

Heat management in aluminium/air batteries: sources of heat

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Abstract

One of the problems with the aluminium/air battery is the generation of heat, during both idle and discharge periods. The main sources of heat are: (i) corrosion of the aluminium anode during the idle period; (ii) inefficient, or less efficient, dissolution of anode during discharge; (iii) Joule heat during discharge, and (iv) non-uniform mass transfer during both discharge and idle periods. These components of heat act in a cumulative way because they are all interconnected. This paper addresses the basic reasons for the origin of these sources of heat. Suitable and practical remedial measures for the effective removal of such heat in the aluminium/air battery are suggested.

Introduction

The aluminium/air battery is a candidate power source for electric vehicles [1–4]. It is also a possible alternative energy source to oil generators whenever an uninterrupted power supply is a prime requisite [5].

The overall reaction in an aluminium/air battery takes place between aluminium, oxygen and water and yields hydrated aluminium oxide as the final product [6], i.e.:



This reaction is exothermic in nature with simultaneous liberation of electrical power. If the heat evolved could be restricted to a very low level, then the energy density and power density of the aluminium/air battery would be very high. This is best done by applying an appropriate 'heat management method'. The actual approach consists in understanding the science and engineering of heat evolution and dissipation in this battery system. The work reported here is directed towards the development of a suitable heat management strategy for the aluminium/air system.

Nature of problems associated with heat management in aluminium/air batteries

The main problem is to discover the origin of the heat evolved in the aluminium/air battery during both idle time and discharge. A second problem is the collection of data in a systematic and scientific study of the individual sources of heat. After

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identifying the sources of heat, remedial measures have to be tried systematically by conducting both long-term and short-term tests. The remedy, however, should be a permanent solution to the problem.

Different aspects of heat evolution in aluminium/air batteries

The following are the four different types of studies on heat evolution in aluminium/air batteries:

- (i) gravimetric and thermometric studies;
- (ii) open-circuit potential measurements;
- (iii) anodic or cathodic galvanostatic polarization, and
- (iv) Joule heat during cell discharge.

Gravimetric and thermometric studies on corrosion of aluminium in alkaline media

Corrosion of the anode material in an aluminium/air battery is a major problem during idle periods. This results in the production of heat and, hence, the development of a marked rise in temperature. Thus, corrosion is the foremost among the different sources of heat liberation in aluminium/air batteries; this portion of heat can be represented by H_1 [7].

Experimental

In order to compare the gravimetric and thermometric methods the following experiments have been carried out. Thermometry involves the measurement of the temperature of the solution (i.e., battery electrolyte) at regular intervals from the moment of introduction of the specimen, whereas the gravimetric method measures the weight loss. In the tests reported here, the specimens were suspended freely and totally immersed in the test solution. The duration of the experiments was 60 min. Other details of experiments were the same as reported in ref. 7. The temperature of the solution was monitored at 15-min intervals.

Results

In Table 1, weight-loss and thermometric data for different grades of aluminium (namely, 57S, 2S and 3S) are compared both in the presence and the absence of an inhibitor (namely, ZnO) in 4 M NaOH solution. It is found that 57S aluminium is the best material with respect to both the rate of temperature rise and the corrosion rate.

From the data in Table 2, it is understood that super-pure aluminium is better than even 57S aluminium with regard to the rates of corrosion and temperature rise, both in the presence and the absence of inhibitor (ZnO) in 4 M NaOH solution. But the super-purity specimens coated with zincate conversion coating are found to be inferior in performance than the zincate-coated 57S. This difference in behaviour may be due to better adhesion of the zincate coating on the 57S aluminium than on super-pure aluminium. With the latter, the formation of Al_2O_3 is faster and more uniform and, thereby, may prevent the formation of an adherent zincate coating.

Table 2 also provides the corrosion behaviour of CECRI quaternary alloy (contains Pb, In and Ga) based on 99.8% pure aluminium (supplied by NALCO, Orissa) in 4 M NaOH solution in the presence and the absence of the ZnO inhibitor. Interestingly,

TABLE 1
Corrosion rate and temperature rise for different anodes

Specimen	Medium	Corrosion rate ($\text{mg cm}^{-2} \text{min}^{-1}$)	Temperature rise ($^{\circ}\text{C}$)
57S	4 M NaOH	0.6923	20
26S	4 M NaOH	0.7147	20
2S	4 M NaOH	0.6809	16
3S	4 M NaOH	1.0640	24
Zincated 57S	4 M NaOH	0.3585	12
Zincated 26S	4 M NaOH	0.4213	13
Zincated 2S	4 M NaOH	0.4350	12
Zincated 3S	4 M NaOH	1.0419	31
Zincated 57S	4 M NaOH	0.1426	1
Zincated 26S	4 M NaOH + 0.6 M ZnO	0.2213	4
Zincated 2S	4 M NaOH + 0.6 M ZnO	0.3686	8
Zincated 3S	4 M NaOH + 0.6 M ZnO	0.5125	13
57S	4 M NaOH + 0.6 M ZnO	0.0173	1
26S	4 M NaOH + 0.6 M ZnO	0.01103	1
2S	4 M NaOH + 0.6 M ZnO	0.0368	1
3S	4 M NaOH + 0.6 M ZnO	0.0996	1

57S=97.7 wt.% Al, 0.3 wt.% Mn and 2.0 wt.% Mg.

2S=99.1 wt.% Al, 0.3 wt.% Si, 0.4 wt.% Fe, 0.1 wt.% Mn and 0.1 wt.% Mg.

3S=98.8 wt.% Al and 1.2 wt.% Mn.

26S=93.3 wt.% Al, 0.8 wt.% Si, 0.8 wt.% Mn, 0.8 wt.% Mg and 4.3 wt.% Cu.

TABLE 2
Corrosion rate and temperature rise for 99.99% aluminium in different media

Specimen	Medium	Corrosion rate ($\text{mg cm}^{-2} \text{min}^{-1}$)	Temperature rise ($^{\circ}\text{C}$)
99.99% pure Al	4 M NaOH	0.168	4.0
99.99% pure Al (zincated)	4 M NaOH		5.5
99.99% pure Al (zincated)	4 M NaOH + 0.6 M ZnO	1.200	3.0
99.99% pure Al	4 M NaOH + 0.6 M ZnO	0.0902	3.0
CECRI alloy	4 M NaOH	0.03395	1.0
CECRI alloy	4 M NaOH + 0.6 M ZnO	0.1202	3.5
CECRI alloy (zincated)	4 M NaOH + 0.6 M ZnO	0.233	3.5
CECRI alloy (zincated)	4 M NaOH	0.7114	1.0

the performance of this alloy in 4 M NaOH solution is better than that of both super-pure and 57S aluminium, whether the alloy specimens carry a zincate coating or not. The corresponding temperature rises also follow the improved corrosion rates. This verifies that the temperature rise is brought about by the corrosion process.

TABLE 3
Corrosion rate as a function of temperature and time

Specimen	Medium	Temperature (°C)						Corrosion rate (mg cm ⁻² min ⁻¹)								
		Time (min)			Time (min)			Time (min)			Time (min)					
		0	15	30	45	60	0	15	30	45	60	0	15	30	45	60
9.99% Al	4 M NaOH	32	33	34.5	36.0	39.0	0.117	0.1174	0.1670							
Zincated 99.99% Al	4 M NaOH	30	31.5	33.0	35.5	37.0	0.265	0.2660								
99.99% Al	4 M NaOH+0.6 M ZnO	30	31.0	32.0	32.5	33.0	0.0938	0.0855	0.0902							
Zincated 99.99% Al	4 M NaOH+0.6 M ZnO	31	32.5	33.5	33.5	34.0	0.1625	0.1245	0.1215	0.1200						
CECRI alloy	4 M NaOH	31	31.5	32.0	32.0	32.0	0.064	0.041	0.047	0.0340						
CECRI alloy	4 M NaOH+0.6 M ZnO	33	35.0	36.0	36.0	36.5	0.144	0.137	0.117	0.1202						
Zincated CECRI alloy	4 M NaOH+0.6 M ZnO	33	34.0	35.0	36.0	36.5	0.368	0.262	0.230	0.2330						
Zincated CECRI alloy	4 M NaOH	30	30.5	31.0	31.0	31.5	0.118	0.087	0.078	0.0710						
57S	4 M NaOH	29	31.0	34.0	40.0	44.0	0.2943	0.3444	0.4557	0.5373						
57S	4 M NaOH+0.6 M ZnO	29	29.0	29.0	29.0	29.0	0.0429	0.0369	0.0279	0.6254						
Zincated 57S	4 M NaOH	30	30.0	33.5	33.5	0.1950	0.1950	0.2579	0.2861	0.3248						
Zincated 57S	4 M NaOH+0.6 M ZnO	29	29.0	29.0	29.0	29.0	0.0620	0.0464	0.0376	0.0481						

TABLE 4

Variation of open-circuit potential (OCP) with time

Time (min)	OCP (V vs. Hg/HgO-OH ⁻)			
	Zn 57S Al/NaOH+ ZnO	57S/NaOH+ ZnO	57S/NaOH+ ZnO	Zn-Al NaOH+ ZnO
0	-1.408	-1.46	-1.440	-1.35
1	-1.407			-1.36
3	-1.404			-1.36
5	-1.397	-1.37	-1.448	-1.36
8	-1.388			
10	-1.386	-1.37	-1.447	-1.37
15	-1.383	-1.37	-1.440	-1.37
20	-1.381	-1.37	-1.435	-1.37
25	-1.380	-1.37	-1.419	-1.37
30	-1.380	-1.37	1.396	-1.37
40	-1.380	-1.37	1.374	-1.37
50	-1.380	-1.37	-1.386	-1.37
60	-1.380	-1.37	-1.384	-1.37

TABLE 5

Variation of open-circuit potential (OCP) with time

Time (min)	OCP (V vs. Hg/HgO-OH ⁻)			
	99.99% Al in 4 M NaOH	Zincated 99.99% Al in 4 M NaOH	99.99% Al in 4 M NaOH+ 0.6 M ZnO	Zincated 99.99% Al in 4 M NaOH+ 0.6 M ZnO
0	-1.736	-1.653	-1.575	-1.375
1	-1.778	-1.657	-1.571	-1.384
2	-1.772	-1.651	-1.452	-1.383
3	-1.768	-1.646	-1.406	-1.382
4	-1.765	-1.638	-1.383	-1.382
5	-1.760	-1.633	-1.386	-1.381
10	-1.711	-1.609	-1.381	-1.379
15	-1.675	-1.599	-1.378	-1.374
20	-1.637	-1.572	-1.382	-1.375
30	-1.612	-1.591	-1.386	-1.373
40	-1.610	-1.582	-1.385	-1.374
50	-1.610	-1.592	-1.385	-1.375
60	-1.610	-1.572	-1.385	-1.375

- (i) (●) the zincated 57S/4 M NaOH+ ZnO system shows a negative shift from -1.35 to -1.37 V
 (●) the zincated CECRI alloy/4 M NaOH+ ZnO system shows a negative shift from -1.356 to -1.442 V
 (●) the CECRI alloy/4 M NaOH system shows a negative shift from -1.584 to -1.630 V

TABLE 6
Variation of open-circuit potential (OCP) with time

Time (min.)	OCP (V vs. Hg/HgO-OH ⁻)			
	CECRI alloy in 4 M NaOH	Zincated CECRI alloy in 4 M NaOH	CECRI alloy in 4 M NaOH + 0.6 M ZnO	Zincated CECRI alloy in 4 M NaOH + 0.6 M ZnO
0	-1.584	-1.621	-1.510	-1.356
1	-1.588	-1.552	-1.479	-1.420
2	-1.592	-1.551	-1.456	-1.444
3	-1.596	-1.553	-1.453	-1.435
4	-1.600	-1.556	-1.451	-1.437
5	-1.603	-1.558	-1.431	-1.436
10	-1.610	-1.562	-1.419	-1.434
15	-1.615	-1.565	-1.412	-1.428
20	-1.619	-1.569	-1.419	-1.441
30	-1.623	-1.574	-1.404	-1.440
40	-1.627	-1.578	-1.405	-1.443
50	-1.629	-1.579	-1.404	-1.442
60	-1.630	-1.579	-1.404	-1.442

(ii) the OCP of the zincated 99.99% aluminium in 4 M NaOH + ZnO medium remains at -1.375 V; this indicates that corrosion is under general control.

Anodic or cathodic galvanostatic polarization and heat evolution during anodic dissolution

When current is drained from the battery, the anode undergoes 'anodic dissolution'. When this reaction takes place at 100% efficiency as via the following equation:



then the reaction promotes three effects, namely:

- (i) dissolution (material loss) of the anode material according to Faraday's law;
- (ii) the evolution of heat, this is evaluated by the accompanying temperature rise of the electrolyte (note, the electrolyte is stirred efficiently and uniformly throughout the experiment), and
- (iii) generation of electric power at the rate of 2980 Ah per kg of aluminium dissolved.

In other words, the anodic dissolution phenomenon is found to give rise to transfer of mass, heat and charge (electric power) from the electrode towards the electrolyte. Of these three effects, mass transfer and charge transfer are complementary to each other. Therefore, at a fixed anodic current density: (i) a 100% efficient mass transfer accounts for a 100% efficient charge transfer, and (ii) 1 g atom of aluminium gives rise to 3F of charge at 100% efficient anodic dissolution. Any thing less than 100% efficiency is wasteful corrosion and leads to heat evolution. This heat is represented by H_2 (i.e., heat due to anodic dissolution).

In fact the purpose of carrying out anodic polarization is to relate the quantity H_2 to the actual process of anodic polarization. Cathodic polarization experiments

were also performed. From the polarization data, the following parameters were calculated: i_{corr} , the corrosion current density; b_a , the anodic Tafel slope; b_c , the cathodic Tafel slope; $i_{o,a}$, the exchange current density of the anodic partial reaction; $i_{o,c}$, the exchange current density of the cathodic partial reaction. The experimental details are reported in ref. 7.

Table 7 gives the polarization data for 57S aluminium, CECRI alloy and 99.99% pure aluminium (with and without zincate coating) in 4 M NaOH in the presence and absence of ZnO. The 57S aluminium shows an overall anodic control of the kinetics of corrosion in 4 M NaOH, both in the presence and absence of ZnO, as well as a zincate coating (over the specimen) either alone or in combination. The 57S aluminium is found to have a higher b_a than b_c value in 4 M NaOH, whether the solution contains ZnO or not and whether the specimen is zincate coated or not. Corrosion of the 57S aluminium is thus under overall anodic control.

The trend in i_{corr} in the different systems is as follows;

(i) 57S/4 M NaOH (12.4 mA cm^{-2}) > zincated 57S/4 M NaOH (12.2 mA cm^{-2}) > 57S/4 M NaOH + ZnO (9.2 mA cm^{-2}) > zincated 57S/4 M NaOH + ZnO (8.4 mA cm^{-2});

(ii) CECRI alloy/4 M NaOH (44.9 mA cm^{-2}) > zincated CECRI alloy/4 M NaOH (44.8 mA cm^{-2}) > zincated CECRI alloy/4 M NaOH + ZnO (38.5 mA cm^{-2}) > CECRI alloy/4 M NaOH + ZnO (37.9 mA cm^{-2}), and

(iii) in the case of 99.99% pure aluminium, $b_a > b_c$ in all four systems. The b_a values of the different systems follow the order: b_a (99.99% Al) > b_a (CECRI alloy) > b_a (57S).

The difference between the heat evolved during anodic dissolution and corrosion (measured as the rise in temperature of the medium) is presented in Table 8. Clearly, the temperature rise (heat evolved) during corrosion is greater than that during anodic dissolution in all the media for the different varieties of aluminium, whether they are zincate coated or not and whether ZnO is present or absent in the test medium.

The temperature rise (heat evolved) during anodic dissolution is compared with that during cathodic polarization for all the systems in Table 9. It is found that the temperature rise is greater for anodic polarization than for cathodic polarization. During cathodic polarization, the electrode is completely or partially protected and hence the above observation. It is an established fact that during cathodic protection, the surface of the working electrode is taken into an immunity region where the metal ions are prevented from flowing out of the lattice into the solution. In other words, the mass transfer from the specimen is prevented, either wholly or partially, due to cathodic protection. Since Al^{3+} ions do not flow out of the lattice of the aluminium surface during cathodic protection, the following reaction:



does not take place and, hence, no heat is evolved. Among the different systems, only two show zero temperature rise during cathodic polarization compared with anodic dissolution, namely; (i) zincated CECRI alloy in 4 M NaOH containing zinc oxide, and (ii) 99.99% pure aluminium in 4 M NaOH. By contrast, about six systems display a rise of 0.5°C or more during cathodic polarization (Table 9).

Thus, corrosion (H_1) and anodic dissolution (H_2) have already been confirmed as two different sources of temperature rise or heat evolution. Moreover, the overall corrosion kinetics of all the systems have been found to be predominantly under anodic control; there is not one exception, as demonstrated by the respective Tafel parameters. Further, it has been proved beyond doubt that in all cases the anodic dissolution

TABLE 7
Electrochemical parameters of anode materials

Specimen	Medium	i_{corr} (mA cm ⁻²)	E_{corr} (V)	Steady-state OCP		Tafel slope	
				(V vs. Hg/HgO-OH ⁻)	Anodic	(mV/decade)	b_a
57S	4 M NaOH	12.4	-1.380	-1.383	-1.362	105	100
57S	4 M NaOH + 0.6 M ZnO	9.16	-1.390	-1.380	-1.404	75	70
Zincated 57S	4 M NaOH	12.16	-1.444	-1.449	-1.444	20	15
Zincated 57S	4 M NaOH + 0.6 M ZnO	8.35	-1.380	-1.369	-1.383	75	40
CECRI alloy	4 M NaOH	44.87	-1.51	-1.473	-1.494	720	560
CECRI alloy	4 M NaOH + 0.6 M ZnO	37.87	-1.40	-1.381	-1.397	420	20
Zincated CECRI alloy	4 M NaOH	44.82	-1.51	-1.510	-1.534	220	560
Zincated CECRI alloy	4 M NaOH + 0.6 M ZnO	38.46	-1.39	-1.385	-1.377	290	20
99.99% Al	4 M NaOH	62.22	-1.87	-1.599	-1.484	2200	800
99.99% Al	4 M NaOH + 0.6 M ZnO	63.33	-1.40	-1.370	-1.360	550	10
Zincated 99.99% Al	4 M NaOH	39.00	-1.82	-1.489	-1.577	1300	630
Zincated 99.99% Al	4 M NaOH + 0.6 M ZnO	55.00	-1.39	-1.375	-1.375	300	55

TABLE 8

Comparison of temperature rise due to anodic dissolution and corrosion

Specimen	Medium	Temperature rise (°C)	
		Anodic dissolution	Corrosion
57S	4 M NaOH	2.0	20.0
57S	4 M NaOH+0.6 M ZnO	1.0	3.5
Zincated 57S	4 M NaOH	2.0	12.0
Zincated 57S	4 M NaOH+0.6 M ZnO	1.0	2.5
CECRI alloy	4 M NaOH	1.0	1.5
CECRI alloy	4 M NaOH+0.6 M ZnO	1.5	3.5
Zincated CECRI alloy	4 M NaOH	1.0	2.0
Zincated CECRI alloy	4 M NaOH+0.6 M ZnO	1.0	3.5
99.99% pure Al	4 M NaOH	1.0	3.0
99.99% pure Al	4 M NaOH+0.6 M ZnO	1.5	3.0
Zincated 99.99% Al	4 M NaOH	1.5	6.0
Zincated 99.99% Al	4 M NaOH+0.6 M ZnO	1.0	3.0

TABLE 9

Comparison of temperature rise for anodic and cathodic polarizations

Specimen	Medium	Temperature rise (°C)	
		Anodic dissolution	Cathodic polarization
57S	4 M NaOH	2.0	1.0
57S	4 M NaOH+0.6 M ZnO	1.0	0.5
Zincated 57S	4 M NaOH	2.0	1.0
Zincated 57S	4 M NaOH+0.6 M ZnO	1.0	0.5
CECRI alloy	4 M NaOH	1.0	0.5
CECRI alloy	4 M NaOH+0.6 M ZnO	1.5	0.5

process gives rise to more heat than the cathodic polarization. In fact, cathodic polarization of different systems has reduced the temperature rise almost to zero or a very low value. This proves that the local anodic partial reaction is mainly responsible for the temperature rise during corrosion and not the cathodic partial reaction.

Joule heat during cell discharge

The third component of heat in an aluminium/air battery is the Joule heating during discharge. This arises from the total internal resistance that is built up in the battery, i.e.:

$$R_{\text{int}} = (V_{\text{total}} - V_{\text{electrolysis}})/I$$

where, R_{int} is the internal resistance at the temperature at which the battery is discharged, V_{total} the actual terminal voltage of the battery, $V_{\text{electrolysis}}$ the thermoneutral

voltage or voltage required for electrolysis, and I the current. Therefore, $H_3 = \text{Joule heat} = I^2 R_{\text{int}}$. This heat is a wasteful part of the total energy expected from a battery and it is manifested more profoundly with increase in the rate of discharge. Since aluminium/air battery is a high density battery this component of heat has greater probability to manifest itself. The Joule heat is directly proportional to the total internal resistance at any fixed rate of current drain, or it is directly proportional to the amount of current drain per unit time (of course, the temperature at which the experiment is carried out is kept constant). This heat component, H_3 , can be minimized by: (i) increasing the ionic conductivity of the battery electrolyte; (ii) decreasing the inter-electrode distance, and (iii) using internal battery components of least resistance. It is very difficult to predict accurately the theoretical value of the Joule heat. A rough estimate can be made, however, if the design parameters of the entire aluminium/air battery assembly are available. This estimate of H_3 can be improved further if the discharge characteristics of the prototype aluminium/air battery are also available.

If the values of H_1 , H_2 and H_3 are available, then the design of an aluminium/air battery with the desired characteristics is possible. The total heat developed in the aluminium/air battery, due to the sum of the H_1 , H_2 and H_3 components, can be reduced by adopting the remedial measures suggested above.

Further reduction is feasible if the relationship between the heat and the mass transfer (i.e., flow of electrolyte) occurring in the battery is known clearly. This aspect of battery operation will be discussed in a future communication.

Conclusions

1. Addition of ZnO to the alkaline electrolyte solution is found to be beneficial in reducing the heat evolution (and, hence, the temperature rise) during corrosion and anodic dissolution of aluminium in an aluminium/air battery.

2. An Al-In-Ga-Pb quaternary alloy developed at CECRI, is found to be the best alloy anode in alkaline media for the aluminium/air battery. It is based on 99.8% pure aluminium.

3. In presence of ZnO in the alkaline electrolyte, 57S and 26S grades of aluminium are found to be very useful as anodes for aluminium/air batteries.

4. Formation of a zincate coating (developed at CECRI) over aluminium (57S and 26S) and the above alloy helps further to improve the electrochemical characteristics of the anode in the order CECRI alloy > 57S > 26S.

5. The heat evolved (in terms of the temperature rise of the alkaline medium) during corrosion is found to be more than that evolved during anodic dissolution for any grade of aluminium or alloy.

6. The overall kinetics of corrosion of all grades of aluminium in an alkaline medium is under anodic control. The local partial anodic reaction that controls the kinetics is responsible for the heat evolution during corrosion. By contrast, cathodic polarization brings about reduction in both the corrosion rate and the heat evolution.

7. There is a definite correlation between inhibition efficiency and the reduction in heat evolution during corrosion in the presence of inhibitors.

8. The best electrode/electrolyte combination for the aluminium/air battery is the zincated CECRI alloy in 4 M NaOH solution containing ZnO. For this system, the corrosion rate is $0.0340 \text{ mg cm}^{-2}$ per month, the temperature rise is the least, and the anode efficiency is as high as 98% and above.

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