



## Seeking effective dyes for a mediated glucose–air alkaline battery/fuel cell



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### HIGHLIGHTS

- Important aspects in solubility and mobility for 45 redox dyes in alkaline solutions are discussed.
- Polarization effects related to mass transport limitation on indigo and its sulfonate derivatives are explained.
- Optimization strategy for power generation using a matrix of dye selection is presented.
- Co-solvent effect on dye solubility and mobility is studied.
- Loss from mass transport limitation versus other kinetic factors was analyzed to guide future work.

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### ABSTRACT

A significant level of power generation from an abiotic, air breathing, mediated reducing sugar–air alkaline battery/fuel cell has been achieved in our laboratories at room temperature without complicated catalysis or membrane separation in the reaction chamber. Our prior studies suggested that mass transport limitation by the mediator is a limiting factor in power generation. New and effective mediators were sought here to improve charge transfer and power density. Forty-five redox dyes were studied to identify if any can facilitate mass transport in alkaline electrolyte solution; namely, by increasing the solubility and mobility of the dye, and the valence charge carried per molecule. Indigo dyes were studied more closely to understand the complexity involved in mass transport. The viability of water-miscible co-solvents was also explored to understand their effect on solubility and mass transport of the dyes. Using a 2.0 mL solution, 20% methanol by volume, with 100 mM indigo carmine, 1.0 M glucose and 2.5 M sodium hydroxide, the glucose–air alkaline battery/fuel cell attained  $8 \text{ mA cm}^{-2}$  at short-circuit and  $800 \mu\text{W cm}^{-2}$  at the maximum power point. This work shall aid future optimization of mediated charge transfer mechanism in batteries or fuel cells.

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### 1. Introduction

The demand for sustainable alternative energy is growing rapidly worldwide [1]. Renewable resources such as solar, wind, or biofuels are attractive to many to achieve carbon neutral energy conversion. Electrochemical energy conversion is one of the most efficient methods to convert chemical energy into electrical. Primary batteries or fuel cells promise high theoretical efficiency [2,3]. Advanced primary battery systems using metal–air (e.g. lithium–air or zinc–air) reactions continue to attract attention by the energy storage community, even though commercialization is still

impeded by technical barriers that inhibit the redox reaction [4–8]. Presently, hydrogen and methanol are each advocated as the future in clean energy, however both cost too much for large-scale applications. Novel fuel cells that harness energy from carbon-rich fuels, such as glucose, provide an affordable alternative. Such biotic fuel cells already show promise at lower power densities [9].

Higher power densities are possible by optimizing the catalysts that function on the cathode and anode. Improved oxygen reduction on the cathode would decrease polarization interference in the existing fuel cell [10]. Non-noble metal catalysts are of particular interest and are currently being explored [11]. Catalysis of carbon-rich fuels on the anode can also be improved. Platinum (Pt) has been the most used catalyst so far [12]. However, Pt is both costly and scarce. Complete oxidization of glucose down to  $\text{CO}_2$  is a

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desirable process difficult to realize in a conventional fuel cell or bio-fuel cell. A new method of harnessing electrical energy from carbohydrates (e.g. glucose) is highly desired [13]. Past experimentation has been seeking for alternative pathways for catalyzed glucose oxidation [9,14–24]. Enzymatic catalysts have been pursued to mimic glycolysis [14–16]. Microbial fuel cells could simplify the energy harvesting process [17,18]. It is known that mediated

charge transfer can achieve effective catalysis [16,19]. With mediated charge transfer, a reducing sugar–air alkaline battery (SAAB) has been demonstrated in our laboratory, which operates at ambient pressure and temperature, producing a peak power density (PPD) on the order of a few  $\text{mW cm}^{-2}$  [20,21]. Partial oxidation of monosaccharides occurs naturally in strong alkaline solution, requiring no catalyst to yield an ene-diol intermediate. What

**Table 1**  
A comparison of 45 redox dyes to function as electron mediators: molar mass, relative solubility in the nominal solution (see key), current density at short-circuit, cell open circuit voltage, formal reduction potential and diffusivity are shown for each dye in the alphabetical order. Nominal test solutions contain 50 mM dye, 1.0 M glucose, and 2.5 M NaOH. Citations for the formal reduction potential are also included in the footnotes.

Dye	Molar mass ( $\text{g mol}^{-1}$ )	Relative solubility	Current density ( $\text{mA cm}^{-2}$ )	Cell voltage (mV)	$E^\circ$ (mV vs SCE)	D ( $\text{m}^2 \text{s}^{-1}$ )	Refs <sup>a</sup>
ABTS	548.7	3	$0.100 \pm 0.007$	$403 \pm 27$	–399	59.14	1
Alizarin red S	342.3	1	$0.30 \pm 0.02$	$538 \pm 36$	–1079	53.50	2
Alizarin yellow GG	309.2	2	$0.90 \pm 0.06$	$90 \pm 6$	–220	60.25	3
Benzenesulfonic acid, Na salt	180.2	3	0.0	$491 \pm 33$	N/A	N/A	–
Brilliant blue R	826.0	2	$0.100 \pm 0.007$	$520 \pm 35$	–236	9.37	4
Brilliant green	482.6	1	0.0	$468 \pm 31$	–485	5.56	3
Brilliant yellow	624.6	3	$0.100 \pm 0.007$	$540 \pm 36$	–381	3.86	3
Bromocresol purple	540.2	2	$0.70 \pm 0.05$	$559 \pm 38$	–117	7.51	3
Cresyl violet	321.3	2	$0.70 \pm 0.05$	$583 \pm 39$	–680.3	5.08	5
DTNB	396.4	3	$1.5 \pm 0.1$	$496 \pm 33$	–441	12.83	3
Erythrosine	835.9	2	$0.100 \pm 0.007$	$374 \pm 25$	–437	29.51	3
Fast green	765.9	2	$0.5 \pm 0.03$	$588 \pm 39$	–183	10.24	3
Hydroquinone	110.1	3	$1.5 \pm 0.1$	$501 \pm 34$	–274	30.01	6
Indigo carmine (IC)	466.3	2	$4.00 \pm 0.03$	$510 \pm 34$	–687	10.09	6
Indigo tetrasulfonate (ItetS)	734.88	3	$3.80 \pm 0.03$	$441 \pm 30$	–402	N/A	7
Indigo trisulfonate (ItriS)	616.7	3	$3.20 \pm 0.02$	$479 \pm 32$	–435	N/A	7
Indocyanine green	775.0	2	0.0	$360 \pm 24$	N/A	N/A	–
Janus green	511.1	0	$0.100 \pm 0.007$	$459 \pm 31$	–469	20.96	8
Light green SF yellowish	749.9	2	$0.70 \pm 0.05$	$600 \pm 40$	–569	0.53	3
Malachite green oxalate	927.0	0	$0.30 \pm 0.02$	$578 \pm 39$	–506.427	0.95	3
<sup>b</sup> Methoxatin disodium salt (PQQ)	374.2	3	$2.0 \pm 0.1$	$450 \pm 30$	–323	N/A	9
Methyl blue	799.8	2	$1.00 \pm 0.07$	$561 \pm 38$	–576	33.61	3
Methyl orange	327.3	1	0.0	$545 \pm 37$	–332.5	5.66	3
Methyl viologen (MV)	257.2	3	$4.20 \pm 0.03$	$626 \pm 42$	–684	36.72	8
Methylene blue	319.9	1	0.3	$470 \pm 32$	–235	39.68	7
Methylene green	433.0	1	$1.5 \pm 0.1$	$504 \pm 34$	–649	N/A	3
Neutral red	288.8	1	$0.5 \pm 0.03$	$207 \pm 14$	–198	30.04	8
Nicotinamide	122.1	2	0.0	$468 \pm 31$	–559	19.17	3
Nile blue A	353.9	0	$0.40 \pm 0.03$	$499 \pm 33$	N/A	N/A	–
Phenolphthalein	318.3	3	$0.30 \pm 0.02$	$473 \pm 32$	–492	28.77	3
Potassium ferricyanide	329.3	3	$0.100 \pm 0.007$	$518 \pm 35$	–1044.6	53.63	3
Pyromellitic dianhydride	218.1	3	$0.30 \pm 0.02$	$509 \pm 34$	–142	6.62	3
Quinalizarin	272.2	3	$1.00 \pm 0.07$	$523 \pm 35$	73.0	7.71	6
Reactive red 120	1470.0	3	$0.5 \pm 0.03$	$485 \pm 33$	–196	5.90	4
Sodium diphenylamine sulfonate	271.3	2	$0.100 \pm 0.007$	$376 \pm 25$	–336	N/A	10
Sudan black	456.5	0	0.0	$489 \pm 33$	–634	13.46	3
Indigo	262.3	0	$0.20 \pm 0.01$	$561 \pm 38$	N/A	4.14	–
Tartrazine	534.3	3	$0.100 \pm 0.007$	$539 \pm 36$	–485	32.14	3
Terephthalic acid	166.1	3	$0.100 \pm 0.007$	$530 \pm 36$	–565	0.84	3
Thiophene-3,4-dicarboxylic acid	172.2	2	$0.100 \pm 0.007$	$434 \pm 29$	N/A	N/A	–
Thymol blue	466.6	3	$0.5 \pm 0.03$	$510 \pm 34$	–459	1.89	3
Thymolphthalein	430.6	2	$0.100 \pm 0.007$	$432 \pm 29$	–252.7	30.07	11
Trypan Blue	872.9	3	$1.00 \pm 0.07$	$620 \pm 42$	–429	7.17	3
Vanadium oxide (V)	181.88	3	$0.70 \pm 0.05$	$421 \pm 28$	–760	N/A	12
2,6-Dichlorophenolindophenol	268.1	2	$1.5 \pm 0.1$	$258 \pm 17$	–412.8	N/A	13

<sup>1</sup> Ref. [30].

<sup>2</sup> Ref. [31].

<sup>3</sup> This work.

<sup>4</sup> Ref. [32].

<sup>5</sup> Ref. [33].

<sup>6</sup> Ref. [34].

<sup>7</sup> Ref. [35].

<sup>8</sup> Ref. [36].

<sup>9</sup> Ref. [37].

<sup>10</sup> Ref. [38].

<sup>11</sup> Ref. [39].

<sup>12</sup> Ref. [40].

<sup>13</sup> Ref. [41].

Solubility key: 0 = not soluble; 1 = slightly soluble; 2 = mostly soluble; 3 = completely soluble.

<sup>a</sup> Notes: Citations for formal potential ( $E^\circ$ ).

<sup>b</sup> Note: The conc. of PQQ used was 2.5 mM.

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