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Technical Progress Report For the Period
October 1, 1990 - March 7, 1991

On the Development of a Chemical
Kinetic Measurement Apparatus and
the Determination of the Reaction Rate Constants
For
Lithium-Lead/Steam Interaction

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ABSTRACT

An experimental set-up for accurate measurement of hydrogen generation rate in Lithium-Lead ($\text{Li}_{17}\text{Pb}_{83}$) Steam or water interactions is being designed. The most important features of the design include a solenoid actuated quick opening and closing butterfly valve used to control the reaction time and the placement of all measuring devices below a water line to minimize leakage of the hydrogen collected.

I. INTRODUCTION

This technical progress report covers work done on this project for the period October 1, 1990 through March 7, 1991. Graduate and Undergraduate Research Assistants were assigned to the project in October of 1990, after which some time was spent to bring the students up to date on some important theoretical and design aspects needed to achieve project objectives.

The greatest challenge in the design is ensuring a system with minimum leakage rate in order to obtain accurate accounting of reaction product gases. This greatly affects the placement of thermocouples, transducers and other instruments needed in the design.

A. Objectives of Research

The objectives of the research are:

- (i) The design and construction of an experimental set up that can be used to obtain accurate measurements of hydrogen generation rate during the initial and subsequent stages of liquid metal/water or steam reactions.
- (ii) The use of data collected in conjunction with an improved liquid metal transport reaction model developed to determine the empirical chemical kinetics reaction rate constants.
- (iii) To involve minority graduate and undergraduate students in this research.

B. Project Schedule

To enable the objectives listed above to be met, the overall project was divided into 10 main steps, namely

1. The search for qualified graduate and undergraduate students for the research and their subsequent introduction to the project.
2. Preliminary design of experimental system and determination of experimental test matrix.
3. Identification of manufactures/vendors and price list for all major components in preliminary design.
4. Modification of preliminary design. The final design of the experimental system.
5. Procurement of components and modification of Liquid Metal Transport Reaction Model for experimental analysis.
6. Construction of experimental system.
7. Testing & correction of experimental system.
8. Data Collection on Hydrogen generation rate:
 - (a) Liquid Lithium-lead/water reaction
 - (b) Liquid Lithium-lead/Steam reaction

9. Analysis of Data using the modified liquid metal Transport Reaction Model to obtain the empirical reaction rate constants.
10. Preparation of final Research Report.

Figure 1 shows the project schedule for the first year of the project. The details of the work done on the first three phases of the project are described in this report.

C. Graduate and Undergraduate Research Assistants.

The first phase of the project involved obtaining qualified graduate and undergraduate students to work on the project. In this regard, James Akpan, an MS graduate student in the Mechanical Engineering Department was appointed a graduate Research Assistant and is working on this research for his MS Thesis. In addition, Robert Tetteh, who has been voted as the most outstanding undergraduate Mechanical Engineering Junior student, expressed great interest in the research work and was hired as an Undergraduate Assistant in the project.

While James will primarily be in charge of the overall research, his concentration will be in the more technical aspects of data acquisition, analyses and chemical kinetics of the work. Robert will be involved in the design of the system, procurement of components and assembly of the system. James and Robert have spent a considerable portion of the first phase of the project in literature research into data acquisition, liquid metal/water/steam reactions, chemical reaction kinetics and fusion safety in general

Figure 1 First Year Project Schedule

II. PRELIMINARY DESIGN OF EXPERIMENTAL SYSTEM

A. Experimental Test Matrix

To enable us determine the operating condition of the system to be designed, a detailed experimental matrix was developed. The primary variable to be controlled during the experiment are:

1. Initial Liquid Metal Temperature T_{LM}
2. (a) Initial Water Temperature T_w or
(b) Initial Vapor temperature T_v
3. Initial System Pressure P_i
4. Total reaction time t_r and
5. Initial Mass of liquid Metal M_{LM}

To ensure that the surface area for reaction is constant throughout an experiment, the area will be the maximum that prevents Taylor instability. The Secondary variables to be measured include:

1. Reaction product gas temperature T_g
2. Reaction product gas pressure P_g
3. Hydrogen Concentration in reaction product gas C_{H_2} .
4. Liquid Metal Temperature.

The complete preliminary proposed experimental matrix is shown in Table 1.

Table 1 Preliminary Experimental Test Matrix

Liquid Metal Temperature C	Initial Water Temperature C	Reaction Time s	Liquid Metal Mass gm
350	90	0.5,1.0,2.0,30	60
350	90	0.5,1.0,2.0,30	30
350	80	0.5,1.0,2.0,30	30
350	70	0.5,1.0,2.0,30	60
400	60	0.5,1.0,2.0,25	50
400	60	0.5,1.0,2.0,25	50
400	90	0.5,1.0,2.0,25	40
400	90	0.5,1.0,2.0,25	70
400	70	0.5,1.0,2.0,25	50
400	80	0.5,1.0,2.0,25	20
400	90	0.5,1.0,2.0,25	20
400	60	0.5,1.0,2.0,25	30
400	60	0.5,1.0,2.0,25	30
500	90	0.5,1.0,2.0,20	60
500	90	0.5,1.0,2.0,20	30
500	70	0.5,1.0,2.0,20	30
600	90	0.5,1.0,2.0,15	50
600	90	0.5,1.0,2.0,15	50
600	70	0.5,1.0,2.0,15	20
650	90	0.5,1.0,10	50
650	80	0.5,1.0,10	50
650	70	0.5,1.0,10	50

B. Original Proposed System

Figure 2 shows the initial conceptual schematic diagram for the experimental system. The main characteristics are a liquid metal chamber A, steam/Water/Reaction product chamber B, data acquisition system components G,H,I, thermocouples J,K,Q; Constant temperature bath L, steam generator M, after heater N and the most important component of the set-up, the programmed sliding leaf valve actuated by a solenoid through a control system. Other components of the set-up include the valve actuator E, the valve controller, F, a pressure transducer V, a convection coil D, argon bottle S, and heaters R and auxiliary product vessel W. For Liquid metal/Steam reaction, steam generated by M is heated in N and introduced into the steam chamber B. The temperature and pressure of the steam is measured by thermocouples J,K, & pressure transducer V respectively. When the liquid metal in reaction chamber A reaches a predetermined temperature, a signal is sent to the leaf valve actuator and the valve E is opened for a predetermined period of time and then closed enabling the reaction time to be controlled precisely. The hydrogen produced during the reaction period collects in chamber B. The steam remaining in Chamber B is condensed using the convection coil & the constant temperature bath and the amount of hydrogen produced determined. Any hydrogen produced after the leaf valve is closed is directed into the secondary hydrogen collection tank W. For liquid metal/water reaction, the process is similar except that an amount of water is measured and poured into the water chamber B. The temperature of the water is

Figure 2 Initial Conceptual Design of Experimental System

controlled by the convection coil and the constant temperature bath. In this design, the thermocouples, Convection Coils and pressure transducer all enter the system through a flanged top section of the apparatus and in direct contact with the gases collected in the top chamber of the system. We anticipated having hydrogen gas leakage problems with this design and as a result several changes have been made.

C. Modified New System Design

Figure 3 shows the current design under consideration. In the new design, significant changes have been made in the upper chamber and mid-section area of the apparatus.

Upper Chamber

The attachment of the transducers, thermocouples, and the convection coil have been located below the water level to minimize leakage of the hydrogen gas produced. The transducer and thermocouple wells are being modified by replacing them with a design that allows direct contact of the thermocouples with the gas and water regions. The convection coil is passed through a flanged part at the base of the upper chamber and below the water level. The principle behind these changes are that liquid leakages are far easier to control than hydrogen gas leakage. Thus we want to ensure that there are no flanged or screwed joints in the region where the gas collects after the reaction.

Figure 3 Current Preliminary Design of Experimental System

Mid-Section of Chamber (butterfly valve)

The solenoid actuated leaf valve in the original design of figure 2 has been replaced by a butterfly valve chosen due to its negligible leakage characteristic, sealing capacity and cycle time (250 ms). The upper and lower sections of the apparatus are connected to the butterfly valve through a flange with a series of o-rings used to minimize leakage. The flange connection also facilitates easy disconnection of the lower portion of the apparatus for the purpose of loading and unloading the liquid-metal into and out of the chamber in a glove box. To ensure that the butterfly valve and its actuator are operated within their specified temperature range, the valve section is water-cooled on the outside.

D. Preliminary Determination of Hydrogen Gas Concentration and Pressure in the Gas Region of the Upper Chamber

In order to select the right type of hydrogen gas analyzer for accurate measurement of concentration of hydrogen gas in the gas region, a preliminary analysis was made to obtain the range of hydrogen gas concentration that may be encountered in the course of the experiment. This analysis was also used to provide estimates of gas pressures in the upper gas region for design purposes.

The analysis was based on the following:

1. The mass of liquid metal initially in reaction vessel is known.
2. The percent of metal reacted was varied from 1% to 20% (a typical range for such non-mixing reactions)

3. The upper chamber gas region volume was varied by varying the water level in the upper chamber (of diameter 5 cm) to produce gas volume heights of from 1 cm to 20 cm
4. Saturated liquid is assumed to be in the upper chamber, and the liquid temperature was varied from 60 to 100 C.

The above information are used in conjunction with the chemical reaction equation for lithium-lead ($\text{Li}_{17}\text{Pb}_{83}$)/water reaction to determine the partial pressure and mole fraction of hydrogen generated during the reaction. Tables 2 and 3 give sample calculation results for ($\text{Li}_{17}\text{Pb}_{83}$)/water reaction. In the tables, the first column gives the height of the gas accumulation section of the upper chamber, the second column gives the partial pressure of hydrogen in the gas column and the last column gives the mole fraction of hydrogen gas in the gas column. The percentage reaction assumed and the initial saturated liquid temperature are also indicated on the Table. Similar tables have been developed for other ranges of initial water temperatures and percent reaction. From the table, it is evident that in addition to the original purpose of the water in serving as a reactant and condensing medium for any water vapor generated as a result of the reaction, the variation of its column height will directly change the gas volume above it and hence can be used as a means of controlling the total pressure generated by hydrogen production and accumulation in the gas region. In doing so, we can also have control over the partial pressure of the hydrogen and therefore its concentration. This will enable us to provide hydrogen gas concentration in a range required by the hydrogen gas analyzer.

Table 2 Pressure and Mole Fractions of Hydrogen Generated for
 $\text{Li}_{17}\text{Pb}_{83}$ /Water Reaction, $T_w = 80^\circ\text{C}$, 5% Reaction

Gas accumulation height (cm)	H2 Partial Pressure (MPa)	H2 Mole Fraction
2	0.079	0.049
3	0.053	0.048
4	0.040	0.047
5	0.032	0.047
6	0.026	0.046
7	0.023	0.045
8	0.019	0.045
9	0.018	0.044
10	0.016	0.044
11	0.015	0.043
12	0.013	0.042
13	0.012	0.042
14	0.012	0.041
15	0.011	0.041
16	0.010	0.040
17	0.010	0.040
18	0.009	0.040
19	0.009	0.039
20	0.008	0.039

$\text{Li}_{17}\text{Pb}_{83}/\text{H}_2\text{O}$ reaction
 Water temperature = 80°C
 Mass of $\text{Li}_{17}\text{Pb}_{83}$ = 43 g
 % Metal Reacted = 5
 Initial System Pressure = P_{sat} at 80°C

Table 3 Pressures and Mole Fractions of Hydrogen Generated for
 $\text{Li}_{17}\text{Pb}_{83}$ /Water Reaction, $T_w = 100^\circ\text{C}$, 10% Reaction

Gas accumulation height (cm)	H2 Partial pressure (MPa)	H2 mole fraction
2	0.166	0.094
3	0.111	0.092
4	0.083	0.089
5	0.066	0.087
6	0.055	0.085
7	0.047	0.083
8	0.041	0.080
9	0.037	0.079
10	0.033	0.077
11	0.030	0.075
12	0.028	0.074
13	0.024	0.071
14	0.022	0.071
15	0.022	0.069
16	0.021	0.068
17	0.019	0.066
18	0.018	0.064
19	0.017	0.063
20	0.017	0.063

$\text{Li}_{17}\text{Pb}_{83}/\text{H}_2\text{O}$ reaction
 Water temperature = 100°C
 Mass of $\text{Li}_{17}\text{Pb}_{83}$ = 43 g
 % Metal Reacted = 10
 Initial System Pressure = P_{sat} at 100°C

E. Finite Element Heat Transfer Model for Lower Portion of System

The experimental determination of the rate of hydrogen production during Lithium-Lead/Water reaction involves two distinct stages, the heating stage and the reaction stage. During the heating stage, the lower portion of the apparatus is electrically heated to melt the lithium-lead and to bring it to the required initial temperature T_{LM} . The reaction stage begins when the butterfly valve is opened and water from the upper chamber makes contact with the liquid metal in the lower section. This stage ends when the butterfly valve is closed at the end of a pre-determined reaction time, t_r . During the heating stage, it is important to ensure that both the butterfly valve and the electrical resistance heating elements are maintained within their stated operating temperature range. It is also important to ensure that right size heaters and adequate insulation is provided to obtain a reasonable heating time (30-45 minutes). These require that a heat transfer analysis be done at the early design stages of the system to ensure that the system under design will satisfy the required constraints. A two-dimensional finite element software is currently being used to analyze the heat transfer characteristics of the lower portion of the apparatus during the heating stage. This will help in the final selection of the right type of heater, insulation and the cooling system for the butterfly valve if needed.

III. EQUIPMENT PROCUREMENT

At this point in the design stage, we have not purchased any of the components needed for the design. However, we have spent considerable amount of time in identifying manufacturers/vendors, obtaining prices and technical information on the components we shall need in the design. Appendix A contains a table that gives the details of equipment needed, names and addresses of manufacturers/vendors and prices for those components.

IV. PROBLEMS ENCOUNTERED AND FUTURE DIRECTIONS OF RESEARCH

A. Problems Encountered In the first Six Months

Two major problems were encountered during the first six months of the project, all being the result of the project contract date occurring almost a month after beginning of the semester. The principal investigator was unable to obtain release time to actively work on the project in the Fall Semester of 1990 (August 28 - December 31, 1990) because of the late contract date. A college technician could not be assigned to work on the project in the Fall Semester of 1990 as a result of the late contract date. We could also not get the college technician to work on the project in the Spring Semester of 1991 (January - May 1991) due to heavy work load already being assigned by the college. Plans are currently being made to obtain the college technician to work 25% time on the project in the Summer Semester (June - August 1991) or hire a part-time outside Technician.

B. Future Direction of Research

We anticipate completing all remaining stages on the first year project schedule in the remaining six months of the first year. In addition, a lot more can be accomplished during the Summer months of June - August 1991, because the principal investigator will work on the project on a 75% time basis during those three months. The overall project objectives have not changed. Figure 4 gives the anticipated second year project schedule. After the design and testing phases of the first year, the second year will be mainly devoted to data collection, analyses of experimental data and determination of chemical reaction rate constants for the lithium-lead/water reaction. The final project report will also be prepared at this time and the graduate student will complete his MS Thesis.

Figure 4 Second Year Project Schedule

V. CONCLUSIONS

The first six months of the research has been very productive. One graduate and one undergraduate research assistants have been assigned to the research. The students have been brought up to date on the theoretical and design aspects of the research. Regular brainstorming sessions, organized by the research team, have lead to several improvements in the original design, the major of which is the decision to let measuring devices penetrate the test section below the water-level and to eliminate any screw or flanged joint in the upper portion above the water level to minimize leakage of hydrogen from the apparatus.

Manufactures, Vendors, and price list for all major components have been identified. A finite element model of the lower section has been developed and is being analyzed for the heating stage. This would enable us to select appropriate heaters, insulators and cooling mechanism for the butterfly valve section. An experimental test matrix has been developed. A detailed Project Schedule has been developed for the first and second years of the project and is being followed. Funding for the second year will therefore, facilitate a successful completion of the project objectives.

APPENDIX A

Table 4 Equipment Procurement List

NAME OF PART	COMPANY NAME ADDRESS/PHONE	COMMENTS/PRICES
1. K-LOK HIGH PERFORMANCE VALVES SIZES: 2.5 - 36in	KEYSTONE VALVE Inc. 9700 WEST GULF BANK DRIVE HOUSTON TX. 77240 713-466-1176	\$ 976.00
GREENWOOD BUTTERFLY VALVE SIZES: 2 - 12in	KEMPER VALVE & FITTING CORP. 11335 TODD ROAD. HOUSTON TX. 77055 713-682-7400	\$ 1036
2. 3-WAY ACTUATED VALVES SIZES: .5 - 36in METAL SEATED FLANGED BALL VALVES. REGULAR VALVES SIZES: 0.5 - 8In TEMP 932 F PRESS 700 Psi	KEYSTONE SALES Inc. 6230 W. 34th STREET HOUSTON TX. 77218 713-680-8800 KTM INDUSTRIES. Inc 16610 BARKER SPRINGS ROAD HOUSTON TX. 77218 713-492-8800	TO BE CUSTOM MADE ACCORDING TO SPECS. PRICE NOT PRESENTLY AVAILABLE

NAME OF PART	COMPANY NAME ADDRESS/PHONE	COMMENTS/PRICES
3. TRANSDUCERS (PRECISE QUALITY) .HIGH TEMP .HIGH PRESS. .H2 COMPATIBLE .HIGH FREQ. RESPONSE.	PRECISE SENSORS, inc 235 WEST CHESTNUT AVE. MONROVIA CA. 91016 818-358-4578/ 800-423-7123 OMEGA ENGINEERING inc. ONE OMEGA DRIVE. STAMFORD, CN. 06907 800-826-6342	\$ 200 - \$400 FOR HIGH PRESS. > 10,000 psi
BRANDT I/P TRANSDUCERS (CURRENT TO PRESS)	KEYSTONE SALES 6230 W. 34th ST.. HOUSTON TX. 77092 713-680-1564	
(4) THERMOCOUPLES/ THERMAL WELLS	GRAYSCO THERMOCOUPLES 2859 WEST SIDE, HOUSTON, TX. 713-941-8540 OMEGA ENGINEERING inc ONE OMEGA DRIVE STAMFORD, CN. 06907 800-826-6342	\$ 360, THEMOCOUPLES \$ 200. WELLS

NAME OF PART	COMPANY NAME ADDRESS/PHONE	COMMENTS/PRICES
5. CONVECTION COIL CONSTANT TEMP. BATH	COLE-PALMER INSTRUMENTS 800-323-4340	\$ 150. COIL \$ 4225. (12-LITER BATH)
(6) STEAM GENERATOR.	COLE-PALMER INSTRUMENTS 7425 N. OAK PARK AV. CHICAGO, IL 60648 800-323-4340	\$ 1970.00
(7) ARGON CYLINDERS . HIGH PURITY . 3500psig . 99.998% min . 386 cu ft.	MATHESON GAS PRODUCTS P. O. BOX 969 LAPORTE, TX. 77672 713-471-2644	\$ 175.00, CYLINDER \$ 400.00, GAS
(8) PIPE & CAP LENGTH: 62cm DIA: 5cm THICKNESS: 1cm	CARBON-STAINLESS STEEL ALUMINUM TANKS/PIPING 4403 THEISS. HOUSTON, TX. 713-821-0543	PURCHASE FROM MOBIL PIPE DISTRIBUTORS AND CUT/WELD ACCORDING TO GIVEN SPECS. PRICE TO BE DETERMINED AFTER JOB IS DONE.

NAME OF PART	COMPANY NAME ADDRESS/PHONE	COMMENTS/PRICES
(9) HEATERS MANTLE-ELECT. >1000 DEG C.	COLE-PALMER INSTRUMENTS 7425 N. OAK PARK AVE. HOUSTON, TX. 800-323-4340	\$ 220.00
(10) INSULATORS (FIBER GLASS)	SPECIALTY PRODUCTS AND INSULATION CO. 6500 LONG POINT HOUSTON, TX. 713-957-2730	\$ 2.11/LINEAR ft. TOTAL: \$ 50.00 FOR PIPE, LOWER CHAMBER HEATER & STEAM GENERATOR
(11) COMPUTER/ DATA ACQUISITION SYSTEM	VALCOM COMPUTER CENTER 3400 FONDREN HOUSTON, TX. 77063 713-781-1900 COMPUCON SYSTEMS 4111 DIRECTORS ROW # 100 HOUSTON, TX. 77092 713-681-2200	\$ 6,340.00 (PC ONLY) \$ 6,149.00 (PC ONLY)
(12) HYDROGEN ANALYZER THERMOR 6N (TYPE)	ROSEMOUNT ANALYTICAL 1201 NORTH MAIN ST. ORRVILLE, OH 44667 216-682-9010	\$ 4,500.00

NAME OF PART	COMPANY NAME ADDRESS/PHONE	COMMENTS/PRICES
(13) LITHIUM/ LITHIUM-LEAD	MERECK CHEMICAL DIV. 111 CENTRAL AVE. TETERBORO, NJ. 07608 201-288-2292	\$ 1,500.00 (LITHIUM-LEAD)
(14) GLOVE BOX INTERNAL DIM. ACRYLOGEN TYPE 42 X 26 X 28in CHEMICAL TYPE 14 X 11 X 3in	COLE PALMER INSTRUMENTS 7425 N. OAK PARK AVE. CHICAGO, IL 60648 800-323-4340	 \$ 9,050.00 \$ 8,450.00
NB: PRICES ARE SUBJECT TO CHANGE WHEN FINAL VENDORS ARE CHOSEN		

END

**DATE
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12/19/91