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"SELECTION AND EVALUATION OF CARBON-RESIN COMPOSITES FOR BIPOLAR PLATES FOR HYDROGEN FUEL CELLS"

FINAL TECHNICAL REPORT

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U.S. ARMY MOBILITY EQUIPMENT RESEARCH & DEVELOPMENT COMMAND

FORT BELVOIR, VIRGINIA 22060

CONTRACT NO. DAAK70-77-C-0151

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REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER

2. GOVT ACCESSION NO.

3. RECIPIENT'S CATALOG NUMBER

4. TITLE (and Subtitle)
Selection and Evaluation of Carbon-Resin Composites for Bipolar Plates for Hydrogen Fuel Cells

5. TYPE OF REPORT & PERIOD COVERED
July 1977 to September 1978

6. PERFORMING ORG. REPORT NUMBER

7. AUTHOR(s)
P.M. Colling
R.V. Norton
J.E. Martin

8. CONTRACT OR GRANT NUMBER(s)
DAAK70-77-C-0151

9. PERFORMING ORGANIZATION NAME AND ADDRESS
Ashland Chemical Company
P. O. Box 2219
Columbus, Ohio 43216

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
6.37.02A, 1L263702DG10, 03, 014

11. CONTROLLING OFFICE NAME AND ADDRESS
U.S.Army Mobility Equipment Research & Development Command, Fort Belvoir, VA. 22060

12. REPORT DATE
1978

13. NUMBER OF PAGES
93 p.

14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)
Final rept. Jul 77-Sep 78

15. SECURITY CLASS. (of this report)
Unclassified

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)
Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)
Bipolar plates, fuel cells, composite graphite, resins, electrical conductivity, phosphoric acid, high temperature stability

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)
Resin-graphites composites were developed for use in molding bipolar plates for fuel cells. The composite is in excess of 150°C, has high electrical conductivity, adequate flex strength and high dimensional stability.

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

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BIPOLAR PLATE FINAL REPORT

S U M M A R Y

"SELECTION AND EVALUATION OF CARBON-RESIN COMPOSITES FOR BIPOLAR PLATES FOR HYDROGEN FUEL CELLS"

The goal of this contract was to identify and to develop resin-graphite composites that are resistant to H_3PO_4 at temperatures in excess of 150°C, that have high electrical conductivity, that have adequate flex strength, and that have high dimensional stability for use as a bipolar plate in a fuel cell.

1. The program evaluated sample coupons (1 x 3 x 1/8") prepared from experimental and commercial resin systems with commercial graphite samples. The interaction of the components was determined and particle packing was used to select the graphite particle size distribution.

2. Carbon-14 labelled phenol-formaldehyde resins were used in preparing composite test coupons that were subsequently extracted with phosphoric acid at >180°C. The radioactive tracer technique demonstrated that a significant level of organic C_{14} materials are leached from the bipolar plates by the phosphoric acid. The extractable substances (unidentified) may be a significant factor affecting the lifetime and efficiency of a fuel cell.

3. A two-step molding procedure for the preparation of bipolar plates was developed. The procedure utilizes a hot outgasing step to release gaseous products of molding. It is recommended that the U.S. Army evaluate manufacturing bipolar plates using this procedure. The procedure eliminates structural flaws that result from gas evolution during the molding step of the coupons.

4. An improved resin-composite system was developed using test coupons 1 x 1/2 x 3".

5. Recommended resin composition for molding of bipolar plates:

Resin: EP-032217-A-33 (Styrene modified Arofene 890)	25 Wt. %
Catalyst: Hexamethylene tetramine	6 Wt. %
Graphite:	
Asbury 7101	50 Wt. %
Asbury 4015	40 Wt. %
Asbury Micro-250	10 Wt. %

Molding Conditions: Two-step process; 4000 psi*
Post curing at 450°F (20 hours)

*(Molding process not optimized for large plates)

I. INTRODUCTION

A. Background:

The U.S. Army, Fort Belvoir, has underway an active research program for the development of fuel cells for portable and remote equipment in the 0.5 to 5 Kw rating class that will have a field service operational life of 2000 to 4000 hours. Performance problems associated with both the electrode surfaces and the bipolar plate have been previously identified as contributing factors to the design lifetime. The bipolar plate, made of a graphite resin composite, serves to separate the individual galvanic cells and to conduct the electrical current from cell to cell.

An earlier study (DAAK02-74-C-0367, June, 1977) cited the bipolar plate as a contributor to the short lifetime of fuel cells and mechanical weakness of the composite bipolar plate was discussed (Final report ERC-7396-IV, June, 1977, page 18). The existing bipolar plates were reported to soften and to swell around the edges when in contact with phosphoric acid in a fuel cell. Problems associated with dimensional stability of the plate resulted in excessively high manufacturing rejects and consequently increased the cost of a bipolar plate.

A program to select and evaluate various resin/graphite composite systems and to determine physical properties of coupons under conditions approximating those in actual use was undertaken by Ashland Chemical Company to improve the bipolar plates. A design condition of 200°C was specified as the most stringent requirement for the 1985 time-frame. If, as a result of this program, bipolar plates could be developed to meet the 200°C phosphoric acid operating conditions, then the present operating problems at 150°C should be significantly diminished.

B. Objective:

The objective of this program was to screen, to select, to prepare and to evaluate various graphite/resin composites that could be molded as bipolar plates for hydrogen fuel cells and identify an optimum composition.

A secondary objective, developed during the six month review of the project with MERADCOM, was to quantify the amount of extractable materials leached from the bipolar plates with hot phospohric acid using radio tracer techniques.

C. Research Program:

The research program, as contracted for, comprised 5 tasks:

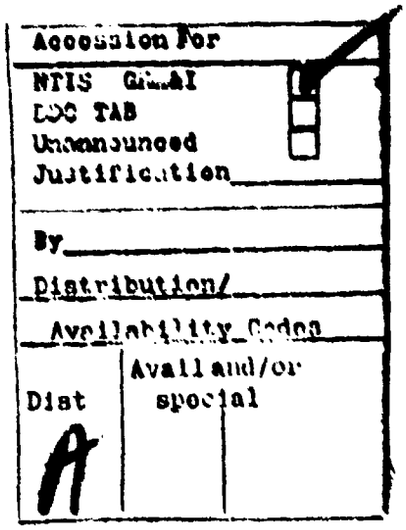
Task 1: Preliminary screening of resin and carbons based upon published sources and in-house information.

Task 2: Screening of resins by phosphoric acid endurance tests to select the two or three resins showing the best phosphoric acid resistance among those tested.

Task 3: Screening of carbons and determination of practical ranges for carbon loading to select those carbon compositions and loadings giving the highest electrical conductivity.

Task 4: Composite evaluation to determine the effects of resin type, carbon type, and carbon loading among those selected in Task 2 and 3.

Task 5: Final report.



II. INVESTIGATION

A. Task 1: Preliminary Screening of Resins/Carbon black/ graphite materials

1. Objective

The objective of Task 1 was to screen and select resins and carbons suitable for use in a bipolar plate based on published properties.

2. Preliminary screening of resins involved elimination of those resins whose properties precluded their use in hot phosphoric acid (See First and Second Monthly Progress Reports). Phenolformaldehyde novolak resins were selected as the chemical system for further evaluation because of their known performance and moldability characteristics.

A laboratory program for thermal gravimetric analysis of a series of phenolformaldehyde resins helped to screen them on the basis of thermal stability . The observed differences were minor so none of the resins were elminiated on the basis of TGA alone. The TGA results are reported in the Fourth and Fifth Monthly Progress Reports.

3. Carbons were evaluated on the basis of electrical conductivity. Graphite was an obvious candidate, but certain carbon blacks were evaluated. Two types of carbon black representing the extreme ends of the range of carbon black properties were evaluated: (a) Cabot XC-72, a high conductive, high structure and large surface area carbon black made especially for electrical applications; (b) medium thermal black, a large particle diameter, low surface area, low structure black. Medium thermal was selected because it was thought that it could effectively fill the interstitial spaces between the larger graphite particles yielding higher packing density and electrical conductivity (Fifth & Sixth Monthly Progress Reports).

Natural and synthetic graphites and blends of different graphites were tested to achieve the desired balance of properties of the molded part. Discussions were held with Asbury Graphite Mills and they recommended several graphites to be tested in the program: Asbury 4533, 4110, A-60, 289-A, and 5131. The Asbury graphites possess a wide range of properties and further development to optimize the particular graphite blend was undertaken as Task 3.

B. Task 2

1. Objective

The objective of Task 2 was to screen resin composite coupons using a phosphoric acid endurance test and to select the two or three resin composites showing the best phosphoric acid resistance.

2. Screening Tests (Fifth, Sixth, Seventh Monthly Progress Reports)

Twelve Novolak resin systems were tested in hot phosphoric acid at 200°C. The 12 samples represented different resin systems at catalyst levels of 6 to 14%. As reported in the Fifth Monthly Progress report (attached) the two compositions that completely failed both contained 14% hexamethylene tetramine. Experimental resin systems were developed and tested in this phase of the program.

(a) Relative Performance of different resins (Seventh, Eighth, Ninth Monthly Progress Reports)

A 33% binder level was selected to represent a commercially practical resin level but the 33% binder level may not have been optimum. Experimentation was also done at lower binder levels to determine the minimum effective amount.

Arofene 875 and 877 at 10% hexa level was found to be slightly more resistant to phosphoric acid than a commercial control sample in terms of weight loss. Arofene 860 and the experimental resin 833 showed approximately the same weight loss as the commercial material, while Arofene 865 exhibited a relatively large weight gain. It appeared that the resins were about equal in resistance to phosphoric acid in the coupon test using a single step molding procedure. The resistance of the coupon was improved by decreasing the catalyst level and using a two-step molding procedure (Fifth and Eighth Monthly Progress reports).

As discussed in Section B3, the improved molding procedure developed later in the program resulted in a dramatic improvement in the performance of the overall coupon and its resistance to phosphoric acid. Using the new molding procedure the different resin systems that heretofore had appeared almost equal, showed different resistances to H_3PO_4. As reported in the Ninth Monthly Progress report, phosphoric acid endurance tests were terminated on the first 30 coupons (single-step molding) after approximately 2000 hours so that flex strength could be measured.

The final results of Task 2b are tabulated in Table I of the Eleventh Monthly Progress report. The breaking strength is generally lower after aging than before. For example: in 61 of the 78 cases, the aged coupon had lower breaking strength than the unaged coupon but scatter was significant. The physical appearance column of Table I (Eleventh Monthly Progress report) is a subjective evaluation based on numerical rating of 0 to 5; and a description of the physical appearance of the aged coupon. As determined by this first phase of the program on different resins, the final tasks used only resins Arofene 872, Arofene 877, Arofene 890, and Arofene 833. Controls were Colloid 844 and Commercial samples #1 and #2. Task 4 involved a detailed evaluation of the composites prepared from these selected resin systems (Seventh and Twelveth Monthly Progress reports).

(b) Effect of catalyst (hexamethylene tetramine) level on physical properties

Table I of the Sixth Monthly Progress report details the results from the study of catalyst levels used for Arofene resins 875, 877, 860, 2869, and the commercial control. All compositions had a 33% binder level. Table I (Eighth Monthly Progress report) summarized the data for the 10 resins systems studied at catalyst levels of 3, 6, 10, and 14 weight percent of catalyst.

The phosphoric acid endurance of the composite was better at lower hexa levels. The 14% catalyst level resulted in failure or excessive cracking in many of the plates. Little or no difference was noted between the 3 and 6% catalyst levels, and therefore, a 6% catalyst level was standardized on for the remainder of the program.

(c) Effect of Binder Level

The effects of binder level and filler composition upon the phosphoric acid screening tests are shown in Table II of the Seventh Monthly Progress report and a reproducibility study is Table III in the Seventh Monthly Progress report.

Final Results Task 2

The final results for Task 2 are tabulated in Tables I and II of the Eleventh Monthly Progress report. This data indicates that two competing phenomona must be balanced against each other to make the most durable bipolar plate. The amount of resin composition should be minimized so as to obtain the maximum electrical conductivity by the graphite filler, but in minimizing the amount of resin binder the hydrogen permeability and the flex strength of the coupon are adversely affected. The Ashland research program concluded that a binder level of 20-25% and a catalyst level of 6% was optimum for the resin systems and graphite samples utilized in the study.

3. Improved molding technique

An improved technique for molding composite samples and post-curing the 1" x 3" x 1/8" coupons were developed and implemented approximately half-way through the program. The two-step molding process allows gases to escape from the mold between the first and second compression step and favors complete filling of all void volume of the mold.

Carbon comprises three different particle sizes, Asbury 4333 (large), Asbury A-60 (medium) and Micro 840 (small). The binder is a phenolic-formaldehyde resin with a range of 20% to 33% level.

Carbon and resin composite is mixed in a Moulinex coffee grinder and blended with a Hobart mixer to give a homogeneous mixture. Twelve grams of the composite is placed in the cavity of the compression mold after the mold has been preheated to a range of 150° to 200°F. Sample is spread evenly in the cavity by means of a spatula. The preform bar (1/4") is inserted into the cavity and pressure applied by means of a heated plate carver press until the bar is fully extended. Pressure is maintained on the mold. The outside mold temperature is measured by means of a magnetic thermometer. When the mold temperature reaches 250°F the pressure is released and the preform bar is removed. This allows the free ammonia or other gaseous products to escape and virtually eliminating the possibility of trapped gases. It is theorized that a breakdown in the hexa occurs forming the gaseous products. In addition to alleviating trapped gases, this preliminary step serves to mold a more uniform sample since in the temperature range of 250°F the sample demonstrates a relative amount of plasticity and can be easily converted to 1/8" diameter.

The second step entails inserting the bar used for forming 1/8" width coupons into the cavity. The mold is placed between the heated plates of the carver press (Plate temperatures in the range of 380°-390°F) and pressure applied until the bar is fully extended with pressure being maintained. The sample is allowed to remain under these conditions for a period of 20 minutes. Sample is removed and placed on an asbestos pad and allowed to cool. Sample is then post cured at 370° to 380°F for a period of 20 hours.

The preliminary step as described earlier serves to eliminate any cracking that might occur from post curing of the samples. Earlier samples in which the preliminary step was omitted random cracking occurred. This cracking was observed to have no trend as to the amount of hexa used.

Before another sample is started, the mold is allowed to cool to the range of 150° to 200°F.

4. Radiotracer studies

As part of Task 2, an Arofene carbon-14 labelled resin was prepared and fabricated into coupons as described in the Ninth Monthly Progress report. The complete experimental description and results are included in the monthly report. The carbon-14 tracer analysis revealed that phosphoric acid extracts a significant quantity of material from the resin that could affect the performance of the bipolar plates. The carbon-14 testing technique is extremely sensitive for analyzing migration of trace impurities from the bipolar plates into the fuel cell, and this technique may prove to be the most valuable for determination of the aging mechanism of the cell and thus improving its lifetime.

C. Task 3: Screening of Carbons and Determination of Practical Ranges of Carbon Loadings to Select Those Carbon Compositions and Loadings Giving the Highest Electrical Conductivity

The detailed experimental results and conclusions are described in the attached monthly progress reports. A new device for measuring volume resistivity was designed and used for this program.

Particle packing theory was used to maximize composite density and decrease the quantity of resin needed to bind the graphite in order to provide the highest electrical conductivity. Graphite of different particle sizes permits the smaller particles to fill the voids between the larger particles. A binder level of about 20-25% was found to be optimum for the particular graphite blend selected. Electrical resistivity was found to increase as the binder level increased; but at low binder levels (below about 20%), permeability to hydrogen was significant.

The program initially molded coupons to a constant thickness with a constant weight of resin/graphite pre-mix. After 7 months into the program, an improved procedure (molding at a constant pressure) was developed and resulted in more uniform coupons and more reproducible results. It was also discovered that substitution of Asbury 7101 graphite for Asbury A-60 in the same proportion gave lower volume resistivity.

In order to accomplish the new molding procedure at constant pressure, additional resin/graphite mixture was added to the mold cavity (13 grams vs. 12 grams) and the pressure during the molding operation was decreased to ensure that the mold would not completely close. It was noted that the coupons molded to constant thickness (old technique) actually varied in thickness due to the shrinkage of resin. Tests were performed at 2000, 3000, and 4000 psig for 12 and 13 grams of mixture to determine the best conditions for molding test specimens. By measuring the thickness of the molded coupons, it was determined that 13 grams of composite afforded sufficient coupon thickness to prevent closing of the mold to the stop. It was noted that coupons molded at 2000 psig exhibited hydrogen permeability at 10 psig differential hydrogen pressure while those molded at 3000 and 4000 psig did not. Hence, 4,000 was chosen for the program of optimum composite selection.

Table II, page 5 of the Eighth report documents the significant effect of graphite composition on density and volume resistivity of the composites.

The principal observation is that the flex breaking strength is lower on the phosphoric acid aged samples than the fresh samples. There were significant differences in the degree of loss of flex strength; and based upon these results, formulations were chosen for Phase 4 testing.

D. Task 4: Composite Evaluation to Determine the Effect of Resin and Carbon Loading for the Resin System Selected from Task 2 & 3.

Eighty coupons representing 10 compositions were prepared for Task 4 aging studies. One half of the coupons were placed in the container of phosphoric acid for the 90-day aging test at 190° ± 5°C and one quarter were stored for later determination of flex strength. The density and volume resistivity of the freshly prepared coupons was determined. Hydrogen permeability tests were completed on selected coupons from each group. Data for the density, volume resistivity and hydrogen permeability are shown in Table 3 of the Tenth Monthly Progress report.

The samples were evaluated for weight change and physical appearance after 1558 hours immersion in phosphoric acid, Table I.

Especially notable was the performance of Ashland A-33 resin compounded with 75% of the filler comprising 50% Asbury 7101, 40% Asbury 4015 and 10% Asbury Micro-250 graphites.

The flexural strength of the coupons, Table II, demonstrates the significant deteriorating effect of phosphoric acid on the resin composition.

TABLE 1

Coupon Number	COMPOSITION Binder	% Hexa	% Binder	Filler	Total Hours Aged	Weight Change %	Final Rating	Physical Appearance Description
	Arofene							
88^2	872	6	25	A	1558	+2.58	3	Good
88^3	872	6	25	A	1558	+1.19	3	Good
88^4	872	6	25	A	1558	+4.92	3	Good
88^5	872	6	25	A	1558	+4.10	3	Good
89^1	877	6	25	A	1558	+2.97	3	Good
89^2	877	6	25	A	1558	+2.07	3	Good
89^3	877	6	25	A	1558	+5.46	3	Good
89^4	877	6	25	A	1558	+4.22	3	Good
90^1	890	6	25	A	1558	+1.13	3	Good
90^2	890	6	25	A	1558	+1.19	3	Good
90^3	890	6	25	A	1558	+0.76	3	Good
90^4	890	6	25	A	1558	+1.30	3	Good
91^1	A-33	6	25	A	1558	-0.28	4	Very Good
91^2	A-33	6	25	A	1558	-0.41	4	Very Good
91^3	A-33	6	25	A	1558	-0.08	4	Very Good
91^4	A-33	6	25	A	1558	+0.53	4	Very Good
92^3	Colloid 8440	-	25		1558	+2.99	2.5	Good
92^4	8440	-	25	-	1558	+9.87	2	Pitted-warped
92^5	8440	-	25	-	1558	+5.71	2	Porous-warped
92^8	8440	-	25	-	1558	+3.92	2	Porous-warped
93^1	Commercial Control #1	-	-	-	1558	+8.93	1.5	Decayed edges
93^2		-	-	-	1558	+6.80	1.5	Warped
93^3		-	-	-	1558	+6.32	1.5	Slightly pitted
93^6		-	-	-	1558	+4.99	1.5	Slightly pitted
94^1	Colloid 8440	-	33	A	1558	+2.95	1	One corner flaking
94^2	8440	-	33	A	1558	+4.71	1	Pitted-warped
94^3	8440	-	33	A	1558	+4.60	1.5	Some pitting
94^4	8440	-	33	A	1558	+3.51	1.5	Some pitting
95^1	Commercial Control #2	-	-	-	1558	+1.77	2.5	Fair to good
95^2		-	-	-	1558	+13.12	0	Terrible-flaked
95^3		-	-	-	1558	+4.67	1	Warped
95^4		-	-		1558	+0.51	2.5	Fair to good
96^1	Colloid 8440	-	25	B	1558	+16.02	0	Warped-flaked
96^2	8440	-	25	B	1558	+16.84	0	Side chipped
96^3	8440	-	25	B	1558	+25.05	0	Side chipped
96^4	8440	-	25	B	1558	+12.07	0	Warped-chipped
97^1	Colloid 8440	-	33	B	1558	+3.13	3	Good
97^2	8440	-	33	B	1558	+5.60	2	Slight warp-pitting
97^3	8440	-	33	B	1558	+3.58	2.5	Good
97^5	8440	-	33	B	1558	+2.07	3	

TABLE II

FLEX STRENGTH OF TEST COUPONS
ASTM-D790

	Sample #	Aged in H_3PO_4	Flex Strength	Aveg.	Std. deviation
Arofene 872	88-2	1558 hrs.	4901 psi		
	88-3	1558 hrs.	4766 "	4133 psi	aged 868 psi
	88-4	1558 hrs.	3052 "		
	88-5	1558 hrs.	3813 "		
	88-1	0	4446 "		
	88-6	0	3549 "	3636 psi	fresh 681 psi
	88-7	0	3757 "		
	88-8	0	2791 "		
Arofene 877	89-1	1558 hrs.	4456 psi		
	89-2	1558 hrs.	4773 "	4149 psi	aged 967 psi
	89-3	1558 hrs.	2712 "		
	89-4	1558 hrs.	4656 "		
	89-5	0	3284 "		
	89-6	0	4642 "	4139 psi	fresh 744 psi
	89-7	0	(6354)		
	89-8	0	4492 "		
Arofene 890	90-1	1558 hrs.	4966 psi		
	90-2	1558 hrs.	7797 "	6665 psi	aged 1258 psi
	90-3	1558 hrs.	7410 "		
	90-4	1558 hrs.	6486 "		
	90-5	0	7596 "		
	90-6	0	6683 "	7026 psi	fresh 889 psi
	90-7	0	7888 "		
	90-8	0	5935 "		
A-33	91-1	1558 hrs.	7351 psi		
	91-2	1558 hrs.	6507 "	6422 psi	aged 715 psi
	91-3	1558 hrs.	5041 "		
	91-4	1558 hrs.	6188 "		
	91-5	0	6704 "		
	91-6	0	6341 "	6310 psi	fresh 940 psi
	91-7	0	7195 "		
	91-8	0	5001 "		
Colloid 8440	92-3	1558 hrs.	4379 psi		
	92-4	1558 hrs.	(1214) "		
	92-5	1558 hrs.	6168 "	5486 psi	aged 967 psi
	92-8	1558 hrs.	5912 "		
	92-1	0	2335 "		
	92-2	0	3922 "	3232 psi	fresh 849 psi
	92-6	0	3071 "		
	92-7	0	3599 "		

	Sample #	Aged in H_3PO_4	Flex Strength	Aveg.	Std. deviation
Commercial Control #1	93-1	1558 hrs.	2912 psi	3418 psi	aged 918 psi
	93-2	1558 hrs.	3045 "		
	93-3	1558 hrs.	4792 "		
	93-6	1558 hrs.	2923 "		
	93-4	0	6800 "	4371 psi	fresh 1810 psi
	93-5	0	4577 "		
	93-7	0	3511 "		
	93-8	0	2596 "		
Colloid 8440	94-1	1558 hrs.	6472 psi	4395 psi	aged 2500 psi
	94-2	1558 hrs.	2517 "		
	94-3	1558 hrs.	1961 "		
	94-4	1558 hrs.	6629 "		
	94-5	0	2745 "	3800 psi	fresh 1959 psi
	94-6	0	2028 "		
	94-7	0	6491 "		
	94-8	0	3935 "		
Commercial Control #2	95-1	1558 hrs.	7770 psi	8223 psi	aged 998 psi
	95-2	1558 hrs.	7680 "		
	95-3	1558 hrs.	7723 "		
	95-4	1558 hrs.	9719 "		
	95-5	0	3942 "	4800 psi	fresh 2180 psi
	95-6	0	4853 "		
	95-7	0	2823 "		
	95-8	0	7903 "		
Colloid 8440 25% binder	96-1	1558 hrs.	811 psi	875 psi	aged 613 psi
	96-2	1558 hrs.	745 "		
	96-3	1558 hrs.	234 "		
	96-4	1558 hrs.	1709 "		
	96-5	0	3887 "	3319 psi	fresh 644 psi
	96-6	0	2416 "		
	96-7	0	3650 "		
	96-8	0	3323 "		
Colloid 8440 33% binder	97-1	1558 hrs.	6549 psi	(4886)psi	aged 1726 psi
	97-2	1558 hrs.	(1751) "		
	97-3	1558 hrs.	3103 "		
	97-5	1558 hrs.	5006 "		
	97-4	0	5015 "	5702 psi	fresh 2411 psi
	97-6	0	No sample		
	97-7	0	8383 "		
	97-8	0	3710 "		

PHOSPHORIC ACID ENDURANCE TEST

The 1" x 3" bipolar composition coupons after post curing are prepared for aging in 200°C H_3PO_4.

Six coupons were aged in each pint jar. A small hole is drilled approximately 1/4" from the end of the coupons and a teflon string is run through the hole of each of the six coupons. A 1/4" teflon spacer is used to separate the coupons, preventing the samples from touching each other. A small vent hole is drilled in the pint jar lid and the teflon string containing the samples is strung through the hole to suspend the samples. The cardboard sealer from the pint jar is removed and replaced with a teflon gasket to prevent corrosion of the lid. The pint jar containing the above described samples is filled to within approximately 1" of the top of the jar with Bakers reagent grade 85% H_3PO_4. Samples are placed in an oven with a pre-set temperature of 200°C with a catch tray placed beneath the samples. The samples are allowed to age for a period of 7 to 10 days before the H_3PO_4 was changed with each weighing.

When the samples are removed from the pint jars, in many instances the sample is coated with a film. This film is removed by holding the sample under a warm stream of running water and wiping the film with ones fingers. After the washing process, the samples are placed on an asbestos pad and dried at 150°C for a period of one hour. The samples are visually inspected and recorded as to their condition. The cycle is then repeated.

HYDROGEN IMPERVIOUSNESS

This apparatus was constructed to check the effect of binder levels of hydrogen permeability. A metal jig was fabricated to 1" x 3" dimensions so that the bipolar plate coupon compositions would fit snug in the cavity. Cured rubber gaskets were cut to fit the outer perimeters of the cavity. The coupon was placed between the rubber gaskets. The top plate contains six holes for inserting bolts and an extended plate 3/16" in thickness that fits into the cavity. The bolts connect the top plate with the bottom chamber and as the bolts are tightened, pressure is created thereby causing the rubber gaskets to expand and form an air-tight seal.

Hydrogen was introduced through the top plate with a vent in the bottom chamber. A piece of tygon tubing was attached to the vent and extended into a beaker of water. A visual observation was noted of the permeability of the coupon by a bubble count.

CALCULATION OF COMPRESSED DENSITY

Various natural and synthetic graphite blends were compression tested to maximize the density of the mixtures by using particle packing techniques. Particle packing is the technique wherein different sized carbon particles are mixed in order to use the small particles to fill the voids between the larger particles. This results in a higher density than can be achieved with either the small particles or large particles by themselves. The filling of voids of the carbon particles should serve to impart better electrical conductivity. Cabot's XC-72 was included in this test because of its high surface area and structure (surface area 254, structure fluffy 185). Sterling MT was also included because of its physical characteristics (surface area 7, structure fluffy 34). The low structure would enhance closer packing which should aid the conductivity if the particles are in contact with each other.

Densities were determined at several pressures up to approximately 11,000 psi. The composites were blended by means of a Hobart mixer. A weighed quantity of the composite (20 grams) was placed in a cylinder (1.5105 inches in diameter). A perforated mild steel block was inserted in the bottom of the cylinder and a microporous stainless steel pad placed on the block. The same set-up was used at the top of the cylinder with the composite in between. This served to alleviate trapped air, but maintain the carbon particles from escaping. A close tolerance steel rod was inserted into the cavity. The apparatus was placed between the plates of a carver press. A base dial micrometer was placed on a 1/2" steel plate and a reading was taken before any pressure was applied. Force was measured at different guage readings from 1,000 to 20,000 and then converted to the true psi.

Vsl Calculation:

Micrometer Reading		Plate Thickness		No Sample Reading		Sample Thickness
A	+	B	-	C	=	D

$D \times 2.54 \times 11.561 = 29.365$

$D \times 29.365 = E \rightarrow$ volume cc

$\frac{\text{Sample Wt.}}{E} = F \rightarrow$ Density gram/cc

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September 9, 1977

To: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, Virginia 22060

From: P. M. Colling

Subject: Monthly Letter Progress Report - 8 August 1977 through 7 September 1977, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

INTRODUCTION AND OBJECTIVES

The purpose of this contract is to screen, select and evaluate carbon resin composites for hydrogen fuel cell bipolar plates to be used in the presence of phosphoric acid for up to 6 months at 150°C to 200°C. The plates must be electrically conductive, impervious to hydrogen and possess certain minimum mechanical properties, including strength and impact resistance. They must maintain these properties under operating conditions for several months. The research program is divided into five tasks. The first three tasks are screening of resins and carbons by various methods. Task 4 will be a more detailed evaluation of composites prepared from the more promising resins and fillers selected during the first three tasks. The last task is the final report preparation. The program is currently in the middle of Task 1. Tasks 2 and 3 have not yet started.

RESULTS AND OBSERVATIONS

A literature search has been started to help determine resin and fillers which would have the greatest potential for this application. Discussions have also been held with Ashland personnel in other research groups concerning resins which are most likely to provide the properties needed. A list of resins to be screened is being formulated now. A portion of this list is shown below. Product data sheets and samples of fillers such as graphite and carbon black are being obtained. Laboratory equipment for compounding and corrosion studies has been set up.

The principal classes of resins which are being considered for experimental screening at this time are phenol formaldehyde, furfuryl alcohol, and teflon. Data on other resins will also be examined and might include the polyesters, polysulfones, polycarbonates and polyamides.

The following novolak resins comprise a wide spectrum that will be evaluated at 6%, 10%, and 14% hexamethylene tetramine (hexa) levels.

Resin Designation	Modifier	Dennis Bar Softening Point °C	Relative Flow
Arofene 872	0	95 - 110	Low
Arofene 875	Aniline	90 - 100	Medium
Arofene 877	0	80 - 95	Medium
Arofene 877LF	0	75 - 85	Medium to High
Arofene 890LF	0	85 - 95	Low
Arofene 2869	Polyvinyl Butyral	93 - 104	Low
Arofene 860	Epoxy	96 - 102	Low to Medium

In addition to the above standard resins, the experimental products below will also be included in the program:

EP-032217-A-33 (Styrene Modified 890)

EP-032217-A-39 (Methoxy functional diphenyloxide Modified 890)

EP-032217-A-35 (High Aniline 875)

EP-032217-A-36 (875 with polyvinyl butyral)

DISCUSSION

The composites for this project must be capable of withstanding hot phosphoric acid at from 150°C to 200°C and must at the same time possess a relatively high level of electrical conductivity and very low permeability to hydrogen. They must also possess a certain amount of mechanical strength. Electrical conductivity would be expected to increase as the loading of the carbon filler increases, but other properties, particularly the strength and permeability, would deteriorate. So a compromise of properties is necessary.

The effect of varying amounts of "hexa" which is a curing agent for the novolak resins, is to provide improved heat resistance and higher viscosities as the "hexa" content increases and improved chemical resistance at low levels of "hexa". A series of novolak resins with different levels of "hexa" will be studied to optimize the "hexa" level.

The effect of mixing during the compounding step is another important variable that must be quantified. One suggestion is to use a low boiling solvent to partially dissolve the solid resins to obtain better and more intimate contact between the resin and the filler particles. This "premix" is to be partially dried and then reground to provide a more uniform composite. The effect of such a technique shall be determined.

Graphites have been used for fillers in these types of materials and are well known for their good electrical conductivity properties. Sometimes, mixtures of two or more particle size ranges are helpful in obtaining high density composites and good conductivity. Similarly, it might be expected that by mixing in certain types of carbon black the conductive properties of the composite could be improved. This will also be studied.

Many of the furfuryl alcohol resins which are to be studied are liquid systems and this will involve some process development to determine effective compounding methods.

Teflon will be evaluated because of its known temperature and chemical resistance. However, it is also known that molding of teflon parts, particularly small intricate parts is extremely difficult. If the basic data provided by this research contract shows significant advantages in using teflon composites, then there would be justification for additional process development in the area of molding of the teflon carbon composites.

FUTURE PLANS

Samples of the novolak resins will be submitted to Ashland Chemical's Analytical Chemistry Section for thermal analysis during the next reporting period. The literature search will also continue in more detail and by the end of the next reporting period, Task 1 should be complete with a complete listing of proposed resins and fillers to be screened in Tasks 2 and 3. It is expected that Tasks 2 and 3 will actually begin somewhat prior to the end of Task 1 in order to help speed up the overall project. This is necessary in order to complete the project work within the 11 months time frame as specified in the revised project statement of work.

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October 19, 1977

TO: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, Virginia 22060

FROM: F. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 September 1977 through 7 October 1977, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

Task 1 is about 90% complete, and Tasks 2 and 3 have been started.

TASK 1

The literature search is essentially complete and a list of resins and resin types to screen in Tasks 2 and 3 has been compiled. Thermal analyses of novolak resins has begun.

TASK 2

Corrosion test facilities have been set up. These are two constant temperature ovens operated at 150°C and 200°C with test specimens to be contained in bottles inside each oven.

TASK 3

Compounding experiments have begun to determine optimum loadings for various carbon fillers. A compression mold has been ordered for this purpose.

RESULTS AND DISCUSSION

TASK 1

The literature search is essentially complete and most articles and patents which were ordered have been received and reviewed. The list of resins and resin types has not changed significantly from those

suggested last month. It includes phenolformaldehyde novolak resins, furan resins, teflon, polyesters, polysulfones, polycarbonates, polyamides, with most emphasis being placed on the novolaks at the outset due to their relative moldibility and superior performance characteristics.

Various natural and synthetic graphites and blends of different graphites will be tested to try to achieve the desired balance of properties of the molded part. Discussions were held with Asbury Graphite Mills and they recommended several graphites including the following Asbury products: Numbers 4333, 4110, A-60, 289-A, and 5131. These graphites possess a wide range of properties and particle sizes. Asbury personnel have given us many helpful suggestions and a meeting is scheduled for October 18th with Asbury to discuss graphites for this application. Certain carbon blacks, particularly those having high surface area and structure such as experimental furnace blacks and acetylene black will be tried as additives to the graphites to impart improved electrical conductivity. Carbon fibers will also be studied.

Thermal gravimetric analyses of the novolak resins has started. Arofene 877 was tested at 3 hexa levels with no significant difference apparent. The remainder of the resins will be tested at one hexa level only.

TASK 2

Two constant temperature laboratory ovens have been set up to conduct the phosphoric acid corrosion tests. These will be done by placing the test piece in phosphoric acid contained in bottles in each of the ovens. The test pieces will be compression molded cured strips measuring approximately 1/8" thick by 1" wide by 3" long (1/8" x 1" x 3").

TASK 3

Preliminary formulations of compounds and curing is underway to determine optimum filler loadings and cure times. A composite sample was received from a commercial supplier of bipolar plates to use as a control in the experimental work. A Carver press with heated plates and a preform mold was obtained and set up for preforming and curing test pieces. Based on the preliminary work done drawings were made for a new compression mold capable of forming a 1" x 3" test strip. These strips can be used for both the corrosion tests and flex tests. A compression mold is being fabricated by a local machine shop.

FUTURE PLANS

The new 1" x 3" compression mold is being constructed and is expected to be received during October. At that time fabrication of the corrosion test strips for Task 2 began using the new compression mold. Task 3 which has already begun with the preliminary compounding work will continue to screen and select suitable carbons and to determine optimum loading range.

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November 17, 1977

TO: Dr. Alayne A. Adams
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Electrical Power Laboratory
Fort Belvoir, Virginia 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 October 1977 through 7 November 1977, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 1

The thermal analysis of the novolak resins has been performed with no large differences up to 400°C being observed.

TASK 2

The compression mold has been received so fabrication of test strips can now start for corrosion testing.

TASK 3

Most of the effort during the past month has been centered on the determination of compressed densities of various mixtures of graphites of different particle sizes. This is being done to select graphite compositions giving the highest densities. Compounding experiments to determine preferred binder level ranges have started using the newly received compression mold.

RESULTS AND DISCUSSION

TASK 1

The thermal analysis work has been completed on the novolak resins which were submitted. In general, only minor changes in weight loss were noted between 200°C and 400°C. Some differences were noted above 400°C, but it

is difficult to predict what relationship, if any, these changes will have to performance in phosphoric acid at 200°C. Therefore, all of the novolak resins will be screened by the phosphoric acid method in Task 2 and none will be eliminated on the basis of the thermal analysis.

TASK 2

Very little work was done on Task 2 because the compression mold was received very near the end of the reporting period. However, work has started and a few test strips have been molded using the new compression mold. These test strips are 1" by 3" long and can be either 1/8" or 1/4" thick.

TASK 3

A meeting was held with personnel from Asbury Graphite Mills on October 18th. As a result of this meeting, the major emphasis during the reporting period has centered around trying to maximize the density of graphite mixtures by using particle packing techniques. Particle packing is the technique wherein different sized graphite particles are mixed in order to use the small particles to fill the voids between the larger particles. This results in a higher density than can be achieved with either the small particles or large particles by themselves. Densities were determined at several pressures up to approximately 11,000 psi. A weighed quantity of the graphite mixture was placed in a cylinder and compressed to a measured pressure. The sample thickness was then measured, the pressure was released and the sample thickness was remeasured to determine how much expansion occurred in the graphite. Table 1 shows the results of these experiments. Additional experiments will be performed to find graphite blends giving higher densities and to check repeatibility of the method.

FUTURE PLANS

TASK 2

During the next reporting period, a three component graphite composition will be selected to use for phosphoric acid screening tests. This composite will not necessarily be the composite giving the maximum density, but it will be one that is close to it. A suitable range of binder level will then be determined for this composition. One inch by three inch corrosion test strips will be prepared using all of the novolak resins selected, and using the same graphite composite and binder level. Test strips will also be prepared from the composite which was received from a commercial supplier as a control. The phosphoric acid screening test will start by emersing these test strips in vessels containing phosphoric acid at 150°C and 200°C.

TASK 3

We will continue to try to maximize the density of three component graphite and graphite/carbon black composites. Then practical ranges for binder level will be determined for this composition by molding 1" x 3" strips and determining density, electrical conductivity, and permeability to hydrogen.

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TABLE 1

COMPRESSED DENSITIES OF GRAPHITE/CARBON BLACK COMPOSITES Grams/cc

	Pressure Expanded	P	E	P	E	P	E	P	E	P	E
Composition: 4333	100%	--		--		--		--			
A-60	--	100%		100%		--		--			
Micro 840	--	--		--		100%		--			
MT	--	--		--		--		100%			
Pressure: PSI-558	1.37 --	1.22	1.18	1.37	1.35	1.22	1.21	1.08	--		
1117	1.37 --	1.28	1.22	1.37	1.36	1.24	1.18	1.08	1.07		
2233	1.43 --	1.50	1.37	1.52	1.53	1.43	1.35	1.15	1.12		
3350	1.56 --	1.64	1.52	1.67	1.53	1.60	1.36	1.20	1.15		
4467	1.65 --	1.73	1.57	1.73	1.58	1.70	1.53	1.23	1.17		
5583	1.72 --	1.80	1.60	1.82	1.63	1.77	1.58	1.25	1.18		
6700	1.79 --	1.85	1.60	1.87	1.64	1.84	1.61	1.28	1.19		
7817	1.83 --	1.89	1.66	1.91	1.66	1.90	1.62	1.30	1.20		
8934	1.88 --	1.93	1.66	1.95	1.67	1.95	1.63	1.31	1.21		
10050	1.92 --	1.96	1.66	1.98	1.68	1.98	1.68	1.33	1.21		
11167	1.96 --	1.99	1.71	2.00	1.71	2.02	1.67	1.34	1.21		
Composition: 4333	80%	80%		60%		50%		20%		30%	
A-60	15%	15%		30%		35%		60%		20%	
Micro 840	5%	5%		10%		15%		20%		50%	
MT	--	--		--		--		--		--	
Pressure: PSI-558	1.40 --	1.41	--	1.41	1.40	1.39	1.37	1.45	--	1.35	--
1117	1.43 1.40	1.41	1.39	1.43	1.40	1.43	1.42	1.46	1.43	1.36	1.34
2233	1.59 1.54	1.55	1.50	1.59	1.51	1.61	1.53	1.59	1.52	1.51	1.47
3350	1.69 1.61	1.63	1.55	1.70	1.60	1.72	1.61	1.72	1.60	1.62	1.57
4467	1.78 1.63	1.68	1.57	1.78	1.62	1.78	1.63	1.81	1.63	1.70	1.61
5583	1.85 1.67	1.72	1.59	1.82	1.64	1.82	1.68	1.87	1.67	1.72	1.64
6700	1.91 1.71	1.76	1.60	1.87	1.68	1.89	1.70	1.92	1.70	1.76	1.66
7817	1.96 1.71	1.79	1.64	1.91	1.68	1.94	1.71	1.96	1.72	1.79	1.70
8934	2.00 1.74	1.82	1.64	1.94	1.70	1.98	1.73	2.00	1.73	1.80	1.71
10050	2.03 1.75	1.84	1.64	1.97	1.72	2.01	1.75	2.02	1.75	1.82	1.72
11167	2.06 1.77	1.86	1.64	1.99	1.73	2.04	1.77	2.06	1.77	1.83	1.74
Composition: 4333	80%	60%		50%		20%		30%			
A-60	15%	30%		35%		60%		20%			
Micro 840	--	--		--		--		--			
MT	5%	10%		15%		20%		50%			
Pressure: PSI-558	1.50 --	1.38	--	1.47	--	1.43	--	1.27	--		
1117	1.50 1.48	1.43	1.41	1.48	1.46	1.43	1.41	1.27	--		
2233	1.53 1.53	1.58	1.52	1.59	1.53	1.59	1.52	1.39	1.26		
3350	1.61 1.52	1.67	1.59	1.67	1.58	1.67	1.57	1.45	1.35		
4467	1.68 1.55	1.72	1.60	1.73	1.61	1.73	1.60	1.48	1.39		
5583	1.72 1.56	1.77	1.62	1.77	1.63	1.78	1.63	1.50	1.41		
6700	1.77 1.57	1.81	1.64	1.81	1.64	1.82	1.63	1.52	1.42		
7817	1.80 1.59	1.84	1.64	1.84	1.64	1.86	1.66	1.54	1.43		
8934	1.83 1.61	1.86	1.64	1.86	1.66	1.88	1.66	1.55	1.44		
10050	1.86 1.62	1.89	1.64	1.88	1.65	1.89	1.56	1.56	1.44		
11167	1.88 1.62	1.90	1.65	1.89	1.67	1.91	1.66	1.58	1.45		

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December 14, 1977

TO: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, Virginia 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 November 1977 through 7 December 1977, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 2

Phosphoric acid screening tests have begun. Weight loss data for three different binder levels were determined.

TASK 3

The electrical conductivity apparatus and a hydrogen permeability apparatus were set up and checked out. A method for the determination of conductivity was developed.

RESULTS AND DISCUSSION

TASK 2

Phosphoric acid screening tests of bipolar plate compositions have begun. Three samples at three different binder levels were aged in phosphoric acid at 200°C using a commercial supplier composite as a control. Initial results indicated a relatively large weight loss for the commercial supplier, but a repeat indicated that a weighing error occurred on weighing the original test strip. The carbon filler was composed of 50% Asbury 4333 graphite, 45% Asbury A-60, and 5% Asbury Micro 840. The binder was a laboratory prepared Arofene 877 novolak containing 9% hexa.

The weight loss percentages are given in the following table.

SAMPLE	CONTROL 1	CONTROL 2	50/45/5	50/45/5	50/45/5
% Binder	33	33	15	20	30
Hours emersed - Percent weight loss (gain) @ 200°C					
92		1.2			
120	8.98*		(0.64)	0.818	1.10
216	9.05*		(1.08)	0.88	1.10
288	9.03*		(1.34)	0.89	1.05
336	8.89*		(1.64)	0.89	1.00
428	8.90*		(1.48)	0.94	1.06

*Weighing error suspected in original weight.

The aging test is continuing to accumulate longer term data on the second control. Additional test strips will be fabricated using different binders during the next month.

TASK 3

An apparatus has been set up and checked out to measure electrical conductivity of the test pieces. Electrical conductivity tests will be started soon.

An apparatus was constructed to measure the hydrogen permeability of the test strips. A checkout test of the apparatus indicated zero measureable flow of hydrogen through a 1" x 3" x 1/8" thick sample containing 30% binder.

FUTURE PLANS

TASK 2

Screening of the novolak resins will begin using the phosphoric acid test at 200°C. The criteria for stability will be initially weight and dimensional stability. Those resins, showing excessive weight loss or dimensional instability will be eliminated from further consideration. Those showing relatively small changes in weight on dimensions will be further screened using flex strength.

TASK 3

The effect of binder levels on hydrogen permeability will be determined this month. Also, the effect of graphite composition and binder level upon electrical conductivity will be determined.

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January 19, 1978

TO: Dr. Alayne A. Adams
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Fort Belvoir, Virginia 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 December 1977 through 7 January 1978, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 1

The thermal gravimetric analysis data which was obtained during the second month of the effort is presented this month.

TASK 2

Twelve novolak resins are being aged in hot phosphoric acid at 200°C. These represent different resins at varying hexa levels from 6 to 14%. Two compositions have failed. Both contained 14% hexa. Additional resins will be tested during January.

TASK 3

Two coupons were tested for hydrogen permeability. No hydrogen flowed through a coupon containing 25% binder at 40 psig, but a significant flow occurred at 15% binder.

RESULTS AND DISCUSSION

TASK 1

The objective of Task 1 was preliminary screening of resins and carbons to select specific ones for further tests.

Although Task 1 was completed during the third month of the program, the thermal gravimetric analysis data has not been reported until now. Since the TGA results showed only subtle differences between the resins tested, none was eliminated from further testing as a result of the TGA data. The TGA data is summarized in Table 1.

TASK 2

The objective of Task 2 is to screen resins on the basis of their resistance to phosphoric acid at 200°C. Physical deterioration is being determined by observing measured changes in size, weight and flex strength. Those resins showing relatively low rates of deterioration will be considered for further testing.

Table 2 shows the results of the phosphoric acid screening tests which have been obtained to date. Some compositions lose weight and some gain weight. Both results are an indication of some sort of deterioration, although the mechanism is not understood at this time. Most coupons contained 33% binder because that was the amount contained in the control sample. This may not be the optimum binder level. The one coupon at 15% binder (Arofene 877) gained weight, while the coupons of 877 at higher levels lost weight. The increase in weight may be due to occulsion of water or acid in a relatively porous matrix.

Arofene 877 and possibly, Arofene 860 (at 10% hexa) appear to have the lowest weight changes.

The tests are continuing and test of additional resins will be started in January.

TASK 3

The objective of Task 3 is to screen carbon fillers and determine the particle range of carbon loading within which to work. This will be determined by measuring electrical conductivity and hydrogen permeability at several levels of binder for a few different carbon fillers.

Two coupons were tested for hydrogen permeability. The binder in both cases was Arofene 877 with 10% hexa and the filler was composed of 50% Asbury 4333, 45% Asbury A-60, and 5% Asbury Micro 840. At 15% binder hydrogen flow was observed at 40 psig hydrogen pressure. The flow rate was not determined. At 25% binder the flow rate was 0 at 40 psig.

Coupons are being prepared with binder levels of from 15 to 33% to be tested during January. Electrical conductivity will also be determined for these coupons.

FUTURE PLANS

TASK 2

Phosphoric acid endurance tests will continue on the compositions presently being tested and tests on the remaining novolak resins that were listed in Table 1 will be started. Twelve coupons of Arofene 877, the most promising resin to date, will be prepared to determine the repeatibility of the various tests, i.e., phosphoric acid endurance, electrical conductivity

and hydrogen permeability. Endurance testing will also be repeated for the two compositions that failed. Phosphoric acid endurance tests will be started on coupons with different levels of binder to determine the effect of binder level.

TASK 3

Electrical conductivity and hydrogen permeability measurements will be made on two series of compositions at different binder levels ranging from 15 to 33%. Repeatability tests will also be performed.

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TABLE I

THERMOGRAVIMETRIC ANALYSES

Sample Description	Observed Curing Temperature, °C	Wt. Loss During Cure	Temperature @ 10% loss* °C	Temperature @ 50% loss* °C	Comments
I. Standard Products					
877 6% hexa	170°	5%	460°	595°	
877 6% hexa	160°	2-1/2%	445°	610°	± 10°C repeatability
877 10% hexa	165°	4%	455°	555°	
877 14% hexa	165°	6%	445°	555°	
877 14% hexa	160°	5%	415°	575°	
877LF 10% hexa	160°	4%	470°	580°	
872 10% hexa	--	2%	470°	--	decomposed @ 540° after only 20% wt. loss
860 10% hexa	150°	6%	420°	580°	inflection @ 415°; rapid decomposition @ 600°
2869 10% hexa	160°	5%	405°	570°	inflection @ 400°; rapid decomposition @ 600°
875 10% hexa	160°	3%	450°	615°	
II. Experimentals					
A-38	140°	5-1/2%	455°	595°	
A-35	140°	3%	445°	610°	
A-36	150°	4%	420°	590°	inflection @ 435°
A-33	160°	3%	420°	--	rapid decomposition @ 580° after only 46% wt. loss
A-37	--	2%	500°	--	rapid decomposition @ 510° after only 13% wt. loss

*300°C is 100%

TABLE II

Resin Designation	% Hexa	% Binder	Hours Aged @ 200°C	% weight change *
Arofene 875	6	33	155	- 0.095
	10	33	538	+ 1.80
	14	33	155	- 0.57
Arofene 877	7	33	155	- 0.68
	10	33	538	- 0.67
	13	33	155	- 1.71
	10	15	1097	+ 3.04
	10	20	1097	- 0.93
Arofene 860	10	33	538	- 0.29
	14	33	63 (failed)	
Arofene 2869	6	33	155	+ 0.96
	10	33	538	+ 5.00
	14	33	63 (failed)	
A-33	10	33	538	- 0.87
Commercial Control		33	761	- 1.04
		33	538	- 1.18
		33	155	- 0.56
		33	155	0.00

* - indicates a loss

+ indicates a gain

Ashland Chemical Company
DIVISION OF ASHLAND OIL, INC.
RESEARCH AND DEVELOPMENT DIVISION • P. O. BOX [illegible] • [illegible]

February 13, 1978

TO: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, Virginia 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 January 1978 through 7 February 1978, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 2

Phosphoric acid screening tests continued with a trend observed showing low hexa levels are preferred. Tests were started to determine the effect of binder level on phosphoric acid resistance. A repeatability test was also started. Work was started to improve molding and curing techniques. Some failures occurred during a post cure step.

TASK 3

The effect of binder level on density, and hydrogen permeability was determined.

RESULTS AND DISCUSSION

TASK 2

Objective:

To screen resins on the basis of their resistance to phosphoric acid at 200°C by observing changes in size, weight, and flex strength.

Conclusions:

The results of the phosphoric acid endurance tests conducted to-date indicate that Arofene 875 and Arofene 877 have the greatest resistance to phosphoric acid among the resins tested, although the differences

between most of the resins are slight. The data also show that those resins containing lower levels of "hexa" possess better phosphoric acid resistance. The hexa level appears to be the most important factor relative to weight loss during the test.

Discussion:

Arofene 875 and 877 at 10% hexa were found to be slightly more resistant to phosphoric acid than the ERC controls in terms of weight loss. Arofene 860 and the experimental Resin A-33 show approximately the same weight loss as the commercial controls, while Arofene 2865 shows a relatively large weight gain. It appears therefore, that several resins are about equally resistant to phosphoric acid and that their resistance can be improved by decreasing the hexa content. Hence, final selection of the resin can be made on the basis of flow and molding characteristics among these several resins.

Table I lists the several resins tested showing the effect of hexa level.

Two groups of coupons were fabricated to test the effect of binder level and filter composition. One group contained a filler composed of 50% Asbury 4333, 45% Asbury A-60, and 5% Asbury Micro 840. The other group contained a filler having the Micro 840 graphite replaced with conductive carbon black, Cabot XC-72.

At low binder levels the coupons gained weight, confirming the results observed last month. Table II summarizes the phosphoric acid tests to-date.

A group of coupons was fabricated to determine repeatability of molding techniques and test results. The binder was Arofene 877 at 33% and the filler was composition A as given in Table II. The density and phosphoric acid endurance test results are given in Table III.

The coupons fabricated during January were post cured for 20 hours at 400-410°F. This represented a change from previous practice and was done to try to improve phosphoric acid resistance. Cracking occurred at the surface of some of the coupons, apparently a result of escaping gases formed by decomposition of hexa. It is believed that this problem can be overcome by molding for a longer time (20 minutes versus 10 minutes presently) and this will be attempted in the future.

TABLE I

PHOSPHORIC ACID RESINS SCREENING TESTS

Sample No.	Resin	% Hexa	Hours Aged @ 200°C	% weight change
11	Arofene 875	6	661	- 0.067
1		10	1044	- 0.65
12		14	661	- 1.66
7	Arofene 877	7	661	- 0.88
5		10	1044	- 1.03
8		13	661	- 4.70
ER-2	Commercial		1267	- 1.04
6	control		1044	- 1.46
14			661	- 0.16
15			661	- 1.24
4	A-33	10	1044	- 1.17
2	Arofene 860	10	1044	- 1.17
13		14	63 (failed)	--
9	Arofene 2869	6	661	+ 0.63
3		10	1044	+ 3.80
10		14	63 (failed)	--

Notes: All compositions at 33% binder

All except Commercial control contain the following graphite composition:

50% Asbury 4333
45% Asbury A-60
5% Asbury Micro 840

TABLE II

PHOSPHORIC ACID TESTS:

Effect of binder level and filler composition

Sample No	Filler Composition	% Binder	% Wt. Change
17	B	10	+ 9.50
19	B	20	- 0.26
20	B	25	--
21	B	30	- 0.38
22	B	33	+ 0.64
23	Commercial control	33	+ 0.42
24	A	15	+ 1.88
25	A	20	- 0.20
26	A	25	- 0.41
27	A	33	- 0.43

Notes:

Filler Compositions: A - 50% Asbury 4333
45% Asbury A-60
5% Asbury Micro 840

B - 50% Asbury 4333
45% Asbury A-60
5% Cabot XC-72 carbon black

Binder Composition: Arofene 877 with 10% hexa

All coupons aged 138 hours at 200°C in H_2PO_4

TABLE III

Repeatability Tests

Sample No	Density g/cc	% Wt. Change (138 hours)
28-1	1.710	- 0.46
28-2	1.695	- 0.46
28-3	1.701	- 0.48
28-4	1.695	- 0.45
28-5	1.659	+ 0.46
28-6	1.685	- 0.41

TASK 3

Objective:

To screen carbon fillers and to determine the practical range of carbon loading within which to work by measuring hydrogen permeability and electrical resistance at several levels of binder.

Conclusions:

A binder level of 25% is about optimum for the filler composition A as described in Table II. At 20% binder and below the coupons are permeable to hydrogen, and as binder level is increased, electrical resistance also increases.

Discussion:

The two groups of coupons that were prepared to test the effect of binder level and filler composition were also subjected to electrical resistance tests and hydrogen permeability tests. Table IV lists the density and hydrogen permeability data.

Electrical resistance measurements were made and although the values correlated with the density, the values appeared to be about 3 orders of magnitude larger than expected. The reason for this is being investigated. The problem may be due to a relative composition contact resistance. Various techniques will be tried including the use of high current density, improved surface preparation methods, and resistance measurements in the longitudinal direction.

FUTURE PLANS

TASK 2

A few of the Arofene resins which have not yet been tested will be fabricated into coupons this month and phosphoric screening will begin. The current tests will also continue. Changes in the molding and curing procedures will be tested to improve reliability of the tests.

TASK 3

Work will commence this month to improve the graphite composition and to maximize density and electrical conductivity density and electrical conductivity.

The investigation to improve the resistance measurements to obtain more reasonable values will continue. The problem may be due to the relatively low voltage used in the present technique. Measurements will be made at high voltage, and various surface preparation techniques will be tried to reduce contact resistance. Measurements will also be made in the longitudinal direction, which should minimize contact resistance effects.

TABLE IV

Effect of Binder Level and Filler Composition upon Density and Hydrogen Permeability

Sample No	Filler Composition	% Binder	Density g/cc	Hydrogen Permeability at 40 psig
20	B	25	1.741	0
21	B	30	1.706	0
22	B	33	1.658	0
23	Commercial control	33	1.702	0
24	A	15	1.829	+
25	A	20	1.846	*
26	A	25	1.784	0
27	A	33	1.688	0
28	A	33	1.697	0

* Not definitely known. Hydrogen flow was detected, but coupon was found to be chipped when removed from jig.

Binder was Arofene 877 with 10% hexa.

TO: R. V. Norton

DATE: March 1, 1978

FROM: P. M. Colling

SUBJECT: INTERIM REPORT - Bipolar Plate Research

REMARKS:

I. INTRODUCTION

The U.S. Army is developing hydrogen fuel cells to power certain portable and/or remote equipment in the 0.5 to 5 KW class. One area in which design and performance improvements are needed is the bipolar plate, a plate made of graphite resin composite which serves to separate the individual cells and to conduct the current from each cell to the next.

The bipolar plates must be electrically conductive, impervious to hydrogen and possess certain minimum mechanical properties including strength and impact resistance. They must maintain these properties under operating conditions of 150°C in concentrated phosphoric acid for several months. Bipolar plates have been fabricated which meet most of these requirements, but manufacturing methods are costly and reject rates are high. Long term stability under operating conditions has not been good. Failures result from swelling due to H_3PO_4 penetration into the plates.

It is also desirable to operate the cells at higher temperature, e.g., 190°C, to improve operating efficiency.

A program to select and evaluate various carbon resin composite in terms of physical properties and operating life under conditions approximating those in actual use has been undertaken to improve bipolar plates, e.g., longer life under present operating conditions, reduction in the rate of rejection during manufacture and ability to operate and survive at even higher temperatures up to 190°C.

II. OBJECTIVES

To screen, select, prepare, and evaluate various carbon resin composites which could be molded as bipolar plates for fuel cells.. The goal of this research is to develop carbon resin composites which are resistant to phosphoric acid at temperatures over 150°C and have high electrical conductivity, good strength, and dimensional stability.

III. RESEARCH PLANS

The research program, as contracted for, was comprised of 5 tasks:

1. Preliminary screening of resin and carbons based upon published sources and in-house information.

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2. Screening of resins by phosphoric acid endurance tests to select the two or three resins showing the best phosphoric acid resistance among those tested.

3. Screening of carbons and determination of practical ranges for carbon loading to select those carbon compositions and loadings giving the highest electrical conductivity.

4. Composite evaluation to determine the effects of resin type, carbon type, and carbon loading among those selected in Task 2 and 3.

5. Final report.

IV. CONCLUSIONS

The following tentative conclusions have been reached from the experimental program at the half-way point in the contract.

1. Particle packing techniques can be used to maximize compound density and to decrease the quantity of binder needed to fill the interstitial spaces to provide better electrical conductivity. Particle packing is achieved by selecting 2 or 3 graphites of widely different particle size ranges in order to allow the smaller particles to fill the voids between the larger particles.

2. Resins compounded at relatively low hexamethylenetetramine (hexa) levels (6%) show lower weight loss in phosphoric acid than those compounded at higher levels (10-14%). Whether this is a true improvement in chemical resistance or whether it is merely a loss of excess hexa has not yet been determined. Flex strength data may shed light on this subject.

3. A binder level of about 25% is probably optimum for the particular graphite composition selected. This may be decreased somewhat by selecting graphites giving greater compressed densities.

4. An improved molding technique has been developed which should help reduce rejection rates due to breaking of ribs and gas bubbles. It involves a two-step molding process which enables gases to escape from the mold between the 1st and 2nd steps.

5. Substitution of medium thermal carbon black for the smaller size graphite (<1μ) decreased the compressed densities of graphite mixtures.

6. Substitution of Cabot XC-72 conductive carbon black for the smaller sized graphite (Micro 840) resulted in slightly lower density and poorer physical properties of the molded coupon.

7. In spite of the conclusions put forth in the sixth monthly report suggesting that Arofene 875 and 877 were more resistant to phosphoric acid than the commercial control, more recent data are inconclusive. The weight change data appear to show a bias among all coupons from a single test container which is large relative to the differences between coupons.

V. DISCUSSION

Particle packing techniques to increase density, and thereby electrical conductivity of the molded part were investigated during the third month of the program. It was demonstrated that compressed densities of certain mixtures of graphite of different particle size were greater than those of single graphite grades. Optimization of the graphite mixture has not yet been completed, although the differences in densities among several mixtures of different compositions were relatively small. It was also shown that substitution of medium thermal carbon black for the smaller size graphite (<1μ) decreased the compressed density of the mixture.

Close examination of the weight change data from the phosphoric acid endurance tests for the first 15 tests revealed a bias among coupons from the same container which is large relative to the differences between coupons. Thus, conclusions based on small weight change differences between coupons are very tentative at this time. Other methods will be used to help detect differences between coupons including dimensional changes and appearance. Destructive testing such as flex strength tests must be postponed as long as possible in those cases in which only one coupon of a kind is available.

Whether the bias is a result of the changes in treatment conditions during the test or in the method used to prepare and weigh the coupons upon removal from the acid is not known. It should also be pointed out that in these cases none of the coupons had been post cured.

Recent coupons have been molded using an improved technique and have been post cured. Insufficient data are available from these more recent tests to detect a bias. Figure 1 is a plot of percent weight change versus hours aged for 11 of the first 15 coupons comprising two test containers. Visual examination of the lines connecting the points reveals the bias. Table 1 gives data in terms of difference from control for 13 of the first 15 coupons. The other two coupons failed after 65 hours of testing.

Even though a bias does exist, examination of the data in Table 1 shows very clearly that larger weight losses occur at higher hexa levels. Whether this is a simple result of loss of excess hexa is not known at present. Visual examination and dimensional measurements might reveal the mechanism.

A binder level of 25% is about optimum for the particular graphite composition selected based on hydrogen permeability tests. At 20% binder level and below, there appears to be a chance of some permeability. As binder level is increased, electrical conductivity would be expected to decrease, so it is advantageous to keep the binder level as low as possible without incurring permeability. Preliminary electrical conductivity measurements in fact showed this relationship, but the data are suspect and a new technique is being tried to reduce the effect of contact resistance. At this point 25% appears to be a safe place in which to operate.

Substitution of Cabot XC-72 conductive black for the smaller particle graphite gave slightly lower density and poorer physical properties to the molded coupon. This result was expected because of the smaller particle size and extremely high surface area of XC-72.

A new molding technique has been developed which may prove to be as important to bipolar plate manufacture and lifetime as resin selection. It involves a two-step process which permits most gases to be vented between the first and second step. It also provides for more uniform flow of the resin throughout the molded part and produces parts which show less tendency to crack and blister.

VI. PLANS

A. Tasks 2 and 3

1. Start phosphoric acid endurance tests on 5 more resins, and begin tests on 2 of the standard resins to determine the effect of 3% hexa level.

2. Develop a new procedure for measuring electrical resistance in the lengthwise direction to reduce contact resistance effects.

3. Carefully examine and measure dimensions of phosphoric acid endurance coupons presently being tested to try to improve reliability of the test.

4. Perform flex tests on selected coupons from the phosphoric acid endurance tests, comparing the results with similar coupons prepared at the same time, but not subjected to phosphoric acid aging.

5. Continue particle packing studies started in the third month of the program to maximize graphite compressed density.

B. Task 4

1. Select 3 or 4 of the resins showing the best performance to-date to compound with the graphite composition of the highest density. Compound at one or two binder levels (around 25%) preparing at least 12 coupons for each composition. Also, compound controls at the same binder level using the graphite composition and resin currently used by the commercial supplier. Subject these to very careful weight, dimensional, and density measurements, and conduct phosphoric acid endurance tests on one half of the coupons of each composition. Perform hydrogen permeability and electrical resistivity tests on coupons before and after aging. After about 3 months of aging, perform flex tests on the coupons.

PMC/cas

K•E 10 X 10 TO ½ INCH 7 X 10 INCHES
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FIGURE 1

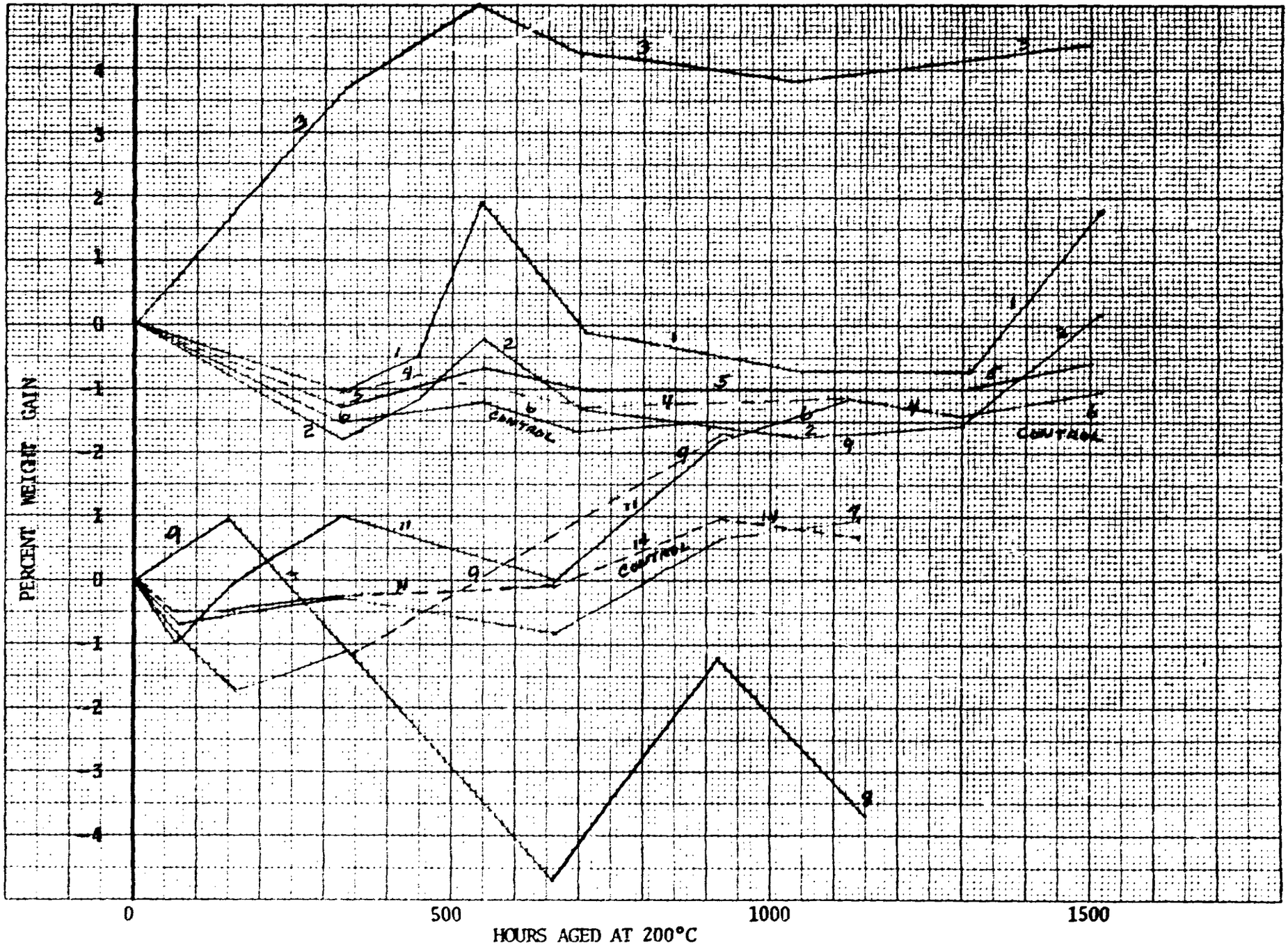

TABLE 1

Percent weight change
Difference from Control

A. CONTAINER 1

	COUPON NUMBER				
	1	2	3	4	5
HOURS AGED	875 (10%)	860 (10%)	2869 (10%)	A33 (10%)	877 (10%)
330	+ .49	- .20	+5.33	+ .5	+ .34
448	+ .99	+ .18	+5.9	+ .61	+ .44
538	+2.98	+ .89	+6.18	+ .31	+ .51
709	+1.43	+ .31	+5.83	+ .33	+ .59
1044	+ .81	- .31	+5.26	+ .29	+ .43
1307	+ .95	+ .01	+5.82	+ .14	+ .58
1521	+3.15	+1.66	+6.05	+ .44	+ .86

B. CONTAINER 2 & 3

	7	8	9	10	11
HOURS AGED	877 (7%)	877 (13%)	2869 (6%)	875 (6%)	875 (14%)
65	- .29	- .38	+ .87	- .47	- .48
155	- .12	-1.15	-1.52	+ .47	- .57
326	0	- .87	- .94	+1.3	- .58
661	- .72	-4.54	+ .79	+ .09	- .42
924	- .28	-2.13	+1.35	+1.25	- .96
1138	+ .22	-4.43	+1.52	+2.13	-1.51

NOTES:

The control is a commercial resin blend.

The numbers under the Coupon Number are the Ashland Arofene numbers.

The numbers in parentheses are % hexa in each resin.

Ashland

Ashland Chemical Company

DIVISION OF ASHLAND OIL, INC.

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March 17, 1978

TO: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, Virginia 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 February 1978 through 7 March 1978, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 2

An improved molding technique has been developed. Phosphoric acid tests continue on a wide variety of phenol-formaldehyde resins.

TASK 3

A new procedure has been developed for measuring resistivity to give more reliable results. The effects of binder level on resistivity were determined. Particle packing techniques for maximum density are discussed.

RESULTS AND DISCUSSION

TASK 2

Objective:

To screen resins on the basis of their resistance to phosphoric acid at 200°C by observing changes in size, weight, and flex strength.

Conclusions:

(1) An improved molding technique has been developed which should help reduce rejection rates due to breaking of ribs and gas bubbles. It involves a 2-step molding process which enables gases to escape from the mold between the first and second steps.

(2) Resins compounded at relatively low hexamethylenetetramine (hexa) levels (6%) show lower weight loss in phosphoric acid than those compounded at higher levels (10-14%).

(3) Repeatability of phosphoric acid endurance test is good with 5 of 6 coupons of the same composition showing weight changes within 0.1% after 776 hours.

(4) Low binder levels result in weight gains due to absorption of acid or water in the relatively porous coupons.

Discussion:

(1) A new molding technique has been developed which may prove to be as important to bipolar plate manufacture and lifetime as resin selection. It involves a 2-step process which permits most gases to be vented between the first and second step. It also provides for more uniform flow of the resin throughout the molded part and produces parts which show less tendency to crack and blister.

(2) Resins compounded at relatively low hexamethylenetetramine (hexa) levels (6%) show lower weight loss in phosphoric acid than those compounded at higher levels (10-14%). (See Table 1). Whether this is a true improvement in chemical resistance or whether it is merely a loss of excess hexa has not yet been determined. Flex strength data may shed light on this subject.

(3) Table III shows the results of the repeatability tests. Weight changes of 5 out of 6 coupons were within 0.1% of each other. One sample showed a significant increase in weight which was due to penetration of acid or water in a crack which developed. The problem of cracking has been resolved by the new 2-step molding technique.

(4) Table II shows the effect of binder level on weight changes. At low levels of binder (10-15%) relatively large wieght increases occur apparently due to absorption of water or acid in the relatively porous coupons.

TASK 3

Objective:

To screen carbon fillers and to determine the practical range of carbon loading within which to work by measuring hydrogen permeability and electrical resistance at several levels of binder.

Conclusions:

(1) A new procedure for measuring volume resistivity was developed which gives reproducible results that appear reasonable.

(2) Particle packing techniques can be used to maximize compound density and to decrease the quantity of binder needed to fill the interstitial spaces to provide better electrical conductivity. Particle packing is achieved by selecting two or three graphites of widely different particle size ranges in order to allow the smaller particles to fill the voids between the larger particles. Substitution of medium thermal carbon black for the smaller size graphite (less than 1 micron) decreased the compressed densities of graphite mixtures.

(3) A binder level of about 20-25% is probably optimum for the particular graphite composition selected. Electrical resistivity increases as binder level increases, but at lower binder levels (below about 20%) permeability to hydrogen may become a problem.

(4) Substitution of Cabot XC-72 conductive carbon black for the smaller sized graphite (Micro 840) resulted in slightly lower density and poorer physical properties of the molded compound.

TABLE 1

Percent Weight Change Difference From Control

A. CONTAINER 1

	COUPON NUMBER				
	1	2	3	4	5
HOURS AGED	875 (10%)	860 (10%)	2869 (10%)	A33 (10%)	877 (10%)
330	+ .49	- .20	+5.33	+ .5	+ .34
448	+ .99	+ .18	+5.9	+ .61	+ .44
538	+2.98	+ .89	+6.18	+ .31	+ .51
709	+1.43	+ .31	+5.83	+ .33	+ .59
1044	+ .81	- .31	+5.26	+ .29	+ .43
1307	+ .95	+ .01	+5.82	+ .14	+ .58
1521	+3.15	+1.66	+6.05	+ .44	+ .86
1780	+1.68	+ .23	+2.05	+ .49	.86
1780 hr. appearance	poor	warped pitting	chip	good	chip

B. CONTAINER 2 & 3

	7	8	9	11	12
HOURS AGED	877 (7%)	877 (13%)	2869 (6%)	875 (6%)	875 (14%)
65	- .29	- .38	+ .87	- .47	- .48
155	- .12	-1.15	+1.52	+ .47	- .57
326	0	- .87	- .94	+1.3	- .58
661	- .72	-4.54	+ .79	+ .09	- .42
924	- .28	-2.13	+1.35	+1.25	- .96
1138	+ .22	-4.43	+1.52	+2.13	-1.51
1397	+ .03	-4.1	+1.69	+2.37	-2.57
1397 hr. appearance	bulge	warped swollen pitted	slightly warped		Flaking along edge

NOTES:

The numbers under the Coupon Number are the Ashland Arofene numbers

The numbers in parentheses are % hexa in each resin.

TABLE II

Phosphoric Acid Tests:

Effect of binder level and filler Composition

Sample Number	Filler Composition	% Binder	Hours aged	% Weight Change 1.38	4.93	7.76	Appearance
17	B	10		+9.50	+6.71	+4.0	Warped Corner bad
19	B	20		-0.26	-0.33	-0.35	Good
20	B	25		--	-0.06	-0.14	Hairline crack
21	B	30		-0.38	-0.46	-0.57	Good
22	B	33		+0.64	+0.88	+1.15	Several cracks
23	Commercial control	33		+0.42	+0.03	+0.50	Cracked and bleeding
24	A	15		+1.88	+2.44	+2.63	Bleeding along edge
25	A	20		-0.20	-0.18	-0.20	Good
26	A	25		-0.41	-0.38	-0.53	Good, one bump
27	A	33		-0.43	-0.44	-0.47	Good

NOTES: Filler Compositions:

A - 50% Asbury 4333
45% Asbury A-60
5% Asbury Micro 840

B - 50% Asbury 4333
45% Asbury A-60
5% Cabot XC-72 Carbon Black

Binder Composition: Arofene 877 with 10% hexa

TABLE III

Repeatability Tests

Sample Number	Density g/cc	% Weight Change (776 hours)
28-1	1.710	-0.53
28-2	1.695	-0.44
28-3	1.701	-0.48
28-4	1.695	-0.40
28-5	1.659	+1.15
28-6	1.685	-0.45

TABLE IV

Percent Weight Loss Vs. % Hexa

Resin	% Hexa 6	10	14
A-33	-0.12g	-0.24g	+2.23 swollen
872	+0.90g	+0.33g	-0.23g
877	+0.83f	-0.67g -0.46g	+3.25 warped, pitted
890	+0.41g	+0.26chip	-0.79g

g - good

f - fair

Discussion:

(1) A new procedure was developed for the measurement of electrical resistivity whereby the resistivity was measured in the longitudinal direction in order to reduce the effect of contact resistance.

(2) Particle packing techniques to increase density, and thereby electrical conductivity, of the molded part were investigated during the third month of the program. It was demonstrated that compressed densities of certain mixtures of graphite of different particle size were greater than those of single graphite grades. Optimization of the graphite mixture is not yet been completed, although the differences in densities among several mixtures of different compositions were relatively small. It was also shown that substitution of medium thermal carbon black for the smaller size graphite (less than 1 micron) increased the compressed density of the mixture.

(3) A binder level of from 20 to 25% is about optimum for the particular graphite composition selected based on hydrogen permeability and resistivity tests. Below about 20% there appears to be a chance of some permeability. As binder level is increased, volume resistivity increases as shown in Table V, so it is advantageous to keep the binder level as low as possible without incurring permeability.

(4) Substitution of Cabot XC-72 conductive black for the smaller particle graphite gave slightly lower density and poorer physical properties to the molded coupons. This result was expected because of the smaller particle size and extremely high surface area of XC-72. Little or no effect on resistivity was detected as seen in Table V.

TABLE V

Coupon No. Number	Filler Composition	% Binder	Density g/cc	Volume Resistivity ohm-cm
17	B	10		0.0170
18	B	15		0.0342
19	B	20		0.0353
20	B	25	1.741	0.0747
21	B	30	1.706	0.1657
22	B	33	1.658	0.4360
24	A	15	1.829	0.0353
25	A	20	1.846	0.0844
26	A	25	1.784	0.0886
27	A	33	1.688	0.4157

PLANS

TASK 2

(1) Visually observe and compare all coupons subject to phosphoric acid tests to select those resins appearing to show the greatest promise.

(2) Perform flex tests on selected coupons from the phosphoric acid endurance tests to help select the most resistant resins.

(3) Continue phosphoric acid screen tests on the new resins recently subjected to aging.

TASK 3

(1) Optimize graphite composition for the aid of particle packing tests.

(2) Optimize binder level using electrical conductivity and hydrogen permeability as criteria.

TASK 4

At the end of the next reporting period (around April 7) three or four of the most promising resins will be selected and compounded with a graphite composition giving a high density. These will be compounded at or near the optimum binder level as determined in Task 3. Controls will also be compounded using a commercial composition. A minimum of 12 coupons of each composition will be compounded to provide plenty of samples for repeatability tests and flex tests. Very careful weight, dimensional and density measurements will be made on these coupons, and then 1/2 of the coupons of each composition will be subjected to phosphoric acid endurance tests for three months. Hydrogen permeability and resistivity tests will be preformed on the coupon before and after aging. Flex tests will be preformed on the aged coupons and the duplicate coupons not aged.

PMC/cas

Ashland

Ashland Chemical Company

DIVISION OF ASHLAND OIL, INC.

5200 PAUL G. BLAZER MEMORIAL PARKWAY, DUBLIN, OHIO 43017 • (614) 889-3333

REPLY TO:
P.O. Box 2219
Columbus, Ohio 43216

April 20, 1978

TO: Dr. Alayne A. Adams
U. S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, VA. 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 March 1978 through 7 April 1978, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 2

Four phenol-formaldehyde resins have been selected for more detailed studies in Task 4 based upon the phosphoric acid aging data of Task 2. They are Arofenes 872, 877, 890, and the experimental resin, A-33. Colloid 8440 will also be included in Task 4 based upon its commercial use in this system.

TASK 3

The molding technique has been altered slightly to mold the compounds at constant pressure rather than constant thickness. It was determined that 3000 psig is a suitable pressure to use for the remainder of the experimental program. This technique should give more reproducible results.

Various graphite compositions were tested to try to minimize volume resistivity. By substituting Asbury 7101 for Asbury A-60 in the formulation, the volume resistivity was decreased with little or no change in density. Volume resistivity can also be decreased by increasing the proportion of the finer graphite, Asbury Micro 840.

A binder level of about 22% appears to be near optimum for the particular graphite compositions used.

TABLE I

RESIN	% Hexa ⟶	3	6	10	14
872		G	G	G	G
875			B	✓	✓
877		G	G	G	B
890			G	G	G
2869			B	B	Fail
860				✓	Fail
A-33			G	G	✓
A-36			G	G	
A-35			Fail	Fail	
A-39			✓	G	

Key:

G - Good: Not more more 1% weight change during aging test and relatively good appearance

B - Bad: More than 3% weight change during aging test and relatively bad appearance

✓ Between 1% and 3% weight change

Fail: Complete failure - generally manifested through complete physical deterioration of compound, e.g., severe cracking, pitting, and dissolution of binder

RESULTS AND DISCUSSION

TASK 2

Objective:

To screen resins on the basis of their resistance to phosphoric acid at 200°C by observing changes in size, weight, and flex strength.

Conclusions:

The following resins have been tentatively selected for further study on the basis of aging studies of Task 2.

Arofene 872
" 877
" 890
A-33

The following resins show inferior performance in the phosphoric acid aging tests.

Arofene 875
" 2869
" 860
A-33

Two other experimental resins showing some promise but not selected for further study are A-36 and A-39.

Discussion:

Table I shows the overall results of the phosphoric acid aging studies based on weight changes and general appearance of the coupons.

Task 2 (cont'd)

Differences among those resins selected for further study appear to be slight. In general, performance was better at the lower hexa level, but little or no difference was noted between 3% and 6% hexa. Therefore, 6% hexa will be used in Task 4.

TASK 3

Objective:

To screen carbon fillers and to determine the practical range of carbon loading within which to work by measuring hydrogen permeability and electrical resistance at several levels of binder.

Conclusions:

Molding at a constant pressure of 3000 psig results in more uniform coupons and more reproducible results. The substitution of Asbury 7101 graphite for Asbury A-60 in the same proportion gives lower volume resistivity.

Volume resistivity increases with increasing proportion of fine graphite such as Micro 840.

A binder level of 22% is near optimum for the particular graphite compositions tested.

Discussion:

Molding Techniques: The molding technique was changed to mold at constant pressure rather than constant thickness. In order to accomplish this, additional resin graphite mixture was added to the mold cavity (13 grams vs. 12 grams) and the pressure was decreased to insure that the mold would not completely close. It was noted that the coupons molded at constant thickness actually varied in thickness due to the elastic behavior of graphite. It was also suspected that the molding pressure of some 6000 psig was too high and could result in crushing of the graphite and would not be practical for molding larger plates because of flexing of the mold. A simple test was performed at 2000 psig and 3000 psig, and at 12 grams and 13 grams of the mixture to determine suitable conditions.

By measuring the thickness of the molded coupons it was determined that 13 grams composite afforded sufficient coupon thickness to prevent closing of the mold to the stop.

It was also noted that most coupons molded at 2000 psig exhibited hydrogen permeability at 10 psig hydrogen pressure, while those molded at 3000 psig did not. Hence, 3000 psig was chosen as the pressure to use for the remainder of the program.

Effect of Graphite Composition on Density and Volume Resistivity

Table II shows the effect of graphite composition on density and volume resistivity.

Substitution of Asbury 7101 for A-60 (see 70 vs. 69) appears to lower volume resistivity in two of three coupons.

Substitution of Colloid 8440 has no significant effect on volume resistivity (see 71 vs. 69).

Increasing the finer graphite (840) decreases volume resistivity (see 73 vs. 70).

Little or no effect on density was noted for the compositions tests.

The binder level of 22% appears to be near optimum for these compositions, but may have to be increased as the proportion of finer graphite is increased.

Compressed Densities of Graphite Mixtures

Compressed densities of four graphite mixtures were determined to try to optimize the graphite composition. The data are shown in Table III.

The substitution of 7101 for A-60 results in a small density increase.

PLANS

TASK 2

Start phosphoric acid endurance test on four additional resins and on two resins using curing agents different from hexa.

Prepare radiochemical labeled coupons and extract with phosphoric acid to determine if labeled species are extracted into the phosphoric acid.

TASK 3

Prepare 3 coupons having different graphite compositions to help select the best composition.

TASK 4

Prepare coupons containing the following resins for detailed study:

Arofene 872
Arofene 877
Arofene 890
A-33
Colloid 8440

These will be compounded with a graphite composition to be selected from the results of Task 3. Binder level will be about 22%.

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TABLE II

Coupon Number	Graphite Composition	Density g/cc	Vol. Resist. ohm-cm
69-1	A4333 (50) A-60 (45) 840 (5)	1.742	0.1219
69-2	" " " " " "	1.740	0.1240
69-3	" " " " " "	1.764	0.1114
70-1	A4333 (50) 7101 (45) 840 (5)	1.760	0.1280
70-2	" " " " " "	1.747	0.0946
70-3	" " " " " "	1.773	0.0835
71-1*	A4333 (50) A-60 (45) 840 (5)	1.804	0.1096
71-2*	" " " " " "	1.746	0.1168
71-3*	" " " " " "	1.751	0.1343
72-1	A4333 (65) 7101 (30) 840 (5)	1.798	0.1171
72-2	" " " " " "	1.759	0.0875
72-3	" " " " " "	1.739	0.0946
73-1	A4333 (50) 7101 (40) 840 (10)	1.743	0.0771
73-2	" " " " " "	1.786	0.0705
73-3	" " " " " "	1.716	0.0672

All coupons molded at 3000 psig.

Binder level 22%.

Binder Arofene 872 with 6% hexa except No. 71

*NO. 71 binder was colloid 8440.

TABLE III

Pressure (psig)	Density g/cc A	B	C	D
1116	1.402	1.456	1.599	1.446
1674	1.519	1.487	1.610	1.567
2232	1.591	1.541	1.633	1.649
2790	1.661	1.591	1.715	1.720
3348	1.720	1.637	1.778	1.778
3906	1.768	1.678	1.832	1.833
4464	1.808	1.706	1.876	1.876
5022	1.845	1.738	1.908	1.914

Compositions:

A 4333 (50) A-60 (45) 840 (5)

B 4333 (65) A-60 (30) 840 (5)

C 4333 (50) 7101 (45) 840 (5)

D 4333 (65) 7101 (30) 840 (5)

Ashland

Ashland Chemical Company

DIVISION OF ASHLAND OIL, INC.

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May 16, 1978

REPLY TO:
P.O. Box 2219
Columbus, Ohio 43216

TO: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, VA. 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 April 1978 through 7 May 1978, Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 2

Carbon-14 labeled coupons were prepared and extracted with hot phosphoric acid to see if the labeled species were extracted into the acid. The results were positive with a small (<1%), but definite portion of the radioactivity transferred to the acid.

The phosphoric acid endurance tests were terminated on the first 30 coupons so that flex tests could be performed. These coupons had been aged from approximately 2000 to 3000 hours.

TASK 3

Several coupons were prepared having varying compositions to help optimize graphite composition and binder level. Although the substitution of a finer graphite for the largest particle graphite in the composition resulted in a coupon having a smoother surface, volume resistivity was increased some 30%.

RESULTS AND DISCUSSION

TASK 2

Objective:

To screen resins on the basis of their resistance to phosphoric acid at 200°C by observing changes in size, weight, and flex strength.

Discussion:

The phosphoric acid tests were terminated on the first 30 coupons so that flex tests could be performed. The results of these flex tests and final weight changes will be reported next month.

Carbon-14 labeled coupons were prepared and extracted with hot phosphoric acid to see if the labeled species were extracted into the acid. The following description of the experiment and results has been excerpted from a report by Dr. Leonard Hughes of the Physical Chemistry Section who preformed the radiochemical extractions. The fact that most of the radioactivity is extracted in the first few hours suggests the use of a phosphoric acid pre-treatment of the bipolar plates before stack assembly.

Preparation of Labeled Resin

This was synthesized from phenol and C-14 labeled formaldehyde using approximately the plant procedure. The melt time of the finished, sieved resin was somewhat off-spec (155 sec), indicating the presence of free phenol. Nonetheless, we decided to make the graphite cell plates with this resin.

Preparation of Plates

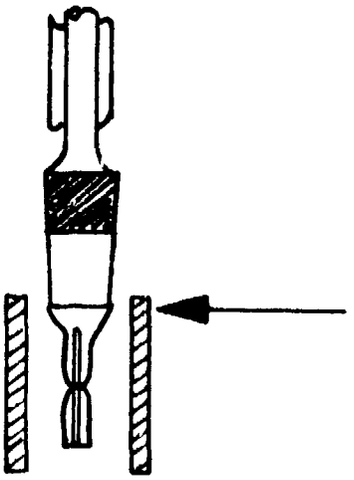
John Martin made six each of 33% and 22% resin plates following his normal procedure. The weights were recorded, with the exception of one 33% plate used in the first leaching experiment.

Leaching Conditions

Eighty-five percent acid boils at about 155°C and so the desired temperature at 200°C could not be attained. To circumvent this, each 50 ml samples of acid was pre-heated in the glass container with an open top until the temperature increased at 200°C. The volume loss usually was between 10 and 15 mls. The test plate was added to the hot acid and a thermometer inserted. An air condenser was used. The electrolyte was kept in contact with the plate for an overnight period without agitation.

Apparatus

The glass vessel shown in the diagram was fabricated:



The squared-off section of the vessel was constructed to minimize the amount of electrolyte required to cover the plate. It was dimpled so that the test plate did not touch the walls, and there was enough space to include a thermometer. The condenser was added to ensure that further water was not lost.

Sampling Procedure

The pre-boiled acid is too viscous to enable a small sample to be accurately pipetted out into a counting vial. This problem was overcome by diluting 2 mls with 2 mls water, thoroughly mixing and cooling and pipetting out 250 µl of the diluted acid for counting. Usually the sample was counted for 100 minutes in dioxane cocktail. The total volume of the acid was noted so that the total, leached radioactivity could be calculated. Each bulk sample of acid was retained in a capped bottle.

It was necessary to calibrate the liquid scintillation counter for acid, water and color quenching. This was done by taking a sample of acid used to treat a non-radioactive plate and spiking solutions of known amounts of C-14 activity (C-14 toluene). Using this standard procedure, a calibration curve was obtained.

The amount of C-14 activity in each plate was calculated from the known specific activity of the resin:

	dpm/plate
33% resin	46.78×10^6
22% resin	31.18×10^6

Experimental Design

The initial objective was to measure any leaching of radioactivity by the acid within a 12-hour period. The experiment was then expanded to include additional treatments of a single plate with fresh batches of phosphoric acid. This was done for the 33% plate first and then for the 22% plate. At each stage the total counts leached was measured.

Results

The first treatment of the 33% plate showed a greater radioactivity loss than the subsequent treatment. Furthermore, during the first treatment, the loss did not increase substantially beyond that at the 12-hour level when the heating time was extended to 70 hours. This suggested either a saturation of the acid or some other limit to extractable resin. The former hypothesis is discounted by the fact that further fresh acid shows less leaching than the first batch of acid. The results are shown in the table.

Acid Batch#	33% Resin Hours	33% Resin dpm's leached	33% Resin %	22% Resin Hours	22% Resin dpm's leached	22% Resin %
1	10½	64,224	0.14	16	78.672	0.25
	70	77,248	0.17			
2	16	8,060	0.011	16	49,077	0.16
3	17	5,776	0.012			
4	16	4,992	0.010			

TASK 3

Objective:

To screen carbon fillers and to determine the practical range of carbon loading within which to work by measuring hydrogen permeability and electrical resistance at several levels of binder.

Discussion:

The results of the tests to help optimize graphite composition and binder level are given in Table 1. The standard two-step molding technique at 3000 psig was used with postcuring.

Most of the coupons show some hydrogen permeability when tested at 5 psig. This suggests that binder level and/or molding pressure need to be increased.

Additional tests are being made to separate the effects of the graphites on volume resistivity. Also, higher molding pressures and increased binder levels will be tried.

The flex strength tests probably have little meaning since they were all single test values. There does appear to be a possible advantage to using finer graphite in terms of flex strength.

PLANS

TASK 2

Complete radio tracer extraction studies on C-14 labeled coupons.

Terminate phosphoric acid endurance tests 31 through 68 and perform flex tests on the coupons.

Start phosphoric acid endurance tests on four additional resins and on two resins using curing agents different from hexa.

TASK 3

Prepare coupons for final selection of graphite and binder levels for Task 4.

TASK 4

Prepare coupons containing the following resins for detailed study:

Arofene 872
" 877
" 890
A-33
Colloid 8440

The effects of binder level and graphite compositions will also be determined by including a 2 x 2 factorial for one of the resins.

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TABLE I

Coupon Number	GRAPHITE COMPOSITION, Wt. % 4333	4015	4110	A-60	7101	840	250	% Binder	Resin	Density	Volume Resistivity	H_2 Flow at 5 psi	Flex strength psi
69	50			45		5		22	A	1.749*	0.1188*	+	
70	50				45	5		22	A	1.760*	0.1020*	+	2492
71	50			45		5		22	B	1.767*	0.1202*	0	2621
72	65				30	5		22	A	1.765*	0.0997*	+	3447
73	50				40	10		22	A	1.748*	0.0716*	Slow	2921
74	45				40	15		22	C	1.748	0.0682	+	1378
75	40				50	10		22	C	1.723	0.0714	+	--
76	35				50	15		22	C	1.707	0.0645	+	1570
77		40			50		10	22	A	1.824	0.1295	Slow	5229
78		40			50		10	25	A	1.717	0.118	--	--
79			40		50		10	22	A	1.845	0.1069	Slow	4113
80			40		50		10	25	A	1.733	0.0936	+	3405
81			40		50		10	25	A'	1.633	0.1244	+	2961
82			40		50		10	25	A''	1.778	0.1043	+	2697
83	30				60		10	25	A	1.786**	0.1178**	Slow	--
84	30				60		10	30	A	1.785**	0.1848**	Slow	--

* Average of 3 values
** Average of 2 values
All other single values

All coupons molded at 3000 psig

Resin Code: A - Arofene 872 with 6% hexa
B - Colloid 8440
C - Arofene 872 with 10% hexa
A' Arofene 872 with 6% hexa + 1% Stearate added
A'' Arofene 872 with 6% hexa + 1% Cab-o-sil added

Ashland

Ashland Chemical Company

DIVISION OF ASHLAND OIL, INC.

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REPLY TO:
P.O. Box 2219
Columbus, Ohio 43216

June 15, 1978

TO: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, VA. 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report - 8 May 1978 through 7 June 1978 Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 2

The phosphoric acid screening tests have been completed except for flex tests which will be reported as the data are received. The radiotracer study has been completed. This short study showed that a greater absolute quantity of radioactivity was leached from the coupon containing the lower quantity of resin and that the rate of loss of radioactivity decreased rapidly with extraction time.

TASK 3

Task 3 has been completed with the preparation and characterization of four additional coupons to help optimize molding pressure and graphite composition.

TASK 4

Task 4 was started with the preparation of 10 groups of 8 coupons each. The 90 day aging test was started on 4 coupons from each group.

RESULTS AND DISCUSSION

TASK 2

Objective:

To screen resins on the basis of their resistance to phosphoric acid at 200°C by observing changes in size, weight, and flex strength.

Discussion:

The phosphoric acid tests have been terminated on the first 68 coupons so that flex tests can be performed. Flex tests are not yet complete. These data will be reported as they are received.

The radiotracer extraction studies started and reported last month were completed. The final results are listed in Table I. Somewhat surprisingly the coupon with the lower binder level is the one with the greatest extraction rate. This may be due to higher porosity of the coupon resulting in increased surface area of binder exposed to the acid.

TASK 3

Objective:

To screen carbon fillers and to determine the practical range of carbon loading within which to work by measuring hydrogen permeability and electrical resistance at several levels of binder.

Discussion:

Four additional coupons were prepared to see if increasing the molding pressure to 4000 psig would increase density and decrease volume resistivity, and to compare Asbury micro 840 with Asbury 250. The results are listed in Table II. Little difference was noted between the different graphite mixtures, but a slight improvement was noted at 4000 psig so it was decided to use 4000 psig molding pressure for coupons in Task 4.

TASK 4

Objective:

To determine the effects of selected resins and graphite upon physical properties and durability in hot phosphoric acid.

Discussion:

Eighty coupons representing 10 different compositions were prepared for the Task 4 aging studies. One-half of these coupons were placed in the containers of H_3PO_4 on June 2 to begin a 90-day aging test. Four coupons of each composition are being aged and four are being stored for determination of flex tests at the end of the aging period.

Density and volume resistivity were determined on each coupon, and hydrogen permeability tests were performed on selected coupons from each group. These data are presented in Table III.

PLANS

TASK 2

Prepare coupons from additional experimental resins as time permits.

Tabulate and report flex strength data when received.

TASK 3

No more work is planned on Task 3.

TASK 4

Weigh aged coupons and replace acid every 10 days for 3 months. After 3 months the coupons will be subjected to flex strength tests along with unaged counterparts.

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TABLE I

Phosphoric Acid Leaching of C-14 labeled Coupons

Percent Resin	dpm/plate	Cumulative hours leached	Cumulative dpm's leached	Cumulative % leached
33	46.78×10^6	10.	66,224	0.14
		80.	77,248	0.17
		96	85,308	0.18
		113	91,084	0.19
		129	96,076	0.21
22	31.18×10^6	16	78,672	0.25
		32	127,749	0.41
		64	169,093	0.54
		80	188,365	0.60
		96	203,125	0.65

TABLE II

Coupon Number	Resin	Filler	Resin %	Molding Pressure psig	Density g/cc	Volume Resistivity OHM-CM	Hydrogen Permeability @5 psig	@ 10 psig
85-1	Arofene 877 (6% Hexa)	E	25	3000	1.760	0.1789	Very slow	Slow
85-2	" "	E	25	4000	1.790	0.1668	" "	"
86-1	" "	F	25	3000	1.769	0.1926	" "	"
86-1	" "	F	25	4000	1.788	0.1771	+	+

FILLER COMPOSITIONS:

E - Asbury 4333 (50%)
7101 (40%)
840 (10%)

F - Asbury 4333 (50%)
7101 (40%)
250 (10%)

TABLE III

Coupon	Resin	Filler	Resin %	Density g/cc	Volume Resistivity OHM-CM	H_2 Flow @ 5 psig	H_2 Flow @ 5 psig
88-1	Arofene 872	C	25	1.708	0.1456	0	0
2				1.744	0.1438		
3				1.769	0.1383		
4				1.760	0.1419		
5				1.755	0.1610		
6				1.750	0.1471		
7				1.733	0.1520		
8				1.743	0.1435		
Ave				1.745	0.14665		
σ				0.0187	0.007		
89-1	Arofene 877	C	25	1.768	0.1975	0	Very slow
2				1.766	0.2049		
3				1.742	0.1944		
4				1.749	0.1996		
5				1.772	0.2106		
6				1.744	0.1965		
7				1.777	0.2100		
8				1.756	0.1885		
Ave				1.759	0.2003		
σ				0.0133	0.0077		
90-1	Arofene 890	C	25	1.758	0.2171	0	0
2				1.765	0.2383		
3				1.773	0.2119		
4				1.746	0.1876		
5				1.783	0.2105		
6				1.735	0.2527		
7				1.760	0.1824		
8				1.765	0.2012		
Ave				1.761	0.2127		
σ				0.0150	0.0238		

Filler Compositions: C - Asbury 4015 (40%)
7101 (50%)
250 (10%)

D - Asbury 99 (73%)
850 (27%)

NOTE: All Arofene Resins contain 6% Hexa.

TABLE III (Cont'd)

Coupon	Resin	Filler	Resin	Density g/cc	Volume Resistivity OHM-CM	H_2 Flow @ 5 psig	H_2 Flow @ 10 psig
91-1	A-33	C	25	1.760	0.1799	0	0
2				1.744	0.2045		
3				1.757	0.1935		
4				1.746	0.1931		
5				1.760	0.2422		
6				1.731	0.2248		
7				1.757	0.2492		
8				1.735	0.1843		
Ave				1.749	0.2090		
σ				0.0115	0.0266		
92-1	Colloid 8440	C	25	1.717	0.1749	+	+
2				1.741	0.2235		
3				1.737	0.2152		
4				1.724	0.2110		
5				1.724	0.1963		
6				1.712	0.1852		
7				1.697	0.1718		
8				1.715	0.2383		
Ave				1.721	0.2020		
σ				0.0141	0.0239		
93-1	Commercial Control			1.683	0.1659	slow	slow
2				1.699	0.2163		
3				1.710	0.1731		
4				1.719	0.1944		
5				1.687	0.1914		
6				1.697	0.2169		
7				1.707	0.1921		
8				1.681	0.1689		
Ave				1.698	0.1899		
σ				0.0136	0.0198		
94-1	Colloid 8440	C	33	1.667	0.4694		
2				1.670	0.4438		
3				1.671	0.4706		
4				1.654	0.5552		
5				1.674	0.3907	+	+
6				1.671	0.3876	+	+
7				1.656	0.4623		
8				1.654	0.3921		
Ave				1.665	0.4465		
σ				0.0085	0.0570		

TABLE III (Cont'd)

Coupon	Resin	Filler	Resin	Density g/cc	Volume Resistivity OHM-CM	H_2 Flow @ 5 psig	H_2 Flow @ 10 psig
95-1	Commercial			1.698	0.2149	0	0
2	Control			1.673	0.1302		
3				1.726	0.1905		
4				1.723	0.2105		
5				--	--		
6				--	--		
7				--	--		
8				--	--		
Ave				1.705	0.1865		
σ				0.0248	0.0390		
96-1	Colloid	D	25	1.749	0.0767	Slow	
2	8440			1.754	0.0745	Slow	
3				1.739	0.0669		
4				1.747	0.0648		
5							
6							
7							
8							
Ave				1.747	0.0707		
σ				0.0062	0.0058		
97-1	Colloid	D	33	1.719	0.1659	0	very slow
2	8440			1.686	0.1804		
3				1.704	0.1469		
4				1.717	0.1844		
5				1.711	0.1728		
6				1.698	0.1464		
7				1.732	0.1607		
8				1.709	0.1366		
Ave				1.7095	0.1618		
σ				0.014	0.0173		

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REPLY TO:
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July 7, 1978

TO: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, VA. 22060

FROM: P. M. Colling

SUBJECT: Monthly Letter Progress Report- 8 June 1978 through 7 July 1978 Contract No. DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 2

The final results of the phosphoric acid aging tests are reported in tabular form. These data include composition of the coupons, number of hours aged, percent weight change, appearance, and flex strength of aged and unaged coupons.

TASK 4

The results of the first months aging tests are reported.

RESULTS AND DISCUSSION

TASK 2

Objective:

To screen resins on the basis of their resistance to phosphoric acid at 200°C by observing changes in size, weight, and flex strength.

Discussion:

The final results of the aging tests are reported in Table I. The data are tabulated in numerical order. No attempt was made to group the data by composition or experimental objective since the conclusions from these tests have been documented in previous reports. The principal observation that has not been previously reported is that generally the breaking strength is lower after aging than before. For example, in 61 of 78 cases the aged coupon had lower breaking strength

than the unaged coupon. There are 13 other examples reported on aged coupons in which no unaged counterpoints were flex tested.

The "Final Physical Appearance" column in Table I is a subjective evaluation by the chemist and consists of a numerical rating (0 to 5 scale) and a description of the aged coupon.

TASK 4

Objective:

To determine the effects of selected resins and graphite upon physical properties and durability in hot phosphoric acid.

Discussion:

Weight change data after about one month of aging in hot phosphoric acid at 200°C are presented in Table II. For each of the 40 coupons being aged, a similar coupon is being retained unaged for flex strength tests to be performed and the end of the aging test in September.

PLANS

TASK 4

Measure electrical resistance through coupons and carbon paper substrate at different pressures. The purpose of this experiment is to determine how volume resistivity of the coupon affects the contact resistance between the coupons and carbon paper at various pressures. Previous data from Energy Research Corporation indicated that about 90% of the total resistance was due to contact resistance between the bipolar plates.

Continue the Task 4 aging tests, weighing the coupons and replacing the acid every 7 to 10 days.

PMC/cas

TABLE I

Coupon Number	COMPOSITION Binder	% Hexa	% Binder	Filler	Total Aged Hours	Weight Change %	FINAL PHYSICAL APPEARANCE Rating	Description	Flex breaking strength (psi) Unaged	Aged
1	Arofene 875	10	33	A	2956	-5.88	1	Warped, bleeding	--	3840
2	860	10	33	A	2956	-4.55	2	Slight warp-flaked	--	4977
3	2869	10	33	A	2956	+4.78	3	Side chipped - OK	6909	4612
4	A-33	10	33	A	2956	-2.02	2	Slight warp	5081	6089
5	877	10	33	A	2956	-1.80	3	Minor chip on edge	7644	7662
6	Commercial Control No. 1				2956	-3.90	2	One crack-ship where drilled		8693
7	Arofene 877	7	33	A	2573	+1.56	2	Bumpy	9500	5152
8	877	13	33	A	2573	-8.38	1	Grainy-warped	9097	5911
9	2869	6	33	A	2573	+2.77	3	Fair to good	7883	4055
10	2869	14	33	A	65	--	0	Severely decomposed after 65 hrs		--
11	875	6	33	A	2573	+3.43	2	Bumpy	8745	4358
12	875	14	33	A	2573	-1.64	2	Side rough & chipped	8580	7140
13	860	14	33	A	65	--	0	Severely decomposed after 65 hrs		--
14	Commercial Control No. 1				2573	+0.85	2		--	6175
15	" " " "				2573	-0.43	2	Pitted and cracked	--	6060
16	" " " "				--	--	-	--	Not tested	
17	Arofene 877	10	10	B	1951	+5.89	1	Warped-shiped bad	3123/4196	1840
18	877	10	15	B	--	--	-	--	Not tested	
19	877	10	20	B	1951	-0.33	3	Good	8345/7074	4830
20	877	10	25	B	1951	+0.07	2	Cracked, bleeding	7526	5953
21	877	10	30	B	1951	-0.60	4	Good to excellent	8220	8703
22	877	10	33	B	1951	+1.92	2	Cracked, bleeding	9073/9294	5077
23	Commercial Control No. 1				1951	+0.79	2	Cracked, bleeding	--	5199
24	Arofene 877	10	15	A	1951	+4.48	3	Side chipped - OK	5981	4224
25	877	10	20	A	1951	0	3	Good	8090/8334	5259
26	877	10	25	A	1951	-0.45	2	Some blisters	7470	8050
27	877	10	33	A	1951	-0.27	3	Good	6102	8582
28-1	877	10	33	A	1951	-0.46	3.5	Good	9048	8417
28-2	877	10	33	A	1951	-0.49	3.5	Good	8712	8194
28-3	877	10	33	A	1951	-0.46	3.5	Good	9533	8067
28-4	877	10	33	A	1951	-0.37	3.5	Good	9140	8853
28-5	877	10	33	A	1951	+1.78	2	Cracked, bleeding	7518	4895
28-6	877	10	33	A	1951	-0.44	3.5	Good		8644
29	Commercial Control No. 1				1951	+1.53	2	Cracked, bleeding	8934	5152
30-1	Arofene 877	10	33	A	1951	+10.44	2	Cracked		5297
30-2	877	10	33	A	1951	+0.33	3	Good	4546	3729

FILLER COMPOSITIONS: A - 50% Asbury 4333
45% Asbury A-60
5% Asbury Micro 840

B - 50% Asbury 433
45% Asbury A-60
5% Cabot XC-72 Carbon Black

TABLE I (Cont'd)

Coupon Number	COMPOSITION Binder	% Hexa	% Binder	Filler	Total Aged Hours	Weight Change %	FINAL PHYSICAL APPEARANCE Rating	Description	Flex breaking strength (psi) Unaged	Aged
31	Arofene 877	10	33	A	2139	-0.34	4	Good	9401	8675
32	A-33	6	33	A	2139	+0.42	4	Good	8380/7102	7431
33	A-33	10	33	A	2139	+0.22	4	Good	7482	7754
34	A-33	14	33	A	2139	+4.16	3	Good	5278/8822	4187
35	Arofene 872	6	33	A	2139	+1.95	3	Good	9113/7323	6656
36	872	10	33	A	2139	+1.33	4	Good	7441/8418	7379
37	872	14	33	A	2139	+0.32	3	Good	8450/9309	6845
38	877	6	33	A	2139	+3.97	2	Bumpy	9948/9806	5293
39	877	10	33	A	2139	-0.17	3	Good	7934/7567	6923
40	877	14	33	A	2139	+7.40	1	Warped	7169/3097	3279
41	890	6	33	A	2139	+3.32	1	Bumpy, bleeding	10291/9077	4924
42	890	10	33	A	2139	+1.10	3	Good	5916/7255	6869
43	899	14	33	A	2139	-0.65	4	Good	7134/5357	8694
44	Commercial Control No. 1				2139	-9.38	1	Bad	--	4292
45	" " " "				2139	-0.91	1	Decayed, porous	--	5490
46	" " " "				1857	+0.24	4	Good	--	9322
47	" " " "				1857	+6.65	2	Warped	--	4188
49	Arofene A-39	6	33	A	1857	+2.24	3	O.K.	7843/6624	5662
50	A-39	10	33	A	1857	+2.13	2	Fair	7451/8016	6346
51	A-36	14	33	A	1857	+4.52	2	Bumpy	7814	4569
52	A-36	6	33	A	1857	+3.38	2	Bumpy	6437	6401
53	A-35	10	33	A	185		0	Failed early	9137	8961
54	A-35	14	33	A	185		0	" "	7219	8210
55	877	6	33	A	1857	+1.26	3	Decent	9667/10399	7664
56	872	10	33	A	1857	+3.55	3	Decent	2867/6708	4708
57	877	3	33	A	1857	+2.01	2	Bumpy, bleeding	9687/9557	5544
58	877	6	33	A	1857	+1.15	3	Decent	10088/7864	7881
59	872	3	33	A	1857	+2.18	2	Bumpy, grainy	9799/6624	5503
60	875	6	33	A	1857	+4.75	2	O.K.	9218/8880	4799
63-2	872	6	20	A	400	+1.16	3	Fair to good	5037	7729
63-3	872	6	20	A	400	+0.75	3	Fair to good	--	6638
64-1	872	6	20	A	400	+5.23	1	Cracked	5486	3476
64-3	872	6	20	A	400	+16.53	1	Warped	--	--
67	872	6	20	A	400	+0.98	3	Good	5685/5701	6985
68	872	6	20	A	400	+10.08	2	Warped, flaked	4462/5266	1366

ALL COUPONS NO. 31-68 were molded using a two-step process. Coupons 31-43 were post cured at 370°F for 16 hours. Coupons 46-60 post cured 20 hours at 385°F. Coupons 63-68 were molded at constant pressure and post cured at 450°F for 20 hours.

TABLE II

Coupon	Resin	Filler	Resin %	Weight Change %
88-2	Arofene 872	C	25%	1.57
3				.73
4				3.15
5				2.42
89-1	Arofene 877	C	25%	1.72
2				1.23
3				4.50
4				3.81
90-1	Arofene 890	C	25%	1.14
2				1.26
3				.88
4				1.36
91-1	A-33	C	25%	.27
2				.23
3				.61
4				1.48
92-3	Colloid 8440	C	25%	3.73
4				14.28
5				5.16
8				3.68
93-1	Commercial Control			8.14
2				5.30
3				5.84
6				4.48
94-1	Colloid 8440	C	33%	2.49
2				3.74
3				3.91
4				2.65
95-1	Commercial Control			1.35
2				1.02
3				3.26
4				.41
96-1	Colloid 8440	D	25%	12.11
2				12.91
3				18.12
4				9.43
97-1	Colloid 8440	D	33%	3.49
2				6.38
3				3.69
5				2.93

Ashland

Ashland Chemical Company

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August 9, 1978

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TO: Dr. Alayne A. Adams
U.S. Army Mobility Equipment R&D Command
Electrical Power Laboratory
Fort Belvoir, Virginia 22060

FROM: J. E. Martin

SUBJECT: Monthly Letter Progress Report - 7 July 1978 through 8 August 1978, Contract DAAK70-77-C-0151 "Selection and Evaluation of Carbon Resin Composites for Bipolar Plates for Hydrogen Fuel Cells"

SUMMARY

TASK 4

The results of the final aging tests are reported.

RESULTS AND DISCUSSION

TASK 4

Composite evaluation

Objective:

Task 4 will be a compounding study to determine the effects of resin type, carbon type, and carbon loading upon physical properties and durability in hot H_3PO_4 of the cured part. The dependent variables will include the following: flex strength, impact resistance, permeability to hydrogen, aging properties in hot H_3PO_4, weight change and dimensional stability, and retention of tensile, flex strength, hardness, and electrical conductivity.

Discussion:

The final results of the aging tests are reported in Table 1.

The "Final Physical Appearance" column in Table I is a subjective evaluation and consists of a numerical rating (0 to 5 scale) and a description of the aged coupon.

Plans:

TASK 4

The four aged samples from each series and the unaged counterparts will be measured and break retention strengths will be compared.

An in-house pneumatic apparatus has been constructed to measure electrical resistance through coupons at different pressures.

TASK 5

Begin preparation for final report.

JEM/cas

TABLE I

Coupon Number	COMPOSITION Binder	% Hexa	% Binder	Filler	Total hours aged	Weight Changed %	Final Rating	Physical Appearance description
88-2	Arofene 872	6	25	A	1558	+2.58	3	Good
88-3	872	6	25	A	1558	+1.19	3	Good
88-4	872	6	25	A	1558	+4.92	3	Good
88-5	872	6	25	A	1558	+4.10	3	Good
89-1	877	6	25	A	1558	+2.97	3	Good
89-2	877	6	25	A	1558	+2.07	3	Good
89-3	877	6	26	A	1558	+5.46	3	Good
89-4	877	6	25	A	1558	+4.22	3	Good
90-1	890	6	25	A	1558	+1.13	3	Good
90-2	890	6	25	A	1558	+1.19	3	Good
90-3	890	6	25	A	1558	+0.76	3	Good
90-4	890	6	25	A	1558	+1.30	3	Good
91-1	A-33	6	25	A	1558	-0.28	4	Very good
91-2	A-33	6	25	A	1558	-0.41	4	Very good
91-3	A-33	6	25	A	1558	-0.08	4	Very good
91-4	A-33	6	25	A	1558	+0.53	4	Very good
92-3	Colloid 8440	-	25	-	1558	+2.99	2.5	Good
92-4	8440	-	25	-	1558	+9.87	2	Pitted-warped
92-5	8440	-	25	-	1558	+5.71	2	Porous-warped
92-8	8440	-	25	-	1558	+3.92	2	Porous-warped
93-1	Commercial	-	--	-	1558	+8.93	1.5	Decayed edges
93-2	control mix	-	--	-	1558	+6.80	1.5	Warped
93-3	" "	-	--	-	1558	+6.32	1.5	Slightly pitted
93-6	" "	-	--	-	1558	+4.99	1.5	Slightly pitted
94-1	Colloid 8440	-	33	A	1558	+2.95	1	One corner flaking
94-2	8440	-	33	A	1558	+4.71	1	Pitted-warped
94-3	8440	-	33	A	1558	+4.60	1.5	Some pitting
94-4	8440	-	33	A	1558	+3.51	1.5	Some pitting
95-1	CC Mix #2	-	--	-	1558	+1.77	2.5	Fair to good
95-2	" " "	-	--	-	1558	+13.12	0	Terrible-flaked
95-3	" " "	-	--	-	1558	+4.67	1	Warped
95-4	" " "	-	--	-	1558	+0.51	2.5	Fair to good
96-1	Colloid 8440	-	25	B	1558	+16.02	0	Warped-flaked
96-2	8440	-	25	B	1558	+16.84	0	Side chipped
96-3	8440	-	25	B	1558	+25.05	0	Side chipped
96-4	8440	-	25	B	1558	+12.07	0	Warped-chipped
97-1	Colloid 8440	-	33	B	1558	+3.13	3	Good
97-2	8440	-	33	B	1558	+5.60	2	Slight warp-pitting
97-3	8440	-	33	B	1558	+3.58	2.5	Good
97-5	8440	-	33	B	1558	+2.67	3	

FILLER COMPOSITIONS: A - 50% Asbury 7101, 40% Asbury 4015, 10% Asbury Micro 250
B - 11 parts Asbury A-99, 4 parts Asbury Micro 850

All coupons were molded using a two-step process at a constant pressure of 4000 psi, coupons were post cured at 450°F for 20 hours.

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