a room temperature O_2 concentrator estimated at 135 watts (for a one-man system) is required. For the solid electrolyte system a 175 watt, 343°C hydrogen separator(6) is required.

Based on the above favorable comparison, it is recommended that further development of this plasma CO_2 reduction technology in the areas described in the next section be pursued.

It is interesting to note that when the heats of formation for CO and CO_2 are used to compute the absolute amount of energy for the reduction of one liter of CO_2, a value of 3.51 watt hours/liter, or 12.64 kilojoules is required at standard conditions of 760 mmHg (101.3 kN/m^2) and 0°C. When this figure is corrected to allow for the greater volume of the gas at 20°C, the approximate temperature at which the experimental work was carried out, an energy requirement of 3.29 watt hours or 11.94 kilojoules per liter of CO_2 reduced is arrived at.

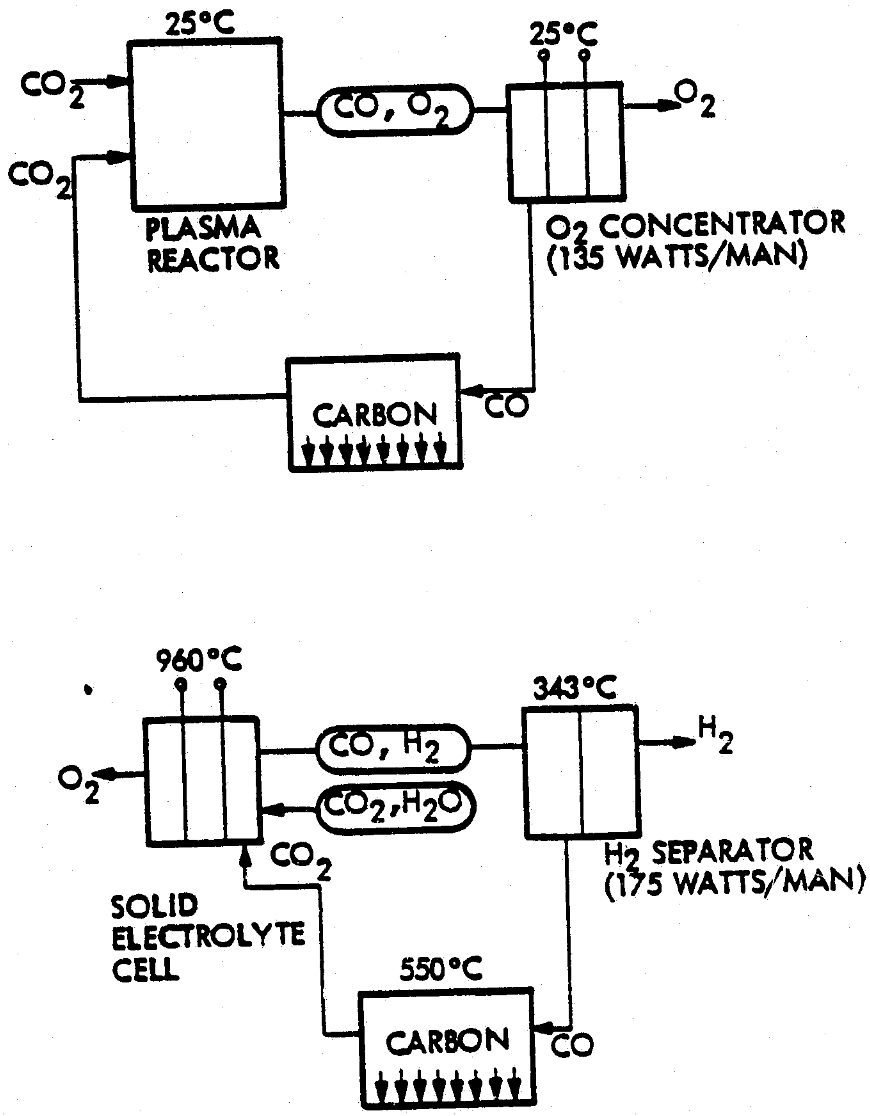


FIGURE 7 BLOCK DIAGRAMS OF THE SILENT ELECTRIC DISCHARGE AND SOLID ELECTROLYTE CO_2 REDUCTION METHODS.

Recommendations

As shown in the Feasibility Study section of this report, the reduction of CO_2 in a silent electric discharge is about a factor of two more costly in energy than the solid electrolyte method which is a potential competitor. The equipment used in the experiment resulting in this report, while adequate for the purpose of investigation, does not represent any effort to refine this CO_2 reduction method to it's practical limit.

Two areas could be explored in further work; the improvement of the electrical subsystem, and the radical development of the reactor vessel to increase the volume processing capability and the efficiency of operation.

The electrical subsystem can probably be improved with a modest effort, since its requirements are determined primarily by the configuration and operating requirements of the reactor vessel and represent no demand on new technology.

It is anticipated that improvements in the reactor vessel would include narrowing the air gap to permit operation at much lower voltages, increasing the area of one electrode while decreasing the area of the other so as to reduce the base capacitance and increase the ionized capacitance, exploration of the effect of thinner insulating materials, and insulating only the larger of the two electrodes. It is possible that a highly evolved reactor device, with an associated inductance, could operate directly from spacecraft 400Hz power, thus eliminating all losses previously encoutered in generating the high voltages. In any event it is possible to foresee many modifications that would be expected to enhance the reactor device efficiency.

In view of the encouraging results obtained during this experiment with an assembly of non-specialized electrical gear and a reactor vessel of rudimentry design it is virtually certain that further study of the process and development of the equipment would lead to a substantial improvement over the efficiency of CO_2 reduction obtained in this study.

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