

# High Rate Batteries

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## ABSTRACT

The capacity and efficiency of a battery increase rapidly as the porosity of the active materials in the plates is increased. At porosities of 85 to 90 percent, ampere-hour efficiencies of more than 50 percent were obtained in discharge times of as little as one minute over a range of temperatures extending down as low as  $-60^{\circ}\text{C}$ .

Metal plates of controlled porosities and high surface areas were produced by the electrolytic reduction of metal compounds subjected to controlled physical pressure during electrolysis. Anodic active materials were prepared in an analogous manner. The active materials thus prepared and tested included zinc, cadmium, lead,  $\text{Ag}_2\text{O}$  and  $\text{AgO}$ , all of which provided highly successful electrode materials. The results of tests on these materials indicate that the technique can be applied to many more active materials, thus pointing the way to the production of a wide range of extremely high-rate, low-temperature batteries.

## PROBLEM STATUS

This is a final report on one phase of the problem; work is continuing on other phases.

## AUTHORIZATION

NRL Problem C05-14  
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## HIGH RATE BATTERIES

### INTRODUCTION

Batteries are an excellent source of power for relatively short periods of time. When the discharge rate is increased, more power is being drawn from a battery, the discharge time becomes less, and the efficiency decreases. This decrease in efficiency becomes quite appreciable in most batteries as the discharge time is decreased from one hour to a few minutes. As the temperature is lowered the problem becomes greatly aggravated, and the efficiency of the battery becomes less even though the discharge rate is greatly reduced.

Many attempts have been made in the past to produce efficient low-temperature, high-rate batteries which can be discharged in a few minutes. These studies have generally been directed toward unusual battery systems which, in general, had undesirable characteristics such as extremely high self-discharge rates, which meant the electrolyte could not be added until the battery was about to be used.

In the work presented in this report, it is shown that the high-rate, low-temperature performance of a battery is dependent upon the physical nature of the active materials in the electrodes. Consequently, it should be possible to develop a number of new types of batteries and also to redesign existing battery systems so they can operate at high efficiencies in discharge times of a very few minutes, even at very low temperatures.

Theoretically, there are a very large number of substances that can be used as the active material in battery anodes or cathodes. From a practical viewpoint, this use is not feasible with a majority of these materials, since they polarize heavily and can operate only at very low current densities. Since polarization is a monotonic increasing function of the current density, all materials have some limit to the current density at which they may be discharged practically, beyond which limit the drop in the value of the potential is too large to be acceptable for practical purposes. Any physical factor that increases the surface area of the active material decreases the polarization by decreasing the current density, expressed in terms of discharge current per unit of surface area, and thus makes it feasible for the material to be discharged at much higher rates.

The authors have shown previously (1) that metal plates of controlled porosities can be produced by the electrolytic reduction of metal compounds which are subjected to controlled physical pressure during the electrolysis. The porosity of the metal plate is a function of the pressure applied during formation and is also dependent upon the particular compound or mixture of compounds being reduced. A wide range of porosities was obtained in this manner from metals of the normal electronegative group such as silver, bismuth, tin, lead, antimony, zinc, cadmium, copper, and mixtures of these, including mercury (1). The porosity,  $P$ , which is often expressed as a percent, is defined as that fraction of the total volume of the material that is occupied by pores or void spaces.

Since the surface area of a metal plate increases with increase in porosity, a number of zinc electrodes of various porosities were prepared and discharged in a AgO-Zn-KOH cell, in an effort to determine the effect of porosity on cell capacity. An excess of AgO was used to insure that the zinc electrode was the limiting factor on cell capacity. Each discharge was made at 20°C and 10 amperes, using 10 cc of 40 percent KOH electrolyte.

The results are shown in Fig. 1, where the ampere-hour efficiency and the cell capacity in minutes are plotted as a function of the electrode porosity. The efficiency increases from close to zero percent at 58 percent porosity, up to a maximum of slightly over 75 percent at 82 percent porosity. As the porosity is increased beyond this peak value, the metal structure becomes flimsier, and anodic solution causes high resistance paths and crumbling of the metal. As a result, the capacity and efficiency then decrease as the porosity increases.

This electrode was over 2 mm thick, exclusive of the grid. By using a thinner plate, which generally results in an improved efficiency, it was felt that an electrode could be constructed which could be discharged at much higher rates and still have high ampere-hour efficiencies for discharge times considerably less than the 15 minutes shown in Fig. 1. The information presented here is the result of an exploratory investigation designed to determine if such high-rate electrodes could be prepared from zinc and also from other active materials; to determine if porosity was a major factor affecting the capacity of such electrodes; and to determine some of the major factors affecting the design, formation, and operation of high-rate electrodes, thus opening the door to future investigations of a more highly detailed and quantitative nature.

#### HIGH-RATE ANODES

A cell or battery consists of components such as an anode, cathode, electrolyte, separators, grids, connectors, etc., all of which must be designed, assembled, and tested under appropriate conditions before the cell can be feasible for consideration in commercial competition with other power sources. The test results, shown here, are not meant to imply that a commercial cell is now available. However, as soon as a high-rate anode and cathode have been tested in a common electrolyte as shown here and found to have suitable characteristics, the major obstacle standing in the path of constructing a high-rate cell or battery has been eliminated.

Various metal anodes were tested by discharging them in a cell especially designed to determine their characteristics. The porous metal anode was suspended between cathodes containing an excess of moderately high rate AgO as the active material. Under

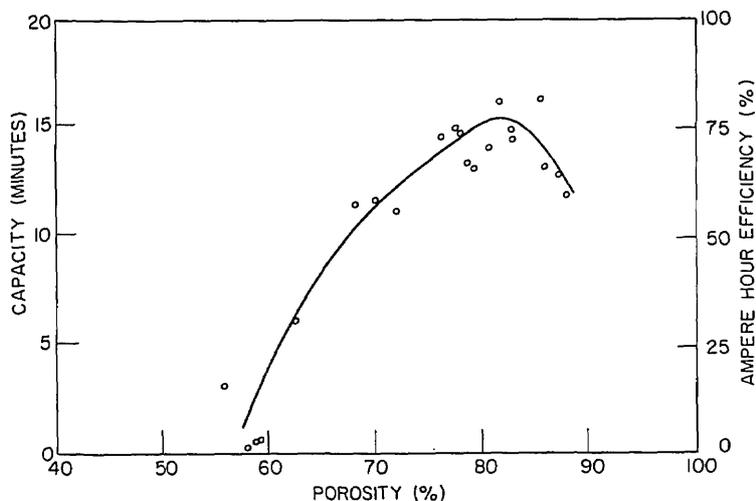


Fig. 1 - Effect of zinc electrode porosity on cell capacity and efficiency, 20 °C, 40% KOH, 730 ma/cm<sup>2</sup>

these conditions the metal anodes were always found to fail first. The effective spacing was 0.075 cm, which was large enough to prevent any physical contact between the electrodes and thus eliminated the need for a separator. The presence of a separator tends to decrease the cell potential and discharge time to a widely varying degree that depends on the physical properties of the separator and the discharge conditions for the cell. Thus, the data taken in the above manner represent an optimum as far as separators are concerned and presents a standard against which the use of various separators can be tested in a specific, commercial cell. This electrode assembly was packed in a snug-fitting cell case and filled with just enough KOH electrolyte to keep the electrodes covered throughout the discharge. The excess of electrolyte in this assembly over what would be present in a close-packed commercial cell using separators did not appear to have any effect on the measured electrode characteristics.

A number of porous zinc electrodes of various porosities were formed by the electrolytic reduction of pasted ZnO and ZnCO<sub>3</sub>, or mixtures of ZnO and ZnCO<sub>3</sub> under controlled physical pressures in 15 percent KOH electrolyte (1). Porosities as high as 92 percent were obtained by reducing ZnCO<sub>3</sub> under the comparatively light pressure of 0.054 kg/cm<sup>2</sup>. A number of discharges were made at constant current covering a range of porosities, current densities, and electrode thicknesses. The results are shown in Fig. 2, where the ampere-hour efficiency is plotted as a function of the electrode porosity for various current densities and amounts of zinc. In each case, the efficiency and consequently the cell capacity decreases rapidly with decreasing porosity. In curve D, discharges are being obtained in slightly over one minute at efficiencies up to 60 percent. Figure 2 shows, as would be expected, that an increase in current density at a given porosity decreases efficiency (curve C versus curve D). It also shows that a given efficiency can be maintained, within limits, by increasing the porosity as the current density increases. The current density is defined here in milliamperes per square centimeter, one square centimeter being defined as the area of an electrode one centimeter square. It would seem more logical to define current density as an active material current density

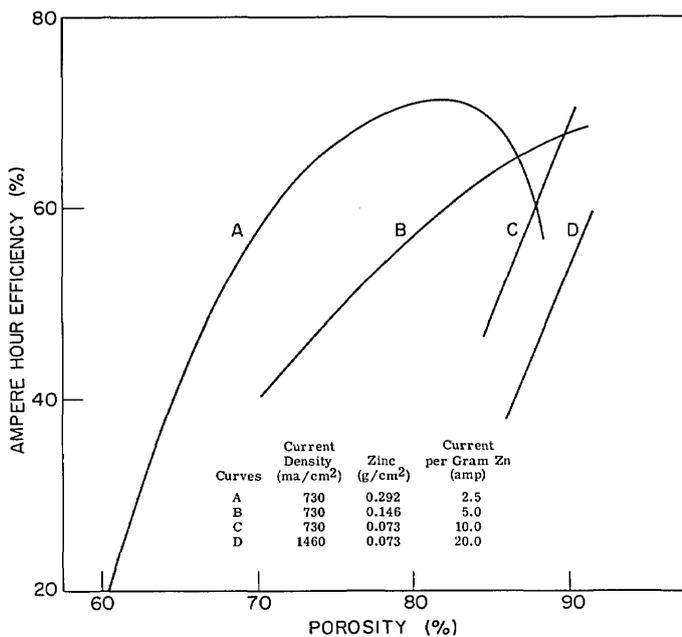


Fig. 2 - Effect of porosities on efficiencies of zinc electrodes at 20 °C

measured in amperes per gram of zinc, since this value would be inversely proportional to the theoretical discharge time at 100 percent efficiency. Consequently the capacity of an electrode, at a given porosity and active material current density, should be fairly constant but would tend to decrease some as the thickness of the electrode increased.

The fact that ampere-hour efficiencies of as much as 60 percent were obtained on cells discharging in as little as one minute indicated that a further study should be made in this extremely high-rate area. A number of cells were evaluated by means of constant-resistance discharges. The average potential and the cutoff potential, as well as the capacity, are difficult to evaluate in a constant-resistance discharge, since the potential tends to drop slowly with time (Fig. 3). This drop is in sharp contrast with the constant-current discharges of Fig. 7, where the potential is fairly constant throughout most of the discharge and drops off rapidly near the end. However, many uses of a cell approximate constant-current-discharge conditions.

Occasionally in other work a cell has been observed, particularly at low temperatures, which was incapable of discharging through any type of load. Once started by extraneous means, a cell of this type would sometimes be capable of discharging on its own with satisfactory characteristics. If such a cell were discharged by the usual method of forcing an extraneous constant current through it and recording the potential, an initial period would be observed where the potential would be less than zero. If this period were too short to be detected by the recording equipment, it would never be known that the cell was incapable of practical operation without being started by an externally applied potential. The constant-resistance discharges showed that this problem did not occur with these high-porosity electrodes at high rates.

A large number of highly porous zinc electrodes of varying porosity were discharged at a number of high rates at temperatures ranging from 23°C to -60°C. A typical discharge is shown in Fig. 3 where the potential, the time, and the current density at 23°C have been plotted as a percentage of the total zinc that has been consumed at any given time. A discharge current equivalent to almost 2000 ma/cm<sup>2</sup> is maintained for nearly a minute at better than 50 percent efficiency. The discharge curve would have been much flatter in the case of a constant-current discharge.

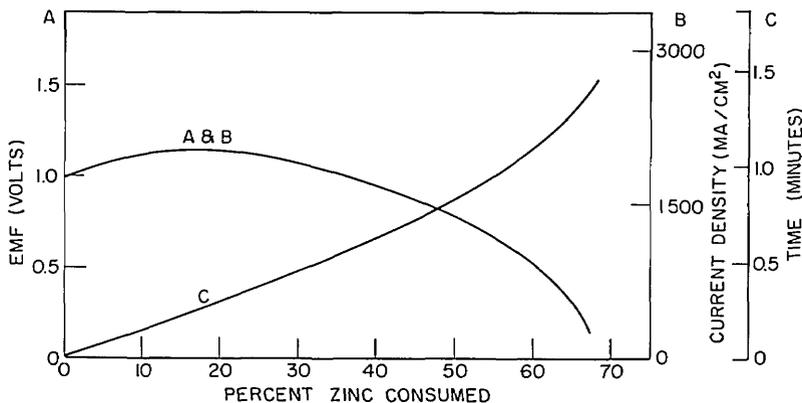


Fig. 3 - Effects of constant-resistance discharge on high-rate zinc electrode at 23°C, AgO-Zn-32% KOH, 0.0577 g/cm<sup>2</sup> 91.6% porous zinc; curve A, potential; curve B, current density; curve C, time

A number of porous zinc electrodes were successfully discharged at high rates at  $-60^{\circ}\text{C}$ . To test their durability, one of them was short circuited for six seconds, followed by an immediate discharge through a constant resistance. The results (Fig. 4) indicate that the short circuit had no apparent effect on the cell characteristics.

The extremely high rates and efficiencies involved here can best be appreciated by comparison with existing types of electrodes. In Fig. 5, curve A, estimated values of the limiting current densities at various temperatures for the high-porosity, high-rate electrode, are compared with those of a conventional electrode, curve B, recently developed for commercial high-rate usage. The high-porosity electrode has limiting current densities one to two orders of magnitude higher than the conventional electrode.

In Fig. 6, where the potential is a function of the constant-current discharge time, a higher efficiency and better characteristics were obtained for the high-rate zinc electrode, which is discharged in about one minute, than were obtained for two commercial cells, which were discharged in about 20 minutes. Commercial cells with higher efficiencies than these may be available, but these were the only two in which the weights of the active materials were known.

If the high surface area of the high-porosity zinc electrode is the chief reason for its ability to discharge with high efficiencies at high rates, then it should be possible to form high-rate electrodes from other metals. High-porosity cadmium electrodes were prepared by the electrolytic reduction of cadmium oxalate in 15 percent KOH electrolyte. They were discharged against AgO cathodes in 32 percent KOH at constant-current densities up to  $1460\text{ ma/cm}^2$ . The results are shown in Fig. 7, where the cell EMF is plotted against time throughout the discharge. High efficiencies were obtained, even in discharges that lasted as little as one minute. (Excellent results were also obtained at low temperatures.) High-porosity lead electrodes were prepared by the electrolytic reduction of lead cyanamid and lead thiocyanate in 15 percent KOH electrolyte. These were discharged against AgO cathodes in a 32 percent KOH electrolyte at constant current densities up to  $1100\text{ ma/cm}^2$  and discharge times of as little as one minute. The results are shown in Fig. 8.

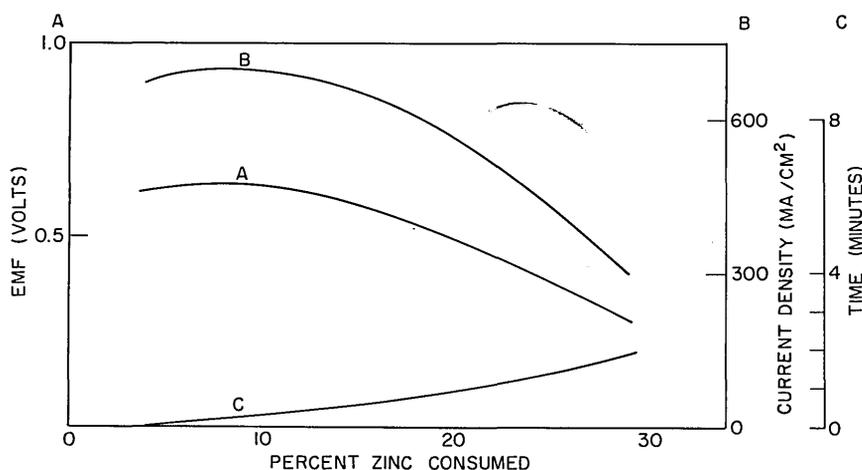


Fig. 4 - Effects of constant-resistance discharge on high-rate zinc electrode at  $-60^{\circ}\text{C}$ , AgO-Zn-32% KOH,  $0.070\text{ g/cm}^2$  91.4% porous zinc; curve A, potential; curve B, current density; curve C, time

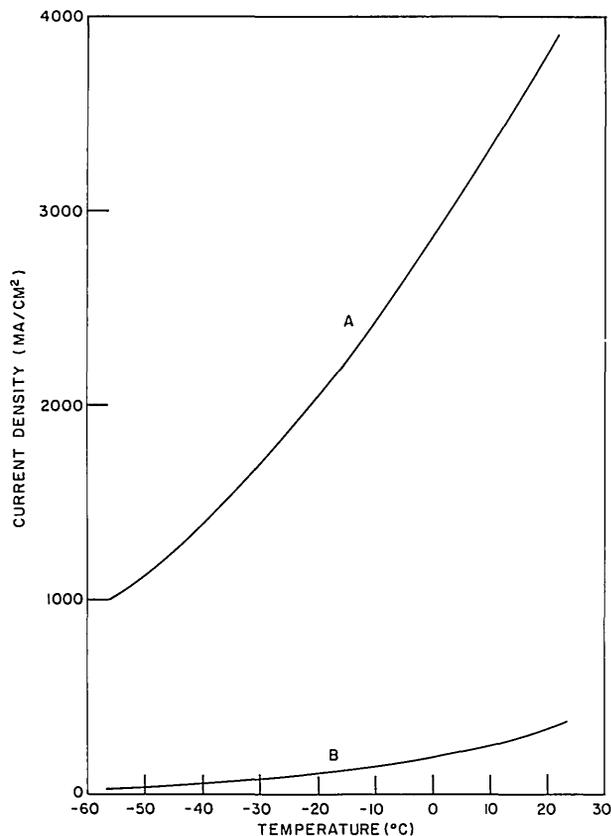


Fig. 5 - Approximate limiting current density for high-rate (curve A) and conventional (curve B) electrodes

Although a plot of efficiency versus porosity should be expected to show a smooth curve of negative slope, it can be seen in Figs. 7 and 8 that the values of the efficiencies are somewhat scattered. These values of the efficiency are erratic apparently for four reasons. First, the electrode thickness varies slightly in the case of cadmium, and in the case of lead it varies quite appreciably in a manner that would be expected to oppose the effect of the variation of the current density on the efficiency. Second, a satisfactory method has not yet been devised for assigning a value to the cutoff voltage, which in turn determines and may seriously affect the value of the calculated efficiency. Third, the reproducibility of discharges made in time intervals of less than four minutes is not very good, particularly in view of the fact that not enough is yet known about the various factors that affect cell characteristics to be sure that a given discharge is under statistical control. Fourth, there is a large increase in temperature, during a high-rate discharge, which can seriously affect cell capacity and other characteristics, particularly at low ambient temperatures, where the change in capacity with change in temperature is relatively large. This increase in temperature, which measured as much as 30°C, can vary widely with changes in cell design and the operating conditions.

The high capacities and efficiencies obtained at extremely high discharge rates from the zinc, cadmium, and lead, high-porosity electrodes indicate that other high-porosity, active materials should also make excellent high-rate electrodes.

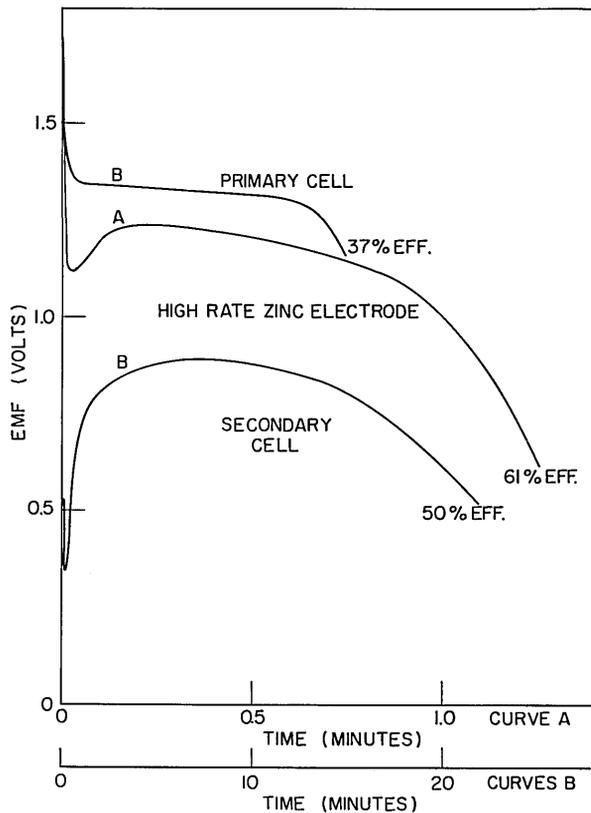


Fig. 6 - Comparison of high-rate zinc electrode battery (curve A) with conventional silver-zinc batteries (curves B)

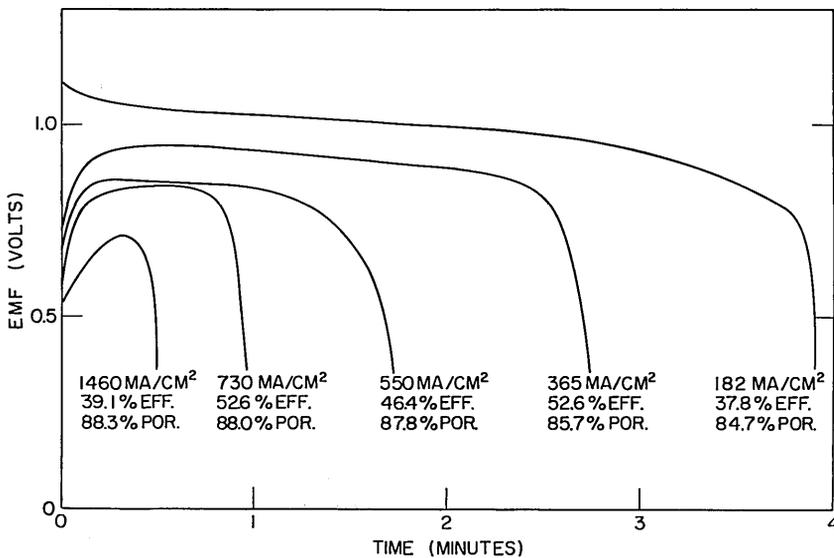


Fig. 7 - Effects of constant-current discharge on high-rate cadmium electrodes at 20 °C, AgO-Cd-32% KOH

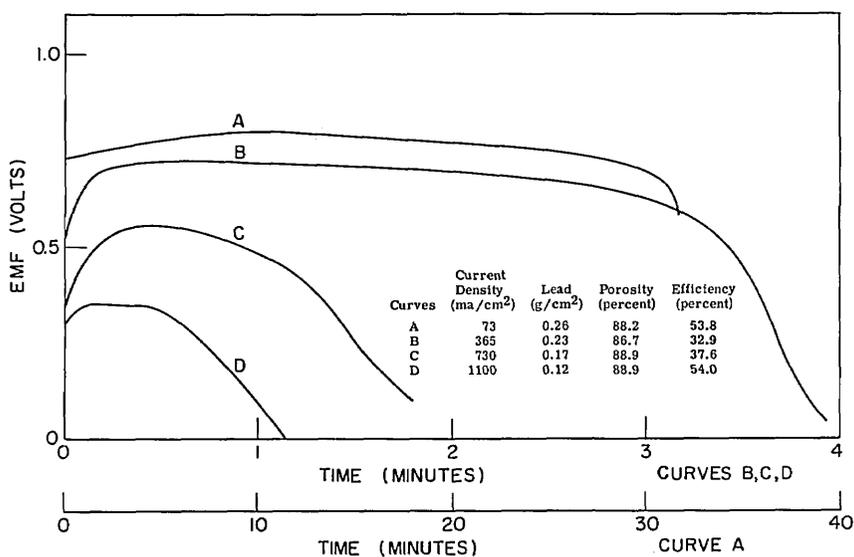


Fig. 8 - Effects of constant-current discharge on high-rate lead electrodes at 20°C, AgO-Pb-32% KOH

## ELECTRODE PASTING

There are a large number of problems involved in the pasting of the grids for the formation of the high-porosity electrodes (2). Since the paste is itself subjected to physical pressure during reduction, it cannot be put in the pockets of a commercial grid but must be set on top of a wire screen. Under these conditions the pasted material must be quite stiff in order to avoid flowing. This type of pasting is not particularly easy and is to a certain extent an art (2). An ideal paste gives on reduction a porous metal plate having optimum properties in regard to such characteristics as strength and cohesiveness. This ideal paste is stiff and is almost dry, is slightly flexible and doughy, is not crumbly or sticky, and is cohesive and will flow to a certain extent when placed under pressure. The percentage of liquid present in a particular powder-liquid combination is either constant or fixed within narrow limits. There is an extremely wide variation in the pasting properties of various compounds as defined in terms of ease of pasting and quality of paste. It was possible to divide all the compounds tested into five arbitrary groups ranging from "very good" to "very poor." This grouping was found to be a function of particle size. In Fig. 9 the average water content is shown as a function of the average of the geometric mean diameter for each of the paste groups. The finest size of particles makes the best pastes and should be used wherever possible.

Since the water content of the paste is critical, slight variations in the paste composition might be expected to have an effect on the electrode porosity. The porosity of the metal obtained by electrolytic reduction of a pasted electrode is shown in Fig. 10 to be a function of the amount of zinc per unit area. The porosity decreases as the amount of zinc increases, which is equivalent to saying that the porosity decreases as the thickness of the electrode increases. Similar results were obtained with lead. Earlier measurements showed the effect of electrode thickness on porosity to be negligible at low porosities. Results are shown in Fig. 10 for two different physical pressures applied throughout the reduction and show, as has been done previously (1), that the porosity decreases as the formation pressure is increased. Each point in Fig. 10 represents an average of several measurements. A comparison shown here of ideally pasted electrodes with electrodes that were pasted slightly on the dry side indicate that the latter give a slightly lower porosity.

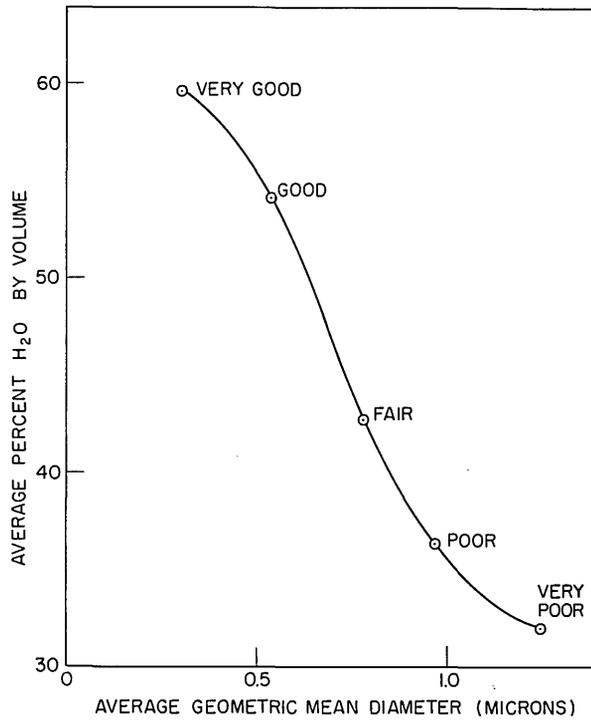


Fig. 9 - Variation of liquid content of paste with particle size for various paste groups. Designations on the curve represent evaluations of paste quality.

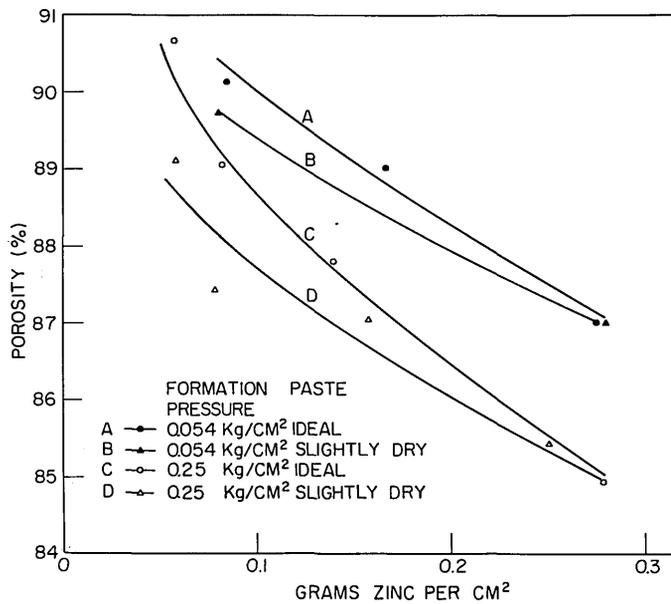


Fig. 10 - Variation of porosity with the amount of zinc per unit area

The pasted electrode is assembled between two flat inert electrodes which are wrapped in a layer of porous cellophane or some other suitable, thin-sheet separator material. Pressure is applied during the reduction to metal. The reduction tends to start at the outer edges of the pasted electrode and moves toward the center. Occasionally, there will be an approximately circular portion in the center of the electrode which does not readily reduce. This is essentially a problem in current-density distribution, which can be remedied by painting the outer edges of the inert electrodes with stop-off lacquer, thus leaving a bare circular portion of appropriate size in the center of the electrode.

The control of metal porosity by the use of controlled physical pressure during reduction presents a number of problems which are now fairly well understood. This method has been successful in preparing porous metals from the normal electronegative group previously mentioned but has not been successful with the transition metals. It has been shown that the porosity of the metal plate is seriously affected by the choice of metal compound being reduced (1). If it is assumed that the volume of the porous metal plate at the end of the reduction at zero pressure equals the volume of the paste at the start, then the weight of metal per cubic centimeter is

$$(1 - P)\rho = (1 - p)dP_m \quad (1)$$

where  $P$  is the porosity expressed as a fraction,  $\rho$  is the metal density,  $p$  is the ratio of the volume of liquid in the paste to the volume of paste,  $d$  is the density of the compound being reduced, and  $P_m$  is the percentage of metal in this compound. Equation (1) can be rewritten in the form

$$1 - P = (1 - p)M_c \quad (2)$$

where  $M_c = dP_m/\rho =$  volume of metal contained in a cubic centimeter of the compound.

A plot of  $1 - P$  vs  $M_c$  should give a straight line with the slope  $(1 - p)$ . Values of the porosity,  $P$ , at zero formation pressures were obtained by extrapolation from porosity-pressure curves and were plotted in Fig. 11 against volume of metal per cubic centimeter of compound for various compounds of zinc, lead, cadmium, silver, tin, copper, antimony, bismuth, and several mixtures of some of these compounds, including mercury. The straight line fitted to this data has a slope of 0.48, which corresponds to a liquid content of 52 percent in the paste, which is about what would be indicated by the data in Fig. 9. There is an appreciable scatter in the data of Fig. 11 due to the fact that each ideally pasted compound has its own particular value of  $p$ . In addition, the relationship is probably affected by such factors as the solubility of the compound in the electrolyte and its particle-size distribution and shape factor. The two zinc compounds fall well below the line and indicate that the slope may also be dependent on the particular metal being formed. The data for the lead compounds alone have been plotted in Fig. 12; the scatter, though definite, is much less than in Fig. 11. Porosities of lead from less than 70 percent up to 94 percent are shown here, and any desired porosity in this range could be obtained by using a single compound or a suitable mixture of two or more of these compounds. All of these compounds were reduced to the porous metal in a 15 percent KOH electrolyte. The same metal porosity was obtained for Pb formed from PbO in 25 percent  $H_2SO_4$  as in 15 percent KOH. It seems likely that the choice of electrolyte will have little effect on the metal porosity as long as the compound will reduce satisfactorily in that electrolyte. The very high porosities are obtained from compounds having a low value of  $M_c$ . Organic compounds containing a low percentage of metal tend to have low densities and consequently a low value of  $M_c$ . Not all compounds will reduce to the metal by electrolytic reduction under pressure. As a rough rule those compounds that are soluble in the electrolyte or are moderately good conductors of electricity tend to reduce. The best chance of obtaining very high porosities appears to be by the use of a metal salt of a high-molecular-weight organic acid.

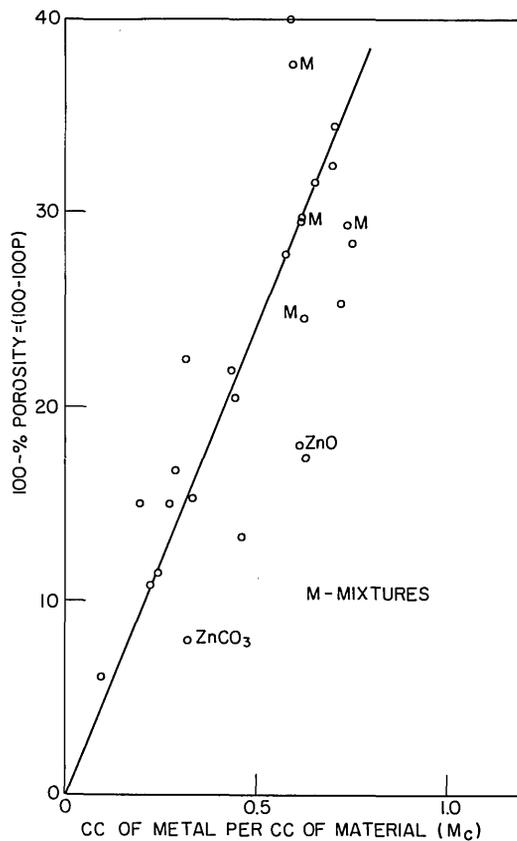


Fig. 11 - Effect of volume of metal per cubic centimeter of material on porosity

If the use of pressure during the electrolytic reduction could be eliminated, the paste could be applied into the pockets of a conventional grid, thus eliminating most of the problems involved in pasting on top of a wire screen grid to form high-porosity electrodes. A careful selection of metal compounds or mixtures thereof obviates the use of pressure to control metal porosity. However, a light pressure during reduction may be necessary in many cases in order to obtain optimum physical properties of the reduced metal electrode. It seems probable that a light pressure could be applied successfully to a pasted conventional grid, particularly if a slight excess of paste were used.

It is now possible to produce a metal electrode at any given porosity from a wide range of metals, and it seems probable that future work will show the feasibility of producing such an electrode on a conventional grid using little or no pressure during reduction.

#### HIGH-RATE CATHODES

If a high-rate battery is to be constructed, a high-rate cathode is needed in addition to the high-rate metal anodes. The problems involved in preparing high-porosity cathodes from such materials as  $AgO$  and  $PbO_2$  are similar to those involved in producing high-rate metal anodes, but appear to be much more difficult, possibly because they have received less study. The major problem involved is the difficulty of getting good adherence between

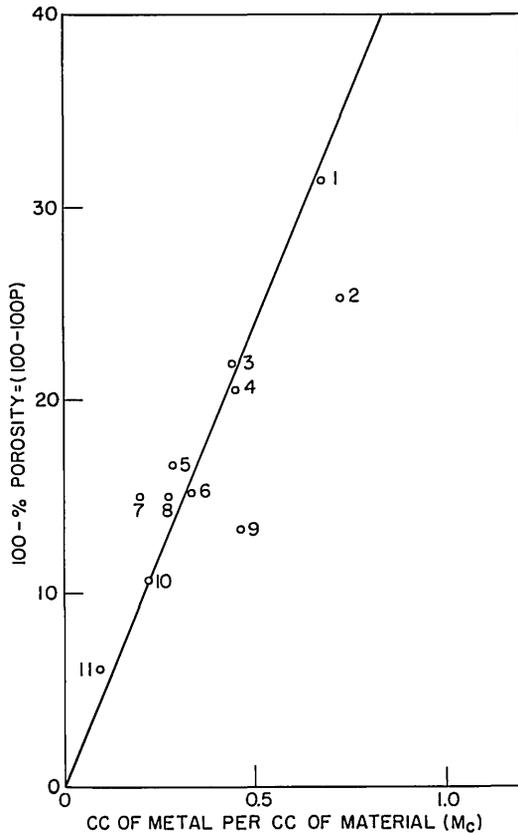


Fig. 12 - Effect of volume of lead per cubic centimeter of compound on porosity

1. Oxide ( $\text{PbO}$ )
2. Oxide ( $\text{Pb}_3\text{O}_4$ )
3. Maleate (tribasic)
4. Carbonate
5. Formate
6. Oxalate
7. Chlorphthalosilicate
8. Cyanate
9. Cyanamide
10. Thiocyanate
11. Stearate

the active material and the grid. Although an  $\text{Ag}_2\text{O}$  electrode can be easily produced that operates at very high efficiencies at extremely high rates, a study was started on the  $\text{AgO}$  electrode, since it is theoretically capable of producing almost double the capacity of the  $\text{Ag}_2\text{O}$  electrode in terms of ampere hours per unit weight.

$\text{AgO}$  electrodes were prepared by oxidation of various silver compounds pasted on a silver screen grid. Cathodes containing up to 87 percent porous  $\text{AgO}$  were formed in this manner. A cell constructed of highly porous zinc and  $\text{AgO}$  electrodes was discharged at  $-40^\circ\text{C}$ . The results are shown in Fig. 13, where the cell potential in volts, the current density, and the time are plotted as a function of the percent of  $\text{AgO}$  consumed throughout the discharge.

A 71-percent porous  $\text{PbO}_2$  electrode was prepared which was capable of discharging at moderately high rates. Both the  $\text{AgO}$ , the  $\text{Ag}_2\text{O}$ , and the  $\text{PbO}_2$  electrodes could be recycled.

The data indicate that the maximum efficiency on an efficiency-porosity curve, such as that shown in Fig. 1, may be obtained at about 92 percent porosity at very high rates. At this porosity there is 11-1/2 cc of electrolyte contained in the electrode for each cubic centimeter of Zn, giving a total weight of active material plus electrolyte of about 27.6 grams. At 84 percent porosity there is 5-1/4 cc of electrolyte per cubic centimeter of Zn, giving a total of about 14.1 grams. Thus, a drop in porosity that might be expected to decrease the capacity about 25 percent gives a decrease in the electrode weight of about 35 percent. Obviously the porosity that gives a peak capacity does not give a minimum weight. A 92 or 94 percent porous electrode is fairly flimsy. Rough calculations indicate that the maximum optimum porosity ever needed will be about 85 or 86 percent. Such an

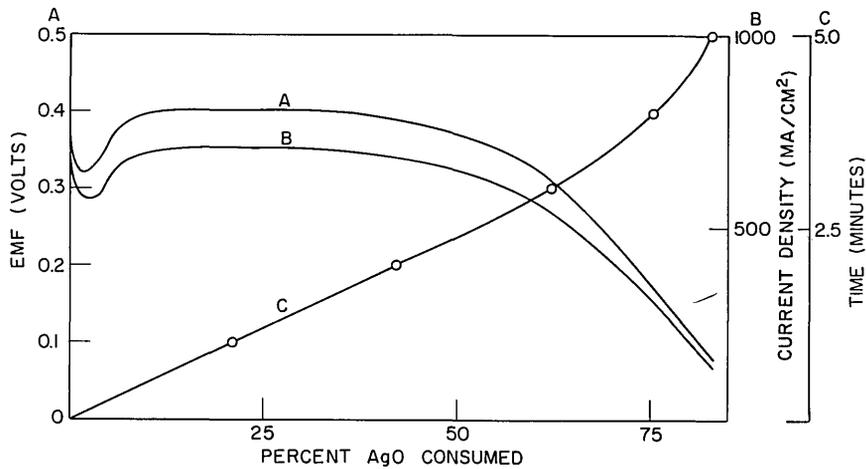


Fig. 13 - Effects of constant-current discharge on high-rate AgO electrode at  $-40^{\circ}\text{C}$ , AgO-Zn-32% KOH,  $0.117\text{ g/cm}^2$  86.9% porous AgO

electrode, prepared on top of a wire screen, has fairly good physical strength which would be even better if a conventional grid were used. High-porosity metal plates can be produced by other methods, such as felt fiber or metallurgical techniques. Whether such electrodes will give the capacities obtained here probably depends on their pore-size distribution and surface area. The successful reduction of a metal compound by the methods described here probably depends on the establishment of a network of relatively low-resistance electrolytic paths filled with electrolyte and leading to virtually all the available active material. Since they must be there for formation to be possible, they will also be there for the purposes of discharge.

## CONCLUSIONS

The results of this exploratory investigation show that high-rate anodes and cathodes can be prepared that are capable of operating at high ampere-hour efficiencies at temperatures down to as low as  $-60^{\circ}\text{C}$ , thus pointing the way to the possibility of eventually producing commercial cells capable of discharging with high efficiencies at extremely high rates. What may be of greater importance is the fact that this work shows that the porosity of the active material is a major factor of utmost importance in determining the capacity and other characteristics of many if not all cells which have porous electrodes.

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13. ABSTRACT  The capacity and efficiency of a battery increase rapidly as the porosity of the active materials in the plates is increased. At porosities of 85 to 90 percent, ampere-hour efficiencies of more than 50 percent were obtained in discharge times of as little as one minute over a range of temperatures extending down as low as -60°C.  Metal plates of controlled porosities and high surface areas were produced by the electrolytic reduction of metal compounds subjected to controlled physical pressure during electrolysis. Anodic active materials were prepared in an analogous manner. The active materials thus prepared and tested included zinc, cadmium, lead, Ag <sub>2</sub> O and AgO, all of which provided highly successful electrode materials. The results of tests on these materials indicate that the technique can be applied to many more active materials, thus pointing the way to the production of a wide range of extremely high-rate, low-temperature batteries.		

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Batteries and components Rate of discharge High efficiency High discharge rate Low temperature electrodes High electrode capacity						

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