Lead-Acid Battery:

Electrochemical Compatibility of Plastics

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Because of the float use-mode of reserve batteries in the Bell System, the cell behavior is quite sensitive to small amounts of electroactive organic substances in the cell electrolyte. Premature cell failure has in the past resulted from the introduction of a new plastic without testing its compatibility with the cell electrochemistry. We describe an accelerated test method by which the extraction rate of organic residues from the plastic container into the cell electrolyte is predicted and the electrochemical effects of these residues is determined. These data predict that in normal service no adverse electrochemical effects from interaction of the electrolyte with the jar or cover will occur in the cylindrical lead-acid cell for a minimum of 50 years.

I. INTRODUCTION

1.1 Materials Selection

The general problem of materials selection for battery use is divided into two areas: (*i*) the test material must meet some specified physical property requirements and maintain them over a period of long exposure to battery electrolyte (usually a strong corrosive acid or base solution), and (*ii*) the material must not introduce any solute into the electrolyte which will upset the electrochemical behavior of the cell.

During the early history of lead-acid battery development, the cell was placed in a container made of a material which experience had shown resistant to the sulfuric acid. Since a wide range of polymeric materials was unavailable, glass and lead-lined containers predominated. During the 1930s, hard rubber containers for lead-acid cells were introduced. There was no compatibility testing of this material but, in retrospect, it can be seen that none was needed since:

(*i*) cell lifetimes were short due to failure mechanisms which were faster operating than cell contamination by the container material;

(*ii*) cells were generally cycled, which tended to decompose organic contaminants due to the high charge potentials;

(*iii*) a lead-antimony alloy was used in the grids and the antimony-caused negative overvoltage lowering was so large that it swamped the much smaller increases caused by organic contaminants. These cells required much higher float currents due to this shift and lead-calcium cells which float at much lower currents are preferred for Bell System use.

These characteristics masked the slow-appearing effects caused by organic contamination. With the advent of extremely long-lived, low float current lead-acid cells, organic contamination from container materials can measurably upset cell behavior.

1.2 *Material Testing*

In recent years various plastics have been used in these applications.[1] Styrene, styrene copolymers and rubber-filled styrene plastics have gained the broadest acceptance.[2] With the advent of large new families of plastics, testing for suitable mechanical properties and monitoring their degradation in the particular electrolyte have become commonplace.[3] The effects of the electrolyte on the properties of the plastic can be measured more rapidly. By increasing the test temperature or increasing the severity of the test conditions in other ways (mechanical stress, radiation, and so on).

The second area of material compatibility, which is the effect of the plastic on the battery, has not to our knowledge been rigorously investigated. Weight-change studies are routinely used to determine the amount of electrolyte attack on a polymeric material[4]; that is, if the loss of the plastic is small over some period of time, the material is assumed chemically compatible with the cell. The effect of the small fraction of "lost" material on the cell electrochemistry has not generally been investigated. For typical cyclic battery service in which the cell is routinely charged and discharged, the effects of these organic contaminants are usually small enough to be innocuous, since the cycling of the cell either promotes the oxidation of the contaminants or covers them up in recharge cycles. In the Bell System, however, batteries which serve as a back-up or reserve power supply are floated, that is, maintained in a fully charged state by a potential of 2.17 volts per cell.

In such a use mode, the effects from electrolyte contamination by organic substances can be very harmful to cell life and performance.

1.3 *Example of Electrochemical Incompatibility*

A history of incompatibility problems has existed in the Bell System. Some standard lead-acid cells with porous polyvinyl chloride (PVC) separators were placed in Bell System use between 1954 and 1965. These separators were cheaper and less brittle than the normally used microporous hard rubber units. All of these cells showed needlelike dendritic lead deposits on the negative plates after a few years of service. These dendrites shorted and discharged the cells, behavior which has since been duplicated in this laboratory in similar cells in which the processes were accelerated by elevated temperatures. The cause of this dendritic growth was traced to the PVC separators by the procedure of leaching separator samples with cell electrolyte and introducing this solution into stably-floating cells, which promptly grew needles. Since solutions leached from pure PVC polymer (resin without additives) show no electrochemical effects; the effects observed must have been due to the residual organic solvents and wetting agents used in fabrication of the separators. It was found that the balance of the polarization (discussed subsequently) was markedly disturbed in the doped cells. No further investigations were undertaken and the new separator material was withdrawn from Bell System use.

Two conclusions are drawn from this experience:

(*i*) New materials for fabrication of battery components must be verified as electrochemically compatible with the cell under use-mode conditions.

(*ii*) A test procedure must be devised in which the compatibility of new materials can be determined in a relatively short time.

Due to the undesirable properties inherent in the presently used styrene-acrylonitrile plastic,[5] a new, rigid PVC copolymer which shows a superior combination of physical properties has been recommended for the jar and cover of the new cylindrical cell. Because of the previous experience with PVC plastic, compatibility testing was mandatory.

There is a need for a test procedure which accelerates the plastic-cell electrochemistry interaction such that a valid choice of materials can be made in a relatively short time (< 12 months). The work reported here is an example of a general test method which we believe to have

broad application in material selection for use in batteries or other electrochemical devices.

II. THE FLOATING CELL

2.1 Description of Float

A battery used as a reserve power source in the Bell System is "floated" when not delivering emergency electrical power; that is, the cells are maintained at full charge by overcharging the cell at a constant voltage of 2.17 V., which is 110 mV above the thermodynamic reversible potential of the cell (Fig. 1). This results in operation in regions where the potentials on the positive and negative plates are largely fixed by the Tafel behavior of the electrodes (the "Tafel lines" on Fig. 1).[6]

2.2 Grid Corrosion and Plate Polarization Balance

It has been shown[7] that grid corrosion in the positive plate is minimized at potentials of 20–30 mV above the reversible potential of the $PbO_2/PbSO_4$ electrode and, to provide a sufficient margin of safety, up to 80 of the 110 mV excess potential should be on the positive plates of the cell, with the remaining 30 mV on the negative plates (Fig. 1).
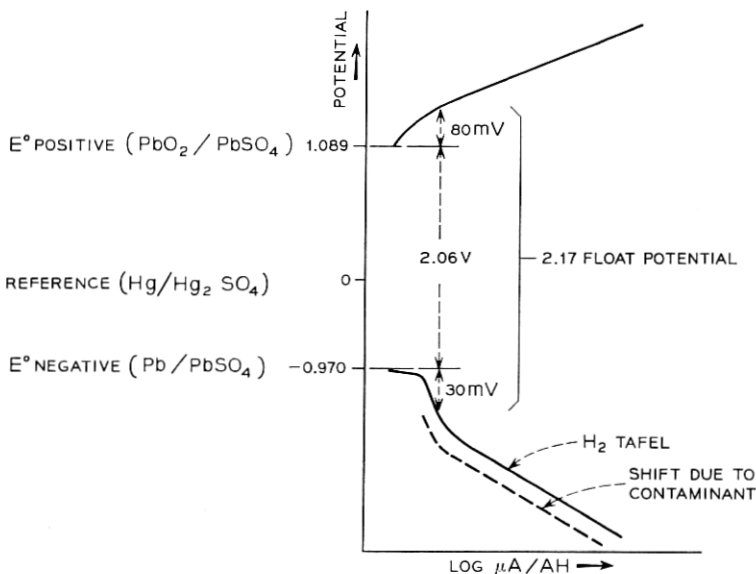

Fig. 1—Float properties of BTL cell, showing effect of contaminants.

The division of this excess potential is determined in large part by the slopes and intercepts of the hydrogen and oxygen Tafel lines[8] and a shift of one or both curves will change the fraction of the excess potential found on each plate. Since these cells must float at the lowest practicable float currents, the cells exist in a potential region where the negative polarization curve has an anomalously steep slope due to oxygen reduction.[8] Because of this, changes in the overpotentials of the cell will have significant effects in the float balance, and an increase in the hydrogen overpotential and/or a decrease in the oxygen overpotential will decrease the fraction of available excess potential on the positive plates of the cell. As the positive polarization drops significantly below 30 mV, the corrosion rate of the grid, which is the failure mechanism of the lead-acid cell on float,[9] increases rapidly, shortening the life expectancy. If the shift were sufficient to place the entire 110 mV on the negative plates, the positive electrodes would remain at their reversible thermodynamic potential and would self-discharge.

In addition to causing overpotential shifts, the organic residues can change the growth habit of the negative electrodes, again affecting cell performance. This can be beneficial in some cases, and organic "expanders" are used to produce a spongy, high surface area, efficient lead anode.[10] The effect can, however, be destructive as in the case of the dentritic structure caused by the PVC separators as described in Section 1.3.

III. ORGANIC CONTAMINANTS AND ELECTRODE BEHAVIOR

A very large number of organic solutes affect electrochemical processes. The role of surface-active organic substances in modifying electrode reactions was noted as far back as 1923[11]; extensive literature has been developed on this subject, though predominantly limited to mercury or platinum electrodes. The usual result of even minute amounts (a few ppm) of electroactive organic substances in an electrolyte is a marked shift in Tafel behavior, especially for hydrogen evolution. A shift of tens of millivolts on a 1 cm^2 electrode is typically caused by less than 10 ppm of an electro-active contaminant.[12] In general, electrode reactions are quite complex and a rigorous description of the mechanisms and identification of the reaction products and intermediate species is always difficult and often impossible. The inclusion of organic substances at the reaction interface complicates matters further; we therefore will not discuss the mechanisms of the organic molecule-electrode interaction but refer the reader to some of the extant literature.[13-18] It is

sufficient to note that organic contaminants in small amounts can affect Tafel behavior markedly and thus may present a problem for long-lived batteries.

IV. INITIAL TESTING

4.1 General Description

Most plastics consist of a mixture of various components. In addition to the polymer resin, stabilizers, antioxidants, lubricants, waxes, impact modifiers (usually another polymer species) and colorants are typically found in plastic formulations.[5] These complicate investigations, for any one or all of these components may introduce sufficient electroactive substances into the electrolyte to upset the float balance. The magnitude of the problem may be seen by noting that a plastic is usually considered resistant to chemical reagents (ASTM D 543) if the percent weight change in the given reagent is 0.1 percent or less over a four-week period under specified conditions and the weight change with time is described by a relatively flat curve.[3] That such a test is inadequate for the present problem is shown by the fact that a weight loss maximum of 0.1 percent from the cylindrical battery jar (normalized to the same test conditions as ASTM D 543) would introduce about 100 ppm organic contaminants into the cell electrolyte. By calculation, this amount of foreign material would cover the actual surface area of the positive and negative electrodes (determined by B.E.T. methods)[19] to a depth more than three monolayers.

To determine if the selected plastic might cause electrochemical problems, equal amounts of thinly-shaved samples of the test plastic,[5] a propylene-PVC copolymer, and the styrene acrylonitrile plastic now in use were subjected to standard sulfuric acid electrolyte (1.210 sp. gr.) for 28 days at 60 C in the apparatus shown in Fig. 2 (this apparatus was used for all subsequent extractions). 60°C was chosen as a temperature high enough to accelerate the chemical processes without getting too close to the glass transition temperature (T_g) of the plastic. (The reasons for this temperature limit are discussed in item ii of Section 5.1.)

Since a measurement of the effect of the test plastic relative to the presently used material was desired rather than an absolute value in this preliminary test, the electrochemical effects were grossly exaggerated by the use of a lead electrode of very small area in a rotating disk configuration.[20] This apparatus was used to measure the hydrogen and oxygen overpotential shifts in the test solutions compared to a pure acid solution. The test plastic caused a shift in the hydrogen over-
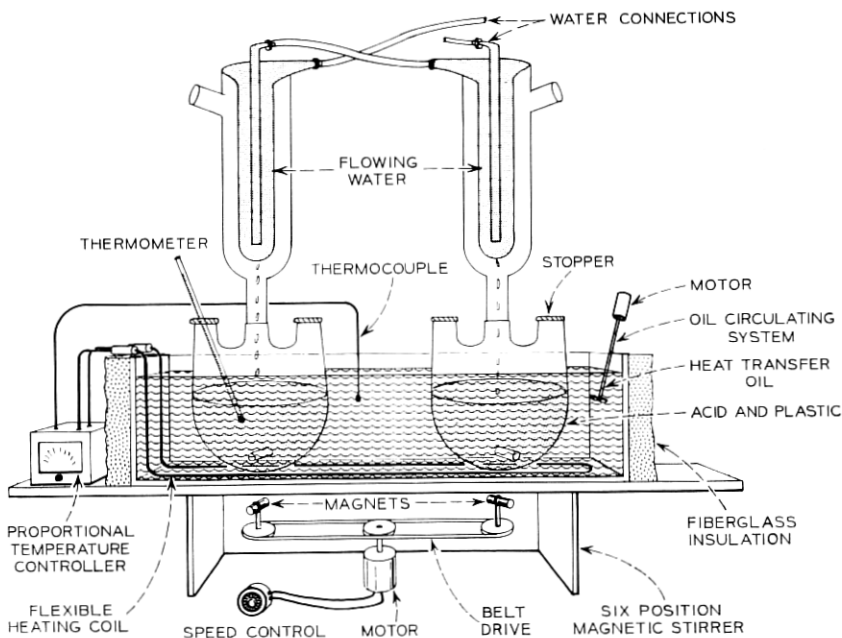
Fig. 2—Extraction apparatus.

potential of about 200 mV while the previously used styrene acrylonitrile caused a 50 mV shift. The oxygen overpotential was not measurably affected, and both solutions caused a slight increase in the lead corrosion current. Thus a source of possible cell deterioration was indicated.

4.2 Relative Effects of Plastic Components

To measure the relative effect of each formulation component independently, samples of the pure components (supplied by the manufacturer) were extracted separately at 60°C with the same volume of 1.210 sp. gr. sulfuric acid as used in the previous test. The amount of each component extracted was equal to the amount of that component found in the thinly shaved test plastic extracted in the previous experiment. As shown in Table I,[21] the individual overpotential shifts are apparently not additive (the mechanism may be competitive—the organic molecule which adheres most tightly to the lead excludes the other species from the main reaction mechanism). It is evident that the organo-tin stabilizer dissolution products produced the major electrochemical effects; in fact, the effects caused by the pure stabilizer were both qualitatively and quantitatively (on a relative scale) identical to

TABLE I[21]—ROTATING DISK ELECTRODE STUDY OF
PLASTICS AND COMPONENTS

Sample	Effects
PVC Co-Polymer	~ -200 mV H_2 overpot. shift slight increase in corrosion current
Styrene Acrylonitrile	~ -50 mV H_2 overpot. shift
PVC Base Resin	No effect
Stabilizer	~ -200 mV H_2 overpot. shift slight increase in corrosion current
Lubricant	~ -100 mV H_2 overpot. shift
Impact Modifier	~ -50 mV H_2 overpot. shift

the effects due to the entire plastic formulation. Therefore, the appearance of electroactive dissolution fragments of the plastic in the electrolyte was monitored by the measurement of the concentration of stabilizer fragments in the sulfuric acid.

This assumption that the electrochemical effect of the plastic leachants was directly related to the stabilizer decomposition was supported by obtaining samples of a plastic compound whose formulation was identical to the test material except for the substitution of a lead-based stabilizer for the normal organo-tin compound. A sample of this experimental plastic and a sample of the lead stabilizer were extracted independently as before, but the extracts both showed identically negligible electrochemical effects. This supports the assumption that the effects produced by the normal test material are effectively caused entirely by the organo-tin stabilizer.*

V. ACCELERATED TEST PROCEDURE

5.1 *Test Assumptions*

The test method evolved from the following facts and assumptions:

(i) The stabilizer fragment concentration in the acid will increase with time at a given temperature, probably by diffusion through

* This experimental lead-stabilized plastic, though electrochemically innocuous, is unsuitable for fabrication of battery jars since it is opaque and is also almost impossible to injection-mold. Small (0.19 g) plaques were molded only with great difficulty.

the plastic to the electrolyte interface. This process can be accelerated by increasing the test temperature. An apparent activation energy can be measured for the process.

(ii) So long as the test temperature does not closely approach the glass transition temperature of the plastic, the extraction processes occur by mechanisms identical with those operative at room temperature, which permits valid extrapolation of the accelerated reaction rates to room temperature.

(iii) The stabilizer was shown to be completely digested by the 1.210 sp. gr. sulfuric acid and the concentration of tin ions in the electrolyte is thus a precise measure of the amount of stabilizer decomposition.

(iv) It was shown that the appearance rate of tin ions is directly proportional to the ratio of the exposed plastic surface area per unit sulfuric acid volume used in the extraction. The leaching can be accelerated by increasing this ratio (Fig. 3). Other geometric effects were compensated for by using plastic samples of the same physical dimensions for all tests.

5.2 Test Procedure

In the test procedure, equivalent quantities of plastic (with equal surface areas) were extracted into equal volumes of standard 1.210 sp. gr. sulfuric acid at 40°, 50°, 60°, and 70°C for up to several weeks. (The time was dependent on the extraction temperature.) Small samples of
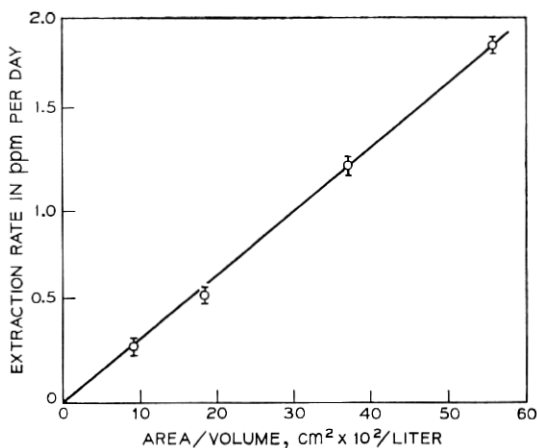

Fig. 3—Extraction rate dependence on plastic surface area/acid volume at 80°C.

acid were removed periodically and analyzed for tin ion.[22] The samples removed were a small enough fraction of the total acid volume so that the change in the plastic surface area/acid volume ratio was small enough that the change in reaction rate which resulted was within the error limits of the measurements.

5.3 Test Results

Resulting concentration data are plotted vs time (Fig. 4) and show linear behavior over the measured temperature range.

A high plastic surface area/acid volume ratio was used to speed up the extraction, and linear extrapolation out to the actual plastic surface area/acid volume ratio present in the BTL cell produced the upper set of time coordinates. A rate equation of the form
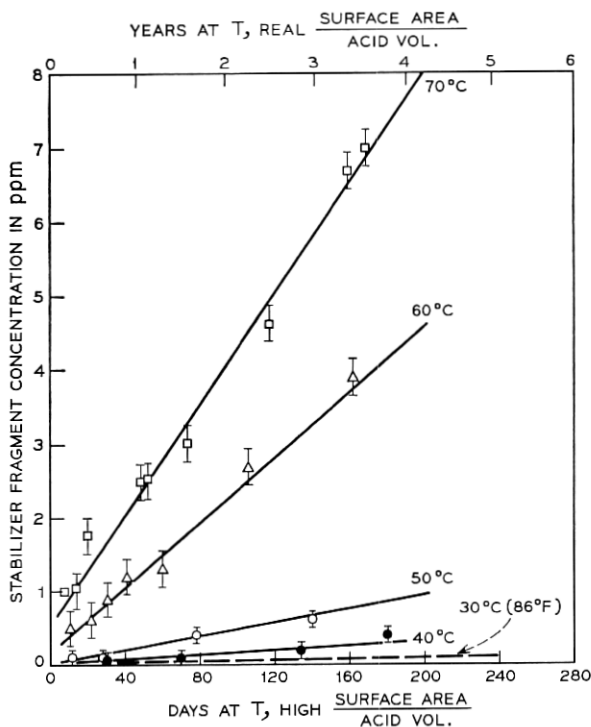
$$\frac{d[Sn]}{dt} = k$$
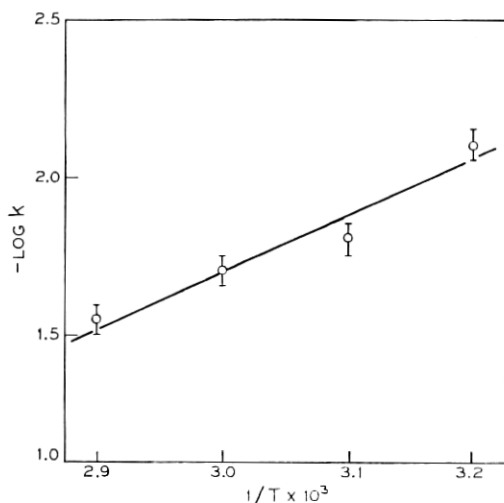

Fig. 4—Extraction rates of stabilizer.

Fig. 5—Arrhenius plot.

was assumed and treatment of these data by the Arrhenius method (Fig. 5) produced an apparent activation energy

$$E_{act} = 10 \pm 1 \text{ kcal/mole.}$$

The use of this value produces the extrapolated room temperature stabilizer decomposition rate described by the dashed line in Fig. 4.

5.4 Application to Real Cells

To determine the relationship between contaminant concentration and electrochemical effect in real cells, stably-floating cylindrical cells were doped with extract aliquots of a known stabilizer fragment concentration and the cell characteristics (electrode potentials, float currents and cell potential) were monitored. Four to six weeks were needed after each extract addition for the cell to restabilize. From these data a plot of hydrogen overpotential shift vs stabilizer fragment concentration in the cell was constructed. (Fig. 6)

An elevated temperature accelerated test was run on cells with large amounts of the test plastic immersed in the electrolyte. The conditions duplicated the testing of the experimental microporous PVC separators in which dendritic lead structures caused premature cell-death. No abnormal lead deposits or altered plate structures were noted in this test nor were any changes in float voltages or currents observed.[23]
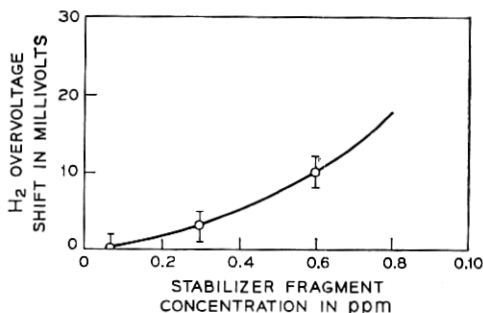
Fig. 6—Overvoltage shift vs stabilizer fragment concentration in BTL cell.

The appearance rate of the stabilizer decomposition products in the battery at room temperature is now known (Fig. 4) and we have established the relationship between the stabilizer fragment concentration and the shift in the hydrogen overvoltage (Fig. 6). We can therefore construct a plot showing the increase in the hydrogen overvoltage (Fig. 7) as a function of time at room temperature. All that remains is the establishment of a maximum allowable overpotential shift and the limit can be read directly from this plot.

Since an increase in the hydrogen overpotential of 50 mV will produce a disastrous increase in the corrosion rate of the positive grid, the allowable shift must be less than this value. It is considered that a maximum of 30 mV is a sufficiently conservative value. Figure 7 shows that even with a worst-case interpretation of the data, the lifetime limit imposed by the organo-tin stabilized test plastic is in excess of 50 years and thus we should not see electrochemical problems in Bell Telephone Laboratories-designed cells due to the plastic case for at least this period of time.

VI. DISCUSSION

Though the experimental work reported here was very specifically related to the immediate problem, we believe it to have wide applicability.

6.1 *Diffusion and Extraction*

Though no assumptions have been made concerning the mechanism of the extraction process and the work described is quite empirical, it is probable that the rate limiting step involves diffusion of the stabilizer
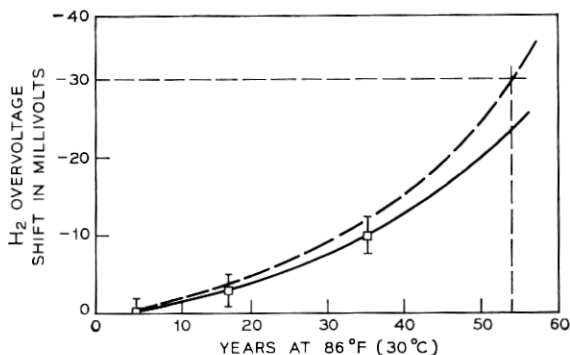
Fig. 7—Life limit of BTL cell due to test plastic.

to the electrolyte/plastic interface. There exists some supporting evidence. When thinly shaved samples of the test plastic were extracted in sulfuric acid, up to 80 percent of the available tin compound was extracted into the acid, but the physical dimensions of the sample shavings were unchanged, and there was little ($<$ 2 percent) weight loss. If the tin extraction were only due to dissolution of the plastic surface, about 80 percent of the sample would have had to dissolve to produce the measured tin concentration. Thus the stabilizer must have diffused through the polymer matrix to the acid. In addition the 10 ± 1 kcal/mole activation energy is comparable with the activation energy of other diffusing species in PVC as shown in Table II.[24,25] The fact that the

TABLE II[24,25]—DIFFUSION IN VARIOUS PLASTICS

Polymer	Diffusant	Activation Energy kcal/Mole
Rigid PVC	N_2	14.79
Rigid PVC	Ar	12.3
Rigid PVC	O_2	13.03
Rigid PVC	CO_2	15.44
Rigid PVC	CH_4	16.8
Rigid PVC	H_2O	9.98
Polyethylene	n-Octadecane	12.4
Polyethylene	DLTP	12.4
Polyethylene	N-ode	12.3
Polypropylene	N-ode	20.5
Polypropylene	DLTP	19.8
Polypropylene	Phenol A	22.3
Poly-4-methylpentene-1	DLTP	14.5
Poly-r-methylpentene-1	Phenol A	14.2

measured 10 ± 1 kcal activation energy is less than any of the values of Table II is comforting since extrapolation based on a low E_{act} will yield conservative time predictions.

6.2 *Future Applications*

If, in the future, methods are available for prediction of diffusion rates in plastics based on the average molecular weight, the physical structure of the polymer, the size and structure of the diffusing molecule and other such parameters, the problem of plastic material selection can be accomplished by *a priori* calculations rather than the time-consuming extraction described in this paper. In addition, plastics may in the future be used as dispensers of a desired additive, slowly releasing small amounts of the dopant which may, for example, be used to maintain a given float balance or retard a particular corrosion process.

Finally, the rotating disk electrode will probably gain importance as a tool for testing compatibility of various solutes in electrochemical devices. Since the electrodes appear to be at least as sensitive as most analytical methods for determination of low solute concentrations, it will probably be calibrated and used directly as an analytical tool, bypassing the time-consuming sampling and separate analytical procedures.

VII. CONCLUSIONS

7.1 *Life Limit of BTL Cell Due to Container Material*

It has been shown that even a "worst-case" treatment of the data results in a projected 50-year minimum life limit for the BTL cell with a container fabricated of the test plastic. This prediction is based on 86°F nominal ambient. If the cells are required to operate at higher temperatures, a corresponding life limit can be calculated by use of the measured 10 ± 1 kcal activation energy.

7.2 *Rotating Disk vs Real Cell Experiments*

The rotating disk provides a quick method for establishing relative levels of electrochemical effects due to a wide range of materials. An example of its utility was demonstrated in early field trials in which a polarization shift was seen in a small number of cells. This resulted in the positive plates floating at their reversible potential and the entire 110 mV excess potential was shifted to the negative plates—a condition that would result in self-discharge of the positive plates. By use of the rotating disk electrode apparatus as a screening tool, it was determined

that cyclohexanone, a solvent used in the jar-cover sealing procedure, was entering the cell electrolyte.

Potential scans through the hydrogen evolution region of the lead disk were made in (1.210 sp. gr.) H_2SO_4 solutions containing various amounts of cyclohexanone. The shift in hydrogen overvoltage (η_{H_2}) relative to uncontaminated acid solution was approximately 50 mV for electrolyte containing 10 ppm cyclohexanone, an amount which was shown to be easily leached from a freshly made cyclohexanone solvent-sealed joint into the battery electrolyte of a BTL cell. The overvoltage shift was toward more negative potentials, the same direction observed on negative plates in the field trial cells. The result of these experiments was the development of an alternate sealing procedure,[26] and the problem has not reappeared in subsequent field trials.

VIII. ACKNOWLEDGMENTS

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