Electronic Supplementary Information (ESI)

Appendix I.

List of review articles on CO_2 electrochemical transformation into useful products.

Author (year)	Publication title	Ref.
A. F. Sammells et al. (1993)	Electrochemical and electrocatalytic reactions of carbon dioxide	[A.1]
M. Jitaru et al. (1997)	Electrochemical reduction of carbon dioxide on flat metallic cathodes	[A.2]
C. M. Sánchez-Sánchez et al. (2001)	Electrochemical approaches to alleviation of the problem of carbon dioxide accumulation	[A.3]
R. P. S Chaplin and A. A. Wragg (2003)	Effects of process conditions and electrode material on reaction pathways for carbon dioxide electroreduction with particular reference to formate formation	[A.4]
M. Gattrell et al.(2006)	A review of the aqueous electrochemical reduction of CO_2 to hydrocarbons at copper	[A.5]
M. Jitaru (2007)	Electrochemical carbon dioxide reduction- fundamental and applied topics (review)	[A.6]
Y. Hori (2008)	Electrochemical CO_2 reduction on metal electrodes	[A.7]
E. E. Benson et al.(2008)	Electrocatalytic and homogeneous approaches to conversion of CO_2 to liquid fuels	[A.8]
J. Lee et al.(2009)	Electrocatalytic recycling of CO_2 and small organic molecule	[A.9]
W. Li (2010)	Electrocatalytic reduction of CO_2 to small organic molecule fuels on metal catalyst	[A.10]
D. T. Whipple and P. J. A. Kenis (2010)	Prospects of CO_2 utilization via direct heterogeneous electrochemical reduction	[A.11]
G. Centi and S. Perathoner (2011)	CO_2-based energy vectors for the storage of solar energy	[A.12]
N. S. Spinner et al.(2012)	Recent progress in the electrochemical conversion and utilization of CO_2	[A.13]
Y. Oh and X. Hu (2013)	Organic molecules as mediators and catalysts for photocatalytic and electrocatalytic CO_2 reduction	[A.14]
H. R. Jhong et al.(2013)	Electrochemical conversion of CO_2 to useful chemicals: current status, remaining challenges, and future opportunities	[A.15]
J. P. Jones et al.(2013)	Electrochemical CO_2 Reduction: Recent advances and current trends	[A.16]
E. V. Kondratenko et al.(2013)	Status and perspectives of CO_2 conversion into fuels and chemicals by catalytic, photocatalytic and electrocatalytic processes	[A.17]
R. J. Lim et al. (2014)	A review on the electrochemical reduction of CO_2 in fuel cells, metal electrodes and molecular catalysts	[A.18]
J. Qiao et al.(2014)	A review of catalysts for the electroreduction of carbon dioxide to produce low-carbon fuels	[A.19]

Appendix II.

Summary of the best performance data for the electrocatalytic reduction of CO_2 to CH_3OH. Potential values (E) have been converted to SCE reference electrode and current density (j) in $mA \cdot cm^{-2}$.

Electrode	Electrolyte/Cell configuration	E (V vs. SCE)	Current density ($mA \cdot cm^{-2}$)	Concentration ($mg \cdot L^{-1}$)	Reaction rate (10^{-6} $mol \cdot m^{-2} \cdot s^{-1}$)	FE (%)	Ref.
Cu-based electrodes							
Pre-oxidized Cu foil (1 h, 130 ºC)	0.5M $KHCO_3$/Undivided	-0.05	0.069	-	3.61×10^{-3} [a]	~240	[A.20]
Pre-oxidized Cu foil (17 h, 130 ºC)		-1.55	7.1	-	23.6[a]	-	
Anodized Cu foil		-1.25	1.4	-	27.8[a]	~120	
Pre-oxidized Cu-TiOx (50 min., 300 ºC)		-1.06	0.74	-	22.5[a]	~180	
Pre-oxidized Cu-TiOx (45 min., 500 ºC)		-0.45	0.30	-	33.3[a]	~30	
Perovskite ($La_{1.8}Sr_{0.2}CuO_4$) based carbon GDE	0.5M KOH/ Two compartments (Nafion 117)	-2.30 to -2.60	180	-	-	2	[A.21]
Air-furnace oxidized Cu	0.5M $KHCO_3$/Undivided -Three electrodes	-1.50	~10	0.12-0.25[a] (10 min)	1.94[a]	2	[A.22]
Anodized Cu foil (Electrochemically oxidized)		-1.40	~5	0.20-0.39[a] (10 min)	3.06[a]	20	
Cu_2O electrodeposited-stainless steel		-1.10	~5	7.64-15.29[a] (10 min)	11.9[a]	38	
$Cu_{88}Sn_6Pb_6$ alloy foil	2M HCl/Two compartments (Nafion 117)	-0.65	0.24	70.99 [a,b] (2 h)	10.6[a,b]	34.3	[A.23]
	1.5M HCl-0.5M NaCl/Two compartments (Nafion 117)	-0.70	0.36	90.93 [a,b] (2 h)	13.5 [a,b]	28.2	
	1.5M HCl-0.17M $MgCl_2$/Two compartments (Nafion 117)	-0.70	0.38	96.77 [a,b] (2 h)	14.4[a,b]	34.1	
	1.5M HCl-0.17M $CaCl_2$/Two compartments (Nafion 117)	-0.70	0.39	103.76[a,b] (2 h)	15.4[a,b]	29.6	
	1.5M HCl-0.17M $BaCl_2$/Two compartments (Nafion 117)	-0.70	0.41	118.59 [a,b] (2 h)	17.6[a,b]	36.3	
	1.5M HCl-0.08M $AlCl_3$/Two compartments (Nafion 117)	-0.70	0.43	120.29[a,b] (2 h)	17.9[a,b]	17.8	
	1.5M HCl-0.08 M $NdCl_3$/Two compartments (Nafion 117)	-0.70	0.61	182.35[a,b] (2 h)	27.1[a,b]	34.6	
	1.5M HCl-0.08 M $LaCl_3$/Two compartments (Nafion 117)	-0.70	0.68	193.79[a,b] (2 h)	28.8[a,b]	35.7	
	1.5M HCl-0.33 M $ZrCl_4$/Two compartments (Nafion 117)	-0.70	0.45	142.51[a,b] (2 h)	21.2[a,b]	23.7	
	0.01M CH_3COOH-0.01M CH_3COONa/Two compartments (Nafion 117)	-0.80	0.29	60.85[a,b] (2 h)	9.06[a,b]	10.2	
	0.01M CH_3COOH-0.003M $BaCl_2$/Two compartments (Nafion 117)	-0.80	0.39	78.61[a,b] (2 h)	11.7[a,b]	7.9	
	0.01M CH_3COOH-0.0016M $LaCl_3$/Two compartments (Nafion 117)	-0.80	0.96	175.57[a,b] (2 h)	26.1[a,b]	7.1	
	0.01M CH_3COOH-0.05M $LaCl_3$/Two compartments (Nafion 117)	-0.80	0.98	186.05[a,b] (2 h)	27.7[a,b]	6.7	
	0.01M CH_3COOH-0.01M CH_3COONa/Two compartments (Nafion 117)	-1.75	5.96	226.56[a,b] (2 h)	33.7[a,b]	0.5	
	0.01M CH_3COOH-0.0016M $LaCl_3$/Two compartments (Nafion 117)	-1.75	6.26	248.99[a,b] (2 h)	37.1[a,b]	0.8	
$Cu_{63.9}Au_{36.1}$ on nanoporous Cu films	0.5 M $KHCO_3$/ Two compartments H-type	- 1.00	~0.85	-	-	15.9	[A.24]
Electroplating of Cu on carbon paper	SPE (Nafion)/ Two compartments	-	11.1	-	-	0.54	[A.25]
	SPE (SPEEK)/ Two compartments	-	8.9	-	-	0.44	
Cu foil electropolishing in H_3PO_4	0.1M $KHCO_3$/ Two compartments (AMV Selemion)	-1.40	~10	-	-	~0.13	[A.26]
Cu nanocluster thermally deposited-ZnO (1010)	0.1M $KHCO_3$/ Two compartments (Nafion)	-1.45	~12	9.44[a] (1 h)	4.25[a]	2.8	[A.27]
Cu nanocluster (111)		-1.45	~12	3.84×10^{-2} [a] (1 h)	0.17[a]	0.1	
Cu_2O electrodeposited on carbon paper	SPE (CMI-7000)/Two compartments MEA	-2[c]	~3.7	-	-	5	[A.28]
	SPE (AMI-7001)/Two compartments MEA		~2.4	-	-	20	
Cu/CuO nanopowder painted carbon GDE	1M $KHCO_3$/ Two compartments (Nafion 117)	-1.40	17.3	-	-	2.5	[A.29]

Ru/Mo-based electrodes							
Electroplated Ru on Cu foil	0.2M Na_2SO_4/ Two compartments (Na_2SO_4/agar bridge)	-0.54	0.08	-	-	42	[A.30]
Teflon-supported Ru on Cu foil		-0.56	0.09	15.84 (18.6 h)	-	-	
Mo foil on Cu wire	0.2M Na_2SO_4/Undivided	-0.80	0.05	-	-	55	[A.31]
	0.05M H_2SO_4/Undivided	-0.69	0.31	-	-	46	
Mo foil-Cu (KOH/HF pre-treated)	0.2M Na_2SO_4/Undivided	-0.80	0.12	-	-	84	
RuO_2+TiO_2 (35/65)-Ti	0.05M H_2SO_4/Two compartment	-0.55	~5	9.7 (-)	-	24	[A.32]
RuO_2+TiO_2 (20/80)-Ti		-0.55	~5	3.5 (-)	-	5	
RuO_2+MoO_2+TiO_2 (25/30/45)-Ti		-0.55	~5	10.2 (-)	-	12	
RuO_2+Co_3O_4+SnO_2+TiO_2 (20/10/8/62)-Ti		-0.55	~5	6.7 (-)	-	7	
RuO_2+TiO_2 (35/65)-Ti	0.2M Na_2SO_4/Two compartment	-1.48	0.06	8.4 (-)	-	76	
	0.5M $KHCO_3$/Two compartment	-1.50	0.1	1 (-)	-	5	
	Phosphate buffer 0.2M/Two compartment	-1.44	0.08	5 (-)	-	35	
RuO_2 + Co_3O_4 + SnO_2 + RuO_2 + Co_3O_4 + SnO_2 + TiO_2 (20/10/8/62)-Ti	0.2M Na_2SO_4/Two compartment	-1.49	0.05	6.8 (-)	-	53	
TiO_2/RuO_2 (75/25)-Cu (deposited 300 mC/cm^2)	0.5M $KHCO_3$/ Undivided-Rotating-disk electrode assembly	-1.00	5	-	-	29.8	[A.33]
RuOx thermally deposited-Ti	0.5M $KHCO_3$/Undivided-Three electrodes	-0.80	~2	~1.60-2.99[a] (2-8 h)	~0.35-0.16[a]	30.5-17.2	[A.34]
RuOx/Cu thermally deposited-Ti		-0.80	~2	~1.39-3.95[a] (2-8 h)	~0.30-0.21[a]	18.2-41.3	
RuOx/Cd thermally deposited-Ti		-0.80	~2	~1.17-4.27[a] (2-8 h)	~0.26-0.23[a]	20.4-38.2	
RuO_2-painted (5-6 layers)-BDD	0.4M Britton–Robinson sol. (H_3BO_3+H_3PO_4+$C_2H_4O_2$)/ Two compartments	-0.80	~5	31.31 (~6 h)	3.44[a]	8.12	[A.35]
RuOx sprayed-Pt	0.5M $NaHCO_3$/ Two compartments H-type cell (Nafion 117)	-0.80	~1.2	-	-	30.5	[A.36]
RuO_2/TiO_2 nanoparticles-Pt		-0.80	~1.2	-	-	40.2	
RuO_2/TiO_2 nanotubes-Pt		-0.80	~1.2	-	-	60.5	
MoS_2-rods in TiO_2 nanotubes	0.1M $KHCO_3$/Undivided, three electrodes	-1.30	~0.75	202.24[a] (6 h)	-	44.9	[A.37]
Other transition metal: Fe, Ti and Hg							
Everitt's salt (ES, $K_2Fe"[Fe"(CN)_6]$) coated-Pt	1,2-dihydroxybenzene-3,5-disulfonate (tiron) ferrate(III) complex-C_2H_6O/Three compartment (glass frit)	-1.00	-	0.32[a] (6 h)	3.86 x 10^{-2} [a]	-	[A.38]
Everitt's salt coated-Pt	$[Fe(C_6H_2(OH)_2(SO_3)_2)_2]$-$CH_3OH$/ Two compartment (glass frit)	-0.90	-	5.15[a] (5 h)	-	14.5	[A.39]
	$[Fe(C_6H_2(OH)_2(SO_3)_2)_2]$-$C_2H_6O$/ Two compartment (glass frit)	-1.10	-	0.04[a] (5 h)	-	0.06	
	$Na_3[Fe(CN)_5(H_2O)]$-CH_3OH/ Two compartment (glass frit)	-0.90	-	4.74[a] (5 h)	-	15.5	
	$[NH_4]_2[CrCl_5(H_2O)]$-C_2H_6O/ Two compartment (glass frit)	-1.10	-	0.04[a] (5 h)	-	0.05	
	$K[Cr(C_2O_4)_2(H_2O)_2)]$-CH_3OH/ Two compartment (glass frit)	-0.90	-	5.09[a] (5 h)	-	14.5	
	$K[Cr(C_2O_4)_2(H_2O)_2)]$-C_2H_6O/ Two compartment (glass frit)	-1.10	-	0.03[a] (5 h)	-	0.04	
Everitt's salt supported-Pt	0.1M KCl-10mM $Na_3[Fe(CN)_5(H_2O)]$)-15 mM CH_3OH/ Two compartment (glass frit)	-0.70	-	2.62-7.20[a] (2-9 h)	1.20-0.69[a]	56.3-45.3	[A.40]
Indigo ($C_{16}H_{10}N_2O_2$)/ graphite-Pt		-0.70	-	1.60-4.35[a] (2-9 h)	0.69-0.42[a]	70.2-37.2	
Alizarin ($C_{14}H_8O_4$)/ graphite-Pt		-0.50 to -0.70	-	2.18-2.50[a] (3 h)	0.63-0.72[a]	44.3-69.4	
ES supported- Fe		-0.30 to -0.70	-	5.47-5.66[a] (5 h)	0.95-0.98[a]	45.5-45.7	
Indigo/ graphite-Fe		-0.70	-	6.24[a] (5 h)	1.08[a]	42.8	
2-aminoanthraquinone/ graphite-Fe		-0.70	-	3.42[a] (5 h)	0.59[a]	31	
Alizarin ($C_{14}H_8O_4$)/ graphite/Fe		-0.70	-	3.55[a] (5 h)	0.62[a]	30.8	

p-benzoquinone/graphite powder supp. on Fe		-0.70	-	4.48[a] (5 h)	0.78[a]	36	
Fe(II) TPP (tetraphenylporphyrin) evaporated-Pt	2-hydroxil-1-nitrosonaphthalene-3,6-disulphonatocobal(II) l in 0.1M KCl-CH_3OH/ Undivided three electrodes (proton exchange membrane)	-0.50	0.36	3.04[a] (5 h)	0.35[a]	12.2	[A.41]
Co(II) TPP evaporated-Pt		-0.50	0.39	4.35[a] (5 h)	0.50[a]	15.1	
Ni(II) TPP evaporated-Pt		-0.50	0.15	3.17[a] (5 h)	0.37[a]	14.4	
Cr(III) TPPCl supported-Pt		-0.50	0.10	1.67[a] (5 h)	0.19[a]	11.6	
Fe(III) TPPCl supported-Pt		-0.50	0.37	6.14[a] (5 h)	0.71[a]	11.1	
Co(II) TPP supported-Pt plate	Aquopentacyanoferrate (II) ($Na_3[Fe(CN)_5(H_2O)$-0.1M KCl-CH_3OH/ Undivided, three electrodes (proton exchange membrane)	-0.50	0.07	3.33[a] (5 h)	0.35[a]	32.9	
Fe-S/Pan/PB electrochemical laminated-Pt (PB: Prussian blue; PAn: polyaniline)	0.5M KCl/Two compartments H-type	-0.80	-	0.04[a] (24 h)	5.62-6.28 x 10^{-4} [a]	6.8	[A.42]
Fe-T/Pan/PB-Pt			-	0.07[a] (24 h)	1.03-1.15 x 10^{-3} [a]	8.3	
Fe-C/Pan/PB-Pt			-	0.11[a] (24 h)	1.64-1.83 x 10^{-3} [a]	12.2	
Fe-C/Pan/PB-Pt-self assembled	0.2M KCl/ Two compartments H-type		-	0.28[a] (24 h)	4.08-4.55 x 10^{-3} [a]	10.1	
			-	0.05[a] (24 h)	7.03-7.85 x 10^{-4} [a]	5.2	
	0.5M KCl/ Two compartments H-type		-	0.04 [a] (24 h)	5.62-6.28 x 10^{-4} [a]	6.3	
Post-transition Ga- based electrodes							
n-GaAs-crystal-(111)As	0.2M Na_2SO_4/ Two compartments (Na_2SO_4/agar bridge)	-1.20 to -1.40	0.16-0.2	-	-	100	[A.43]
n-GaAs-crystal-(111)Ga			0.34	-	-	30-80	
n-GaAs-crystal-(110)Ga			0.13	-	-	14	
n-GaAs-crystal-(100)Ga			0.15-0.16	-	-	1-0	
Pt metal group metal- based electrodes							
Pd with Cu electrodeposited (coverage of 10)	0.1M $KHCO_3$/-	-1.60	-	-	-	2.28	[A.44]
Pd-H with Cu electrodeposited (coverage of 10)		-1.60	-	-	-	6.97	
Pd disk (HNO_3/NaOH treated)	0.5M $NaClO_4$+10mM Pyridine/ Two compartment (glass frit)	-	0.04	-	-	30	[A.45]
Pt/C powder deposited-carbon paper	SPE (Nafion 117)/Two compartments MEA flow cell	-0.35	-	-	-	40	[A.46]
Pt/C powder deposited-carbon paper	SPE (Nafion 117)/Two compartments MEA flow cell	-0.40	~20	0.40[a] (1 h)	~7.44[a]	35	[A.47]
Pt-Ru/C powder deposited-carbon paper		-0.45	~15	10^{a} (1 h)	~186[a]	75	

[a] Calculated from reported data

[b] Data for total CO_2 reduction to various products (including CH_3OH)

[c] Reference electrode not provided

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