PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A
MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

The Nickel-Zinc Battery for Electric Vehicle [and Discussion]

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The nickel–zinc battery for electric vehicles

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The paper first deals briefly with the justification for the present world-wide interest in electric vehicles, and summarizes progress made in this field over the last decade. It uses particularly the work carried out at Lucas to illustrate the state of development and current performance of such vehicles. It then emphasizes the fact that battery performance and life are the limiting factors restricting the increased use of these vehicles. A brief review of possible contenders from among a wide range of electrochemical systems is given, pointing out the major advantages and disadvantages of each. The reasons for preferring the nickel–zinc couple at this time are dealt with.

The characteristics of the nickel–zinc system are described in some detail. An outline of the current research and development programme at the Lucas Research Centre is given, with emphasis on its major aims and objectives, and some of the achievements against these objectives are described. Reference is made to the chief remaining problems. Some of the commercial prospects of a successful battery are stated.

1. Introduction

A major reason for current work on advanced battery systems is the great interest in electric vehicles. The first electric vehicle was produced more than 140 years ago, but the advent of cheap petrol led to the eclipse of the electric vehicle by the internal combustion engined vehicle.

The electric vehicles that survived advanced only slowly in technology over a 50-year period, and took the form of slow simple milk floats and delivery vans. However, the economics of cheap oil, which once almost destroyed the electric vehicle, have obviously now changed dramatically. Recent problems of oil supply, the rapid and continuing rise in oil prices since 1973, and growing political instability in the Middle East, which contains the bulk of the world's oil deposits, combined to focus attention on the uncertainty surrounding the running costs of the internal combustion engined vehicle, and the overall need to reduce the demand for liquid fuels. Projections of the growing gap between oil supply and demand for a number of industrialized countries show that the most dramatic is that for the U.S.A. The situation in the U.K. will be similar, but will lag behind that of the U.S.A. by about 10 years because of North Sea oil. Consequently, there will have to be a steady increase in the use of coal and nuclear power to fill the increasing energy gap.

Considering the question of fuel economy of electric vehicles, it is probable that an electric car employing lead–acid batteries would use rather more primary energy than a similar petrol-engined vehicle. However, the use of modern drive-train engineering and advanced batteries, allied with electricity generation techniques such as combined heat and power sources or fluidized-bed combustion, could mean significantly increased efficiency for electric transport (Agarwal 1980; Mueller & Wouk 1980*a*, *b*). When the comparison is made with liquid fuel from coal, the advantage for electric vehicles could be as high as a factor of two (Collins 1979).

In addition to arguments based on efficiency of primary energy use and overall availability of fuel to justify interest in electric vehicles, there are important arguments concerning electrical generating capacity. Electricity generating plants, and in particular the increasing proportion of plants that will be accounted for by nuclear power, run best under steady operating conditions. This means there will be a night-time peak of spare power, and this is precisely when most of the electricity required to recharge electric vehicle batteries would be required.

Table 1. Comparison of electric vehicles

	milk float type	modern Lucas van
battery	tubular lead–acid, 20 W h/kg	advanced flat plate, 40 W h/kg
controller	carbon pile resistance and contractors	thyristor chopper
motor	d.c. series wound	d.c. shunt field wound
top speed	24 km/h	89 km/h
range (urban driving)	40 km	113 km
range (steady 48 km/h)	56 km	233 km
vehicle gross mass	4 t	3.5 t
payload	1.5 t	1.0 t

Table 2. Lucas/Bedford delivery van

urban driving range	113 km
vehicle gross mass	3.5 t
payload	1.0 t
battery mass	1.0 t

Battery mass 28.6 % of gross mass; battery mass equal to payload.

The Electricity Boards can thus provide this energy without significant capital investment in new generating plants (Select Committee on Science and Technology 1980). As a result of such considerations, the amount of research and development work on electric vehicles has increased rapidly during the last decade.

To summarize the progress made in design and performance of electric vehicles over the period, this is best illustrated by a 'before and after' description of an older vehicle compared with today's best. The traditional milk float used a tubular lead–acid battery with an energy density of perhaps 20 W h/kg, an inefficient carbon-resistor controller and a costly d.c. motor. A typical performance would be a top speed of 24 km/h, a range of 40 km and a payload of 1.5 t.

Today's advanced electric vehicle, as illustrated in table 1, uses an advanced lead–acid battery of 35–40 W h/kg, an electronic controller and a highly efficient drive train, none of which obtrude into the payload space.

While many families have a 'second car' that normally covers only a few miles per day, such a vehicle is required occasionally to cover a much greater mileage at short notice. A Lucas hybrid vehicle at present under development has a top speed of 113 km/h, and a virtually unlimited range.

However, battery performance is by far the greatest remaining problem. Table 2 shows the proportion of vehicle gross mass occupied by an optimized high-performance lead–acid battery, and which nevertheless leaves much room for improvement as regards power : mass ratio. Additionally, such a battery, with a power : mass ratio of 38–40 W h/kg, has a cycle life (80%

depth of discharge (d.o.d.)) of about 300 cycles, which represents an in-service life of only about 34000 km, implying several battery changes over the chassis life of the vehicle.

Considerations such as these lead to the conclusion that an improved battery system is essential to enable electric vehicles to gain increasing commercial acceptance.

2. COMPARISON OF POSSIBLE BATTERY SYSTEMS

An intensive literature survey was carried out in 1978 to compare the properties of a wide range of electrochemical couples, and to assess their potential as an advanced battery for electric vehicle purposes.

Tables 3 and 4 summarize some of the parameters of the systems analysed, and, from a careful consideration of such factors, the nickel oxide–zinc couple was chosen as the system most likely to succeed, when both commercial and technical considerations were taken into account.

TABLE 3. ELECTROCHEMICAL SYSTEMS INVESTIGATED

existing batteries (*ambient temperature*)
- lead–acid, traction
- lead–acid, flat plate
- nickel–cadmium
- nickel–iron

proposed batteries (*high temperature*)
- sodium–sulphur
- sodium–$SbCl_3$
- lithium–FeS_x
- lithium–chlorine

proposed batteries (*ambient temperature*)
- nickel–zinc
- silver–zinc
- iron–air
- zinc–air
- zinc–chlorine
- zinc–bromine
- nickel–hydrogen
- lithium–organic electrolyte
- —TiS_2

TABLE 4. PARAMETERS CONSIDERED

- electrolyte type
- open circuit voltage
- operating voltage, 2 h rate
- operating temperature range
- overall energy efficiency
- energy density (W h/kg)
- energy density (W h/dm^3)
- peak power density
- maximum sustained power density
- sealed/unsealed
- self-discharge rate
- state of charge indication
- deep cycling life
- normal in-vehicle life
- recharge time
- cost (£/kW h)
- cost (£/kW h per cycle)

TABLE 5. CHARACTERISTICS OF ELECTRIC VEHICLE BATTERIES

	lead–acid traction	nickel–iron	nickel–zinc	sodium–sulphur
operating voltage	1.9	1.2	1.6	1.65–1.35
operating temperature range/°C	−20 to 50	10–50	−30 to +40	300–400
overall energy efficiency (%)	80	<60	75	70–75
energy density (1 h rate)/(W h/kg)	20	40	70	120
energy density (1 h rate)/(W h/dm^3)	65	100	140	170
maximum sustained power density/(W/kg)	20	220	200	120
self discharge time to 50% (days)	200	75	200	n.a.
deep cycles (80% d.o.d.)	1500	2000	300–400	thousands
cost/(£/kW h)	50	65	*ca.* 50	*ca.* 50

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To demonstrate more clearly the relative positions of such systems, an abbreviated comparison is shown in table 5, whereby the well known lead–acid couple, and also the sodium–sulphur couple are used for comparison.

Table 6 lists the main advantages and disadvantages of the nickel–zinc system.

Table 6

advantages	disadvantages
good gravimetric energy density	currently low cycle life
good volumetric energy density	increased self-discharge at high temperatures
high power output	cost of nickel
stable discharge voltage profile	
good low temperature performance	
low self-discharge at room temperature	
possible sealed construction	
aqueous electrolyte	

Some advantages summarized in table 6 can be considered.

(*a*) *Gravimetric energy density*. At 70 W h/kg at the 1 h rate, the nickel–zinc battery is about 3.5 times better than the lead–acid traction battery, and two-thirds that of the sodium–sulphur battery.

(*b*) *Volumetric energy density*. At 130–140 W h/dm^3 at the 1 h rate, that of the nickel–zinc battery is more than twice that of the lead–acid traction battery, and nearly equal to that of the sodium–sulphur battery.

(*c*) *High power output*. With a peak power density of 400 W/kg and a maximum sustained power density of 150–200 W/kg, the nickel–zinc battery is particularly good in this respect, giving retention of energy capacity at high discharge rates. It is about five times better than the lead–acid traction battery in this respect, and nearly twice as good as the sodium–sulphur battery.

(*d*) *Stable voltage profile during discharge*. The nickel–zinc battery has a reasonably flat voltage profile over its full capacity range with a sharp end-of-discharge fall. In this respect it shows a distinct advantage over many other systems, particularly the sodium–sulphur or the lithium–sulphur battery, where the voltage decreases by 0.3 V between the 60% discharged and the fully discharged state.

(*e*) *Good low-temperature performance* (*to* −30 °C). The nickel–zinc battery is particularly good as regards low-temperature performance at −30 °C, whereas lead–acid and especially nickel–iron batteries suffer a significant reduction in performance in such conditions. Of course, the sodium–sulphur battery must be maintained at about 350 °C for optimum performance.

(*f*) *Low self-discharge*. Nickel–zinc batteries, in common with modern lead–acid and nickel–cadmium batteries, undergo a very low rate of self-discharge. The time to 50% self-discharge is about 200 days for these ambient temperature batteries, and this is perfectly acceptable for electric vehicle application. The sodium–sulphur battery exhibits virtually no self-discharge.

(*g*) *Sealed construction*. There appears to be a good possibility of developing a sealed system, which would lead to low maintenance requirements.

3. Characteristics of the nickel oxide–zinc battery

The battery in the charged state consists of a nickel oxide cathode and a metallic zinc anode. The electrode reactions may be written as shown in table 7.

Table 7. Cell and electrode reactions

cathode	+0.49 V	$NiOOH + H_2O + e^- \longrightarrow Ni(OH)_2 + OH^-$ 3.42 g/A h
anode	−1.2 V	$Zn + 2OH^- \longrightarrow Zn(OH)_2 + 2e^-$ 1.22 g/A h (followed by $Zn(OH)_2 \longrightarrow ZnO + H_2O$)
overall	1.73 V/(326 W h/kg)	$2NiOOH + Zn + 2H_2O \longrightarrow 2Ni(OH)_2 + Zn(OH)_2$

Note: although NiOOH is the generally accepted formula for positive active material, there is considerable variation in stoichiometry and the average electron number required for reduction is often greater than 1.

Electrode construction

(*a*) *Nickel electrode*

Two types of electrode are being studied, a sintered and a pressed electrode. The sintered electrode consists of a matrix of metallic nickel, formed by sintering nickel powder into a coherent plaque and then electrochemically impregnating with nickel hydroxide. The pressed nickel electrode is constructed by pressing a mixture of nickel, a conductive diluent, typically graphite, and a binder on to a current collector. The sintered electrode has the advantage of long cycle life, but high cost, while the pressed nickel electrode has lower cost but also lower cycle life.

(*b*) *Zinc electrode*

The zinc electrode consists of a finely porous matrix of metallic zinc on a current collector, fabricated by pressing a zinc oxide paste onto a metallic grid and electrochemically converting to metallic zinc.

(*c*) *Separator material*

An absorbent layer is placed against the zinc electrode to maintain good electrolytic contact over the electrode surface. Next a microporous separator is employed to prevent the growth of dendritic zinc crystals, which would short-circuit the cells. Cellulosics such as cellophane and, more recently, ‘Celgard’, a microporous polypropylene sheet, have been the materials most commonly used, although a variety of substrates are under investigation including polyvinylacetate (PVA) and nylon as well as polypropylene. These are often grafted, either by chemical or radioactive means, or coated to improve performance characteristics such as wettability and stability under strongly basic conditions.

An absorbent layer is also employed against the positive plate to maintain good electrolytic contact. A secondary aim is to protect sensitive materials, such as cellulosics, from highly oxidative conditions. Materials commonly used include non-woven felts of polyamide or polypropylene fibres.

(*d*) *Electrolyte*

The electrolyte is typically aqueous potassium hydroxide of 30–40 % concentration by mass.

4. WORK PROGRAMME

The work programme is aimed at achieving the performance from the nickel–zinc battery outlined in table 5 as well as a cycle life in service to 80% d.o.d. of 500 cycles, and a manufacturing cost that will be commercially acceptable. This would provide an electric vehicle with a top speed of about 110 km/h, an excellent acceleration, and a range of about 400 km at a constant 65 km/h.

To date, the results are as indicated in the 'actual' column of table 8.

TABLE 8. WORK PROGRAMME: ACHIEVEMENT AGAINST FORECAST

	May 1980	May 1982	actual
sintered nickel electrode			
capacity/(A h/g)	0.14	0.18	0.16
pressed nickel electrode			
capacity/(A h/g)	0.17	0.19	0.20
small cells (2 A h)			
cycle life	1000	2000	1200–1500
large cells (250 A h)			
cycle life	200–300	600–800	100
energy density/(W h/kg)	> 70	> 80	> 70
full vehicle pack			
cycle life	—	200 (500 in 1984)	—

Since the battery is cathode-limited, the power/mass and power/volume targets have therefore been achieved. However, this still leaves unsolved the major problem of cycle life.

Modes of failure

Most cell failures are due to deterioration of the zinc electrode and separator system. Three phenomena are involved.

(i) *Inter-electrode short circuits due to zinc dendrites.* This is due (*a*) to the preferred manner of growth of zinc nuclei, and (*b*) to favourable mass transfer to the tips of growing dendrites in the concentration gradients set up on charging zinc electrodes.

(ii) *Shape change effects* ('*slumping*') *of zinc electrodes.* This is a movement of active material from the edges to the centre of the electrode during cycling and can cause shorting and capacity loss. It is due to (*a*) variations in mass transfer rates and higher effective current densities at the edges compared with the centre of the electrodes, and (*b*) electro-osmotic pumping effects.

(iii) *Capacity loss due to other factors.* This is manifested as a gradual capacity loss even when the previously mentioned shape change is absent. It is due to a general thickening of the electrode, crystal growth and passivation effects.

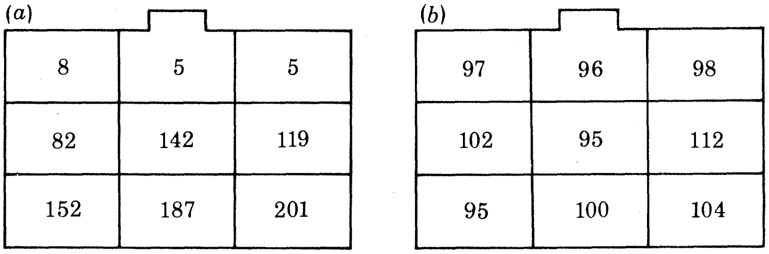
Possible solutions to zinc electrode problems

Owing to improvements in separator systems, dendrite penetration is not as serious as previously expected. Modern separators, together with careful control of the charging régime, seem well on the way to solving dendrite problems. As well as actually restraining dendrite growth, some separators contain nickel or other materials that, by producing local cell action with dendrites, corrode away the growing tips.

The use of excess zinc electrode material (as ZnO) is helpful in reducing shape change effects, as also is the use of foil (as opposed to mesh) current collectors.

Organic binders such as PTFE hold the zinc active material together and reduce shape change and densification effects.

Metallic additives (Hg, Pb, Cd, Sn compounds) can be used to reduce both dendrite growth and shape change (crystal growth effects). Synergistic effects are important with certain classes of these additives.



(*a*)

8	5	5
82	142	119
152	187	201

(*b*)

97	96	98
102	95	112
95	100	104

FIGURE 1. Change of shape of an electrode after 300 cycles to 70 % d.o.d. in 10 A h cathode-limited cells: (*a*)early electrode; (*b*) recent electrode with additives. The figures in the electrode segments are the percentage of the original segment mass remaining.

E.S.B. Inc. in the U.S.A. (following work at A.G.A. in Sweden) have used a radically different zinc electrode system. The substrate is a specially contoured plate that is vibrated in the vertical plane at a frequency of 10–100 Hz by a rotating shaft and eccentric system. The microturbulence produced in the electrolyte at the electrode surface is claimed to give an even, compact deposit free from dendrite and shape change effects for over 1000 cycles. The disadvantages are mechanical complication and excess electroyte, which reduce energy/mass and energy/volume ratios quite significantly.

The progress made on the shape-change phenomenon is illustrated in figure 1, where the degree of change of shape in an improved electrode is compared with that of an early electrode.

Nickel electrodes

Numbers of these have been tested to several thousand cycles. The sintered type is particularly good in this respect, although more costly. The cheaper composite types also appear to be capable of giving adequate cycle life.

Costs

The question of battery costs cannot be fully assessed until solutions to some of the technical problems become clearer. However, a careful study has been made of probable production methods, production costs, capital investment needs, component supplies and other factors.

At an annual production level of 10^5 50 kW h batteries, the estimated installed cost, based on currently available technology and components, is £50–60/kW h, which compares favourably with current volume-produced lead–acid batteries. It is possible that cost reductions could be obtained as further improvements are made in the technology. Moreover, no allowance has been made for the significant materials-recycling capability of this battery, which would help to reduce the overall production cost and also cushion the effect of possible future increases in basic raw material prices. Although at first sight a cost of £50–60/kW h may seem low for a battery based on nickel, it should be borne in mind that only 3.2 kg of nickel (sintered electrode) or 1.7 kg of nickel (pressed electrode) is used per kilowatt hour compared with 17.5 kg of lead per kilowatt hour for a lead–acid battery.

Analysis of the cost of the comparable battery systems indicates that a 500 cycle, £50/kW h nickel–zinc battery is only marginally more expensive than a possible sodium–sulphur alternative.

Consideration has been given to the probable availability of nickel and zinc to meet a major battery programme. Making reasonable assumptions about battery life, use for electric traction, power appliances and commercial applications, the degree of market penetration and so forth up to the year 2015, no problems are foreseen as regards the ready availability of either nickel or zinc, particularly since deposits of these materials are widely distributed and there seems little prospect of cartel action.

Table 9. Batteries for land transport in the E.E.C. (Jensen *et al.* 1979)

(Projected annual market in millions of pounds sterling.)

year	SLI	electric vehicle
1977	600	0
2000	860	1000
2025	1200	4500

5. Market prospects

A major market for a successful nickel–zinc battery would be as a traction power source for commercial vehicles and electric cars. However, this would be by no means the only market available. Such a battery could capture a large share of the automotive market currently held by lead–acid batteries. An assessment of the size of the E.E.C. market for these applications is given in table 9. Further substantial markets would exist in power storage (uninterruptible supplies for telecommunications and computer applications, estimated as currently worth £25M annually approximately in the U.K.) and industrial traction (estimated as worth £30M annually sales in U.K. currently). Much of the market at present held by nickel–cadmium batteries would be available, and the lower cost should open up many new markets for secondary batteries. Likely applications include power tools and domestic appliances (televisions, radios, tape recorders) as well as specialized uses such as military hardware and marine technology. Four areas of consumer durables will be considered in some detail, namely calculators, portable power tools, portable electronic devices, and portable domestic devices.

The situation concerning calculators is interesting, with annual sales in the U.K. of over 7×10^6, of which more than 10^6 are rechargeable. The move towards more complex units encourages this trend, and a cheaper secondary battery would accelerate the growth of this market.

The field of portable power tools is open to exploitation by a cheap battery with improved power density, and such a battery would be expected to capture a large share of the current 1.5×10^6 units annual market. Export opportunities for use in portable power tools in developing countries would appear to be very great.

In portable electronic appliances, the potential would be very good, and could lead to growing use for televisions, radios and cassette recorders, where the application of batteries is currently restrained because of the relatively high cost.

The domestic appliance sector, for applications such as hair dryers, shavers, fans, etc., is not generally a promising one for rechargeable batteries.

For consumer durable applications, it seems that, provided the sealing problems can be solved and an acceptable cycle life achieved, very substantial markets would be available for batteries from the 1 A h size upwards.

Overall, the potential market for a nickel–zinc battery of adequate cycle life, in the fields of electric vehicles, industrial applications, power storage and consumer durables, is very large and would have excellent growth prospects.

6. SUMMARY

Although several major improvements are still required in the operating characteristics of the nickel–zinc battery, nevertheless it is still the most likely candidate to succeed in the medium term, particularly for electric road-vehicle applications. Among the features in its favour are good performance at room temperature, use of a simple aqueous electrolyte, relatively straightforward fabrication techniques already well understood in the battery industry, and its improved power/mass and power/volume capabilities. If cycle life can be brought up to an acceptable level, the prospects for wide-scale commercial application look extremely promising.

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Discussion

D. S. FLETT (*Warren Spring Laboratory, Stevenage, U.K.*). I was very impressed with the progress on the nickel–zinc battery for electric vehicles as described by Dr Reasbeck. It would seem that we might expect commercialization within a decade or so. Quite large quantities of nickel and zinc will then be required. Current estimates of world reserves of nickel and zinc show dynamic resource lives of 200–403 years and 100–200 years respectively (Nutting 1977). However, just as in the lead–acid battery business it is clearly environmentally unacceptable and bad housekeeping to discard failed batteries, and thus batteries are recycled through secondary lead smelters, so will consideration have to be given to recycling old nickel–zinc batteries? Technology will have to be assembled and capital made available. I should like to know if Dr Reasbeck has given any thought to the recycling problem and what conclusions he has come to.

Reference

Nutting, J. 1977 Presidential address. London: The Metals Society.

P. REASBECK. The problem of recycling spent nickel–zinc batteries has been considered in detail, particularly in America (the reader is directed to the 1978 Cost and Design Studies published by E.R.C., Gould, Eagle Picher and Yardney).

No great problems are foreseen in reclaiming both the nickel and zinc contents in acceptably high yields (over 90%), although the cost is liable to be somewhat higher than with present lead recovery from spent batteries. Careful thought is, however, having to be given to minor components sometimes used in nickel–zinc batteries, such as mercury and iron, which can present difficult separation or health hazard problems.

Lucas are currently involved with a major company with extensive interests in the recycling of spent lead–acid batteries to evaluate more fully the necessary extractive processes.

Leaving aside the environmental problems mentioned by Dr Flett, there are no reasons apart from cost saving for recycling spent nickel and zinc until the annual level of production of battery packs is in the millions. Resources will be adequate for present levels of production.

J. B. Goodenough (*Inorganic Chemistry Laboratory, University of Oxford, U.K.*). Are Dr Reabeck's pressed $NiO_{1-x}(OH)_{1+x}$ electrodes particle hydrates? When he sinters his $NiO_{1-x}(OH)_{1+x}$ electrodes he presumably retains a porous electrode. Does he have an estimate of the change in electrode surface area on sintering? How does that effect the proton concentration in the interparticle solution on rehydrating?

P. Reasbeck. Analysis of the nickel hydroxide used in pressed nickel electrodes indicates an approximate formula of $Ni(OH)_2 \cdot \frac{1}{4}H_2O$. This is somewhat dependent upon the drying route employed. However, there is some evidence that further drying of the nickel hydroxide leads to a product with less electrochemical activity. As for the degree of hydration of the oxidized form NiOOH or the hydration of $Ni(OH)_2$ on cycling, not enough is known to comment.

Lucas–Harwell sintered nickel electrodes are made by sintering nickel powder (usually INCO255) in a reducing atmosphere and then electrochemically impregnating the resulting porous structure (plaque porosity is between 80 and 85%). From work with various nickel powders there seems to be little direct relation between the surface area of the initial powder and the finished plaque for a given strength of sinter. However, one parameter critical to obtaining a high utilization of the active material (130–140% corresponding to a 1.3–1.4 electron change) is the pore size distribution of the porous plaque.