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Direct Electron Transfer-Type Dual Gas-Diffusion H_2/O_2 Biofuel Cells

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Supporting Information

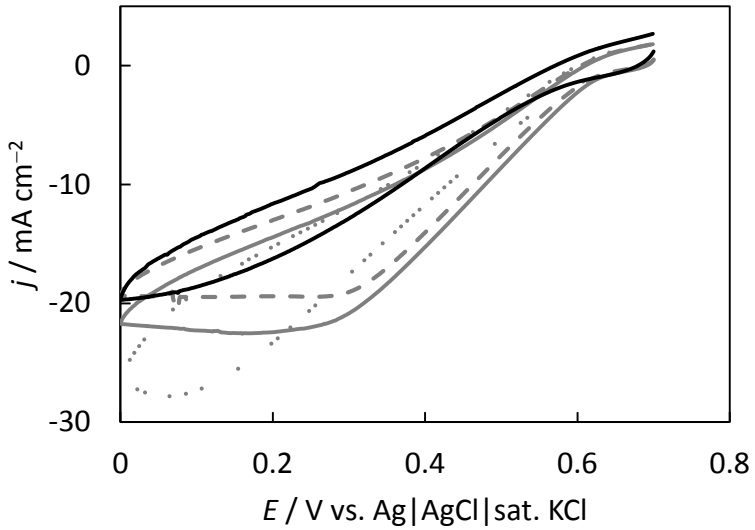

Fig. S1 CVs at the BOD-adsorbed KB/PTFE/WPCC (black line) and the BOD-adsorbed BL/KB/PTFE/WPCC (gray line). The scan rate was 10 mV s^{-1}. The bilirubin concentrations were 1 (dotted line), 3 (solid line), and 5 mM (broken line). Measurements were carried out in 1.5 M citrate buffer (pH 5) at 40 °C under quiescent and O_2-atmospheric conditions.

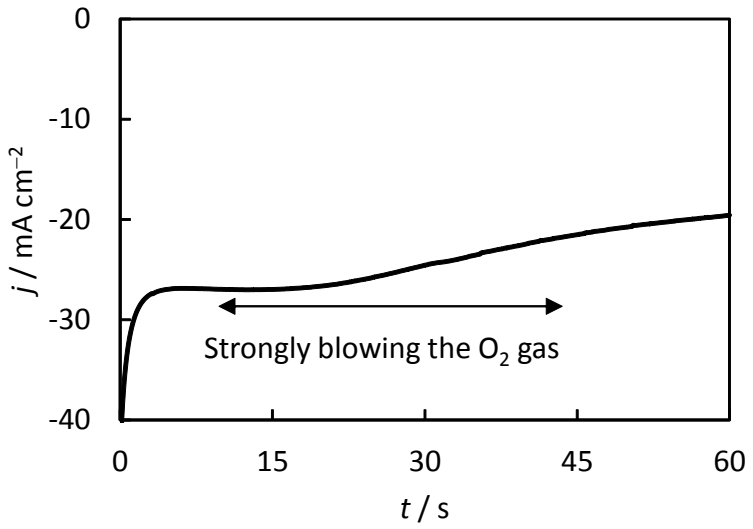

Fig. S2 CA at the BOD-adsorbed KB/PTFE/WPCC. The electrode potential is 0 V. O_2 gas was strongly blown on the surface of the gas-diffusion-type electrode. Measurements were carried out in 1.5 M citrate buffer (pH 5) at 40 °C under quiescent and O_2-atmospheric conditions. The PTFE content was 50%.

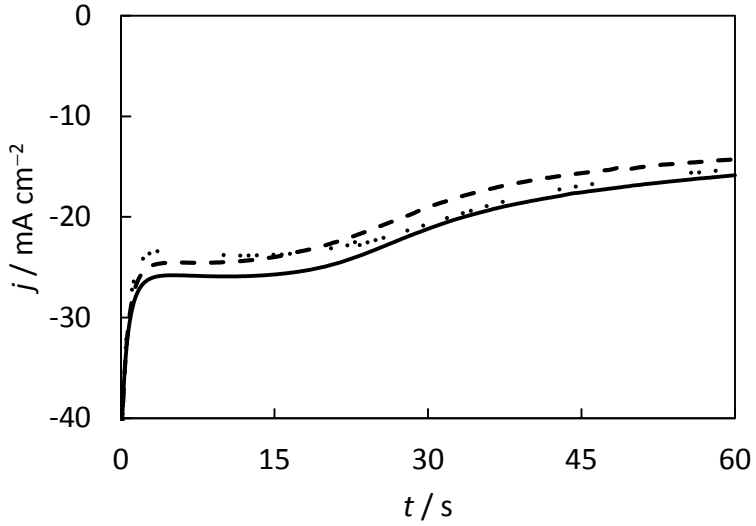

Fig. S3 Repeated CAs at the BOD-adsorbed KB/PTFE/WPCC. The electrode potential is 0 V. Solid, broken, and dotted lines indicate the first, second, and third measurements. The electrode was left in open-circuit for 5 min before the second and third measurements. Measurements were carried out in 1.5 M citrate buffer (pH 5) at 40 °C under quiescent and O_2-atmospheric conditions. The PTFE content was 50%.

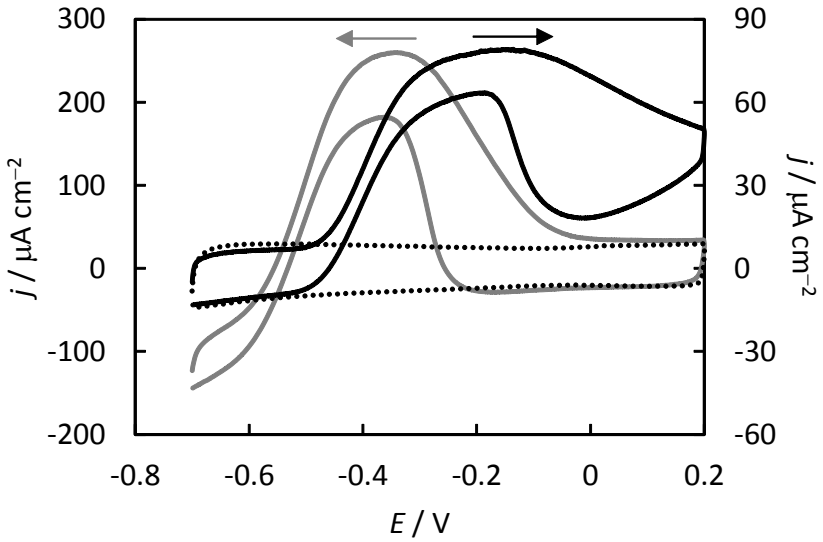

Fig. S4 Rotating disk cyclic voltammograms at *Hm*MBH-adsorbed (black solid line, right axis) and *Dv*MF-adsorbed (gray solid line, left axis) KB-modified glassy carbon electrodes. The dotted black line is the results at KB-modified glassy carbon electrode without enzymes (with the right axis). The scan rate was 10 mV s^{-1}. Measurements were carried out in 0.1 M phosphate buffer (pH 6) at 40 °C, a rotating rate of 4000 rpm under H_2-atmospheric conditions. The electrodes were prepared according to the literature.[80]

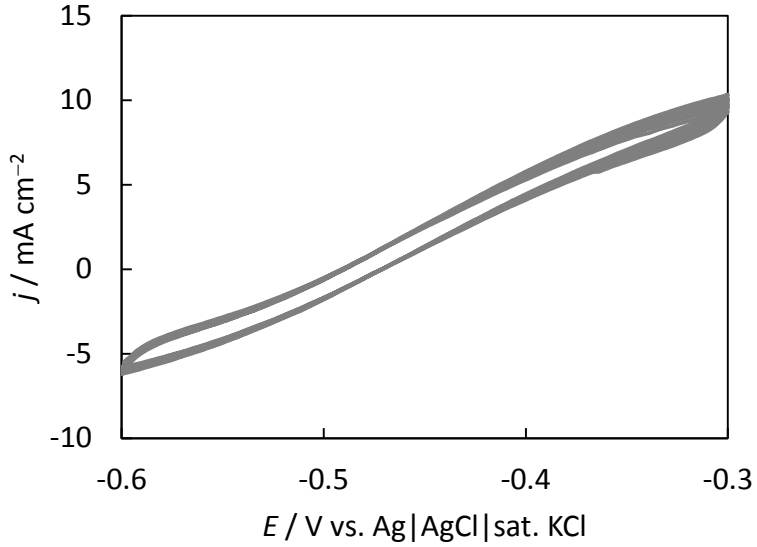

Fig. S5 Multi-sweeped CVs at the *Dv*MF-adsorbed KB/PTFE/WPCC for 10 cycles. The scan rate was 10 mV s^{-1}. Measurements were carried out in 1.5 M citrate buffer (pH 5) at 40 °C under quiescent and H_2-atmospheric conditions. The PTFE content was 50%.

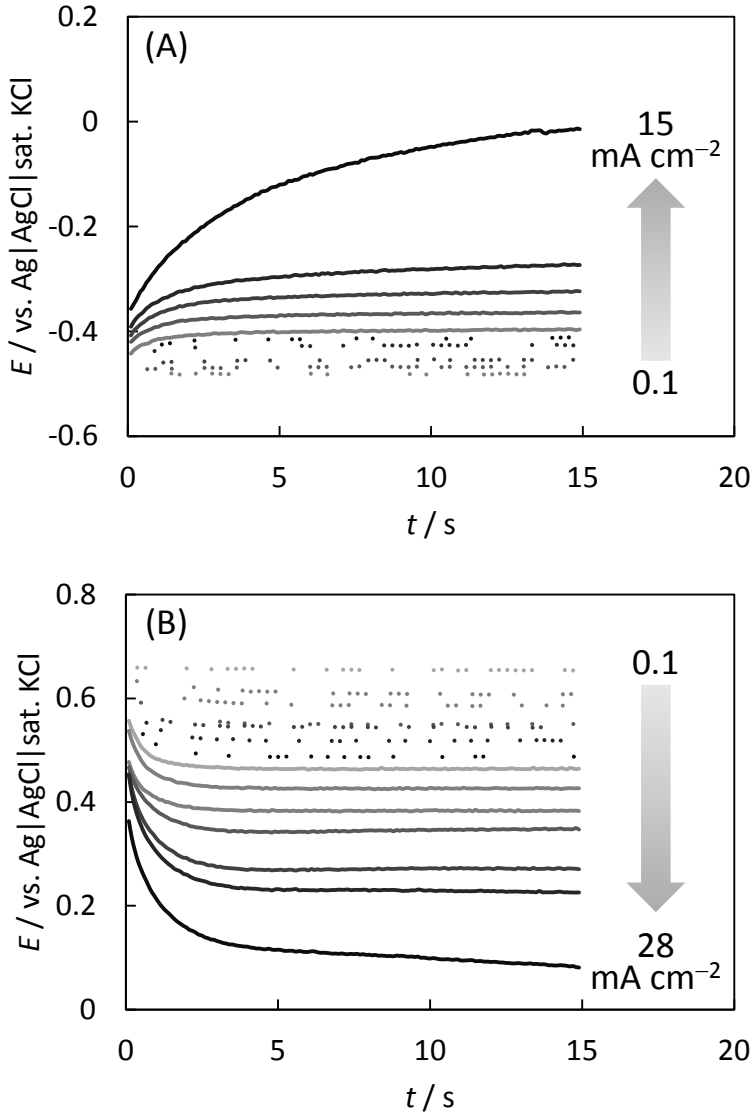

Fig. S6 (A) Chronopotentiograms at the *Dv*MF-adsorbed KB/PTFE/WPCC. The oxidation current densities were 0.1, 1, 2, 4, 5, 6, 8, 10, 12, and 15 mA cm^{-2} (from bottom to top). (B) Chronopotentiograms at the BOD-adsorbed BL/KB/PTFE/WPCC. The reduction current densities were 0.1, 1, 2, 4, 5, 6, 8, 10, 12, 15, 16, 20, 22, and 28 mA cm^{-2} (from top to bottom). Measurements were carried out in 1.5 M citrate buffer (pH 5) at 40 °C under quiescent and (A) H_2- or (B) O_2-atmospheric conditions. The PTFE content and the bilirubin concentration were 50% and 3 mM, respectively.

Table S1 Details of recent development in power of biofuel cells

H_2/O_2 Biofuel Cells						
Date	Power density / mW cm^{-2}	Anode catalyst	Cathode catalyst	Conditions	Notes	Ref.
2001	0.4	*Dv*MF cells	*Mv*BOD	pH 7, room temperature (RT), 100% H_2, 100% O_2	MV and ABTS are used as mediators. Using a membrane as a separator	17
2002	0.32	Hase	LAC	pH 8 (anode), pH 4.2 (cathode)	Using a membrane as a separator	18
2006	0.005	*Rm* CH34 MBH	*Tv*LAC	pH 5, 100 mM citrate, 3% H_2 in air	Low H_2 concentration was selected for avoiding an explosion	19
2010	0.063	*E. coli* Hyd1	*Mv*BOD	pH 5, 100 mM citrate, RT, 100% H_2, 100% O_2	Using a membrane as a separator	20
2012	0.12	*E. coli* Hyd1	*Mv*BOD	pH 5, 100 mM citrate, RT, H_2/air (80:20 mixture), under quiescent conditions	N.A	21
2012	0.3	*Aa*MBH	*Mv*BOD	pH 6.8, 50 mM Hepes, 60 °C (anode), 25 °C (cathode), 100% H_2, 100% O_2	Using a membrane as a separator	22
2013	a) 0.56 b) 1.67	*E. coli* Hyd1	*Mv*BOD	pH 6, 100 mM phosphate, 25 °C 78% H_2, 22% air, under quiescent conditions	a) The cathode surface area is equal to anode. b) The cathode surface area is 3-times larger than anode.	23
2014	1.5	*Aa*MBH	*Bp*BOD	pH 6, 100 mM phosphate, 60 °C 100% H_2, 100% O_2, under quiescent conditions.	Using a membrane as a separator	24
2015	0.72	*Aa*MBH	*Bp*BOD	pH7.2, phosphate buffer, 45 °C Air-breathing/H_2 under quiescent conditions	Gas-diffusion-type electrode is used for the cathode	25
2016	8.4	*Dv*MF	*Mv*BOD	pH 5, 1.5 M citrate, 40 °C, 100% H_2, 100% O_2, under quiescent conditions	Dual gas-diffusion-system	This study
DET-type Biofuel Cells with other substrates						
2007	a) 0.4 b) 0.85	FDH	*Ts*LAC	pH 5, McIlvaine, RT with 200 mM fructose, O_2 saturated	a) Under quiescent conditions b) With stirring	89
2009	a) 0.66 b) 0.87	FDH	*Mv*BOD	pH 6, 100 mM acetate, 25 °C with 200 mM fructose, O_2 saturated	a) Under quiescent conditions b) With stirring	90
2011	1.3	GOD and catalase	*Tv*LAC	pH 7, 100 mM phosphate, with 50 mM glucose, air saturated	No information about stirring.	91
2011	1.8	FDH	*Ts*LAC	pH 5, McIlvaine, with 200 mM fructose, under O_2 saturated stirring conditions	N.A.	59
2013	a) 0.95 b) 2.55	FDH	*Mv*BOD	pH 5, McIlvaine, with 500 mM fructose, air saturated, under quiescent conditions.	a) Single-layer cell, b) Triple-layer cell Gas-diffusion-type electrode is used for the cathode.	83
2014	2.6	FDH	*Mv*BOD	pH 5, 1.0 M citrate, RT, with 500 mM fructose, air saturated, under quiescent conditions	Gas-diffusion-type electrode is used for the cathode.	56