In-Silico High-Throughput Screening of Ag-Based Electrocatalysts for Anion-Exchange Membrane Fuel Cells

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SUPPORING INFORMATION

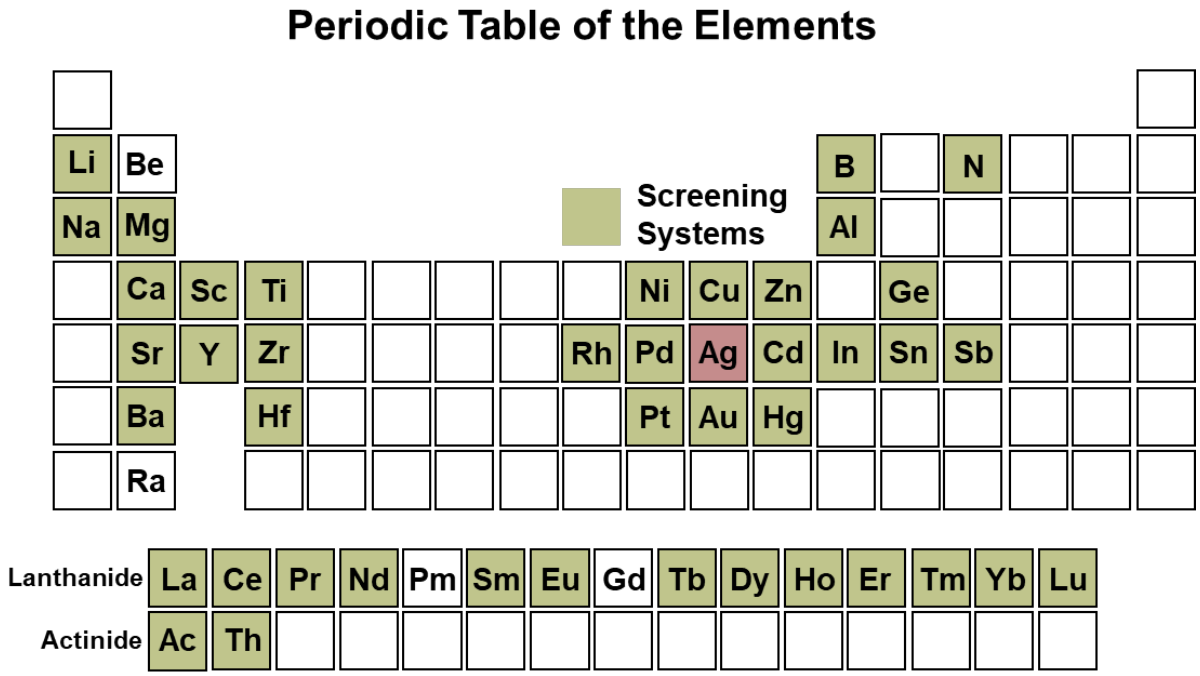

Figure S1. Elements in the Ag-X bimetallic alloys that we explored through high-throughput screening considering various crystal structures based on the Materials Project database. The green elements indicate the X elements of the screened Ag-X bimetallic alloys.

No	Material	Space Group	Crystal Structure	No	Material	Space Group	Crystal Structure
1	Ag_3Sb	Pmmn	Orthorhombic	**54**	$SrAg_5$	P6/mmm	Hexagonal
2	Ag_3Sn	Pmmn	Orthorhombic	**55**	La_3Ag	$P6_3/mmc$	Hexagonal
3	YbAg	Pnma	Orthorhombic	**56**	Si_3Ag	$P6_3/mmc$	Hexagonal
4	$AlAg_2$	Cmcm	Orthorhombic	**57**	$ReAg_3$	$P6_3/mmc$	Hexagonal
5	CaAg	Cmcm	Orthorhombic	**58**	$AgAu_3$_2a	$P6_3/mmc$	Hexagonal
6	$CaAg_2$	Imma	Orthorhombic	**59**	AgSe	F-43m	Cubic
7	$NdAg_2$	Imma	Orthorhombic	**60**	AgO	F-43m	Cubic
8	$PrAg_2$	Imma	Orthorhombic	**61**	AgN	F-43m	Cubic
9	$SrAg_2$	Imma	Orthorhombic	**62**	AgC_1	F-43m	Cubic
10	$YbAg_2$	Imma	Orthorhombic	**63**	$InAg_3$	Pm-3m	Cubic
11	$LuAg_4$	I4/m	Tetragonal	**64**	$ZnAg_3$	Pm-3m	Cubic
12	$ScAg_4$	I4/m	Tetragonal	**65**	AcAg	Pm-3m	Cubic
13	Ti_2Ag	I4/mmm	Tetragonal	**66**	MgAg	Pm-3m	Cubic
14	$ErAg_2$	I4/mmm	Tetragonal	**67**	CdAg	Pm-3m	Cubic
15	$TmAg_2$	I4/mmm	Tetragonal	**68**	PrAg	Pm-3m	Cubic
16	$LiAg_3$	I4/mmm	Tetragonal	**69**	YAg	Pm-3m	Cubic
17	Ag_3Pd	I4/mmm	Tetragonal	**70**	DyAg	Pm-3m	Cubic
18	Ag_3Pd_a	I4/mmm	Tetragonal	**71**	SmAg	Pm-3m	Cubic
19	Li_3Ag_1	I4/mmm	Tetragonal	**72**	HoAg	Pm-3m	Cubic
20	Zr_2Ag	I4/mmm	Tetragonal	**73**	TmAg	Pm-3m	Cubic
21	$TbAg_2$	I4/mmm	Tetragonal	**74**	NdAg	Pm-3m	Cubic
22	$DyAg_2$	I4/mmm	Tetragonal	**75**	ScAg	Pm-3m	Cubic
23	Hf_2Ag	I4/mmm	Tetragonal	**76**	ErAg	Pm-3m	Cubic
24	$ScAg_2$	I4/mmm	Tetragonal	**77**	TbAg	Pm-3m	Cubic
25	YAg_2	I4/mmm	Tetragonal	**78**	$AgAu_3$_1	Pm-3m	Cubic
26	$HoAg_2$	I4/mmm	Tetragonal	**79**	YbAg	Pm-3m	Cubic
27	$LuAg_2$	I4/mmm	Tetragonal	**80**	$MgAg_3$_1	Pm-3m	Cubic
28	$TbAg_3$_1	I4/mmm	Tetragonal	**81**	LiAg	Pm-3m	Cubic
29	$ZrAg_2$	I4/mmm	Tetragonal	**82**	Ag_3Pt	Pm-3m	Cubic
30	$NaAg_3$	I4/mmm	Tetragonal	**83**	$AgPt_3$	Pm-3m	Cubic
31	$NiAg_3$	I4/mmm	Tetragonal	**84**	Ag_3Hg	Pm-3m	Cubic
32	Ag_3P	I4/mmm	Tetragonal	**85**	InAg	Pm-3m	Cubic
33	UAg_3	I4/mmm	Tetragonal	**86**	GaAg	Pm-3m	Cubic
34	Si_3Ag	I4/mmm	Tetragonal	**87**	AgHg	Pm-3m	Cubic
35	$TcAg_3$	I4/mmm	Tetragonal	**88**	Ag_3Ge	Pm-3m	Cubic
36	Ag_3N	I4/mmm	Tetragonal	**89**	Zr_3Ag	Pm-3m	Cubic
37	In_2Ag	I4/mcm	Tetragonal	**90**	K_3Ag	Pm-3m	Cubic
38	Th_2Ag	I4/mcm	Tetragonal	**91**	Rb_3Ag	Pm-3m	Cubic
39	AgH	$P6_3mc$	Hexagonal	**92**	$TmAg_3$	Pm-3m	Cubic
40	AgB_2	P6/mmm	Hexagonal	**93**	Ti_3Ag	Pm-3m	Cubic

41	$EuAg_5$	P6/mmm	Hexagonal	**94**	$NiAg_3$	Pm-3m	Cubic
42	$BaAg_5$	P6/mmm	Hexagonal	**95**	$BeAg_3$	Pm-3m	Cubic
43	$BaAg_2$	P6/mmm	Hexagonal	**96**	Ag_3Rh	Pm-3m	Cubic
44	AgN	$P6_3/mmc$	Hexagonal	**97**	V_3Ag	Pm-3m	Cubic
45	$AgAu_3\_2$	$P6_3/mmc$	Hexagonal	**98**	AgN	Pm-3m	Cubic
46	$MgAg_3\_2$	$P6_3/mmc$	Hexagonal	**99**	$AgAu_3\_1a$	Pm-3m	Cubic
47	$CdAg_3$	$P6_3/mmc$	Hexagonal	**100**	Ag_3Pt_a	Pm-3m	Cubic
48	$AcAg_3$	$P6_3/mmc$	Hexagonal	**101**	$AgPt_3\_a$	Pm-3m	Cubic
49	Li_3Ag_2	$P6_3/mmc$	Hexagonal	**102**	Ag_3Hg_a	Pm-3m	Cubic
50	Mg_3Ag	$P6_3/mmc$	Hexagonal	**103**	AgN	Fm-3m	Cubic
51	$TbAg_3\_2$	$P6_3/mmc$	Hexagonal	**104**	AgC_2	Fm-3m	Cubic
52	$CuAg_3$	$P6_3/mmc$	Hexagonal	**105**	Pt	Fm-3m	Cubic
53	$AgPb_3$	$P6_3/mmc$	Hexagonal	**106**	Ag	Fm-3m	Cubic

Table S1. Materials, space groups, and crystal structures were explored through high-throughput screening considering various crystal structures. AB, AB_1 and AB_2 denote materials with the same composition but different space groups. AB_a denotes materials that are modeled as a AgX skin due to the high dissolution potential of the X atom.

Thermodynamic overpotential calculations

The oxygen reduction reaction (ORR) forms $4OH^-$ from molecular oxygen O_2, H_2O and electrons.

$$O_2\,(g) + 2H_2O\,(l) + 4e^- \leftrightarrow 4OH^- \quad \Delta G = -4.92\ eV$$

The $4e^-$-transfer oxygen reduction reaction mechanism is composed of four steps:

$$O_2 + H_2O + e^- + * \rightarrow OOH^* + OH^- \quad (1)$$

$$OOH^* + e^- \rightarrow O^* + OH^- \quad (2)$$

$$O^* + H_2O + e^- \rightarrow OH^* + OH^- \quad (3)$$

$$OH^* + e^- \rightarrow OH^- + * \quad (4)$$

The conditions were T= 300 K, pH = 14, P_{H_2O} = 0.035 bar and P_{O_2} = 1 bar. Under these conditions, H_2O(g) and H_2O(l) were in thermodynamic equilibrium, and we used the following assumptions to calculate the overpotential. [1, 2]

Assumption S1: Computational hydrogen electrode

$$2(H^+ + e^-) \leftrightarrow H_{2(g)},\ \Delta G(U = 0\ V) = 0\ eV$$

Assumption S2: Corrections for entropy, zero-point vibrations, solvation and coverage effects

$$\Delta G_{O*} = \Delta E_{O*} + 0.01\ eV$$

$$\Delta G_{OH*} = \Delta E_{OH*} - 0.25\ eV$$

Assumption S3: Scaling relation

$$\Delta G_{OOH*} = \Delta G_{OH*} + 3.2\ eV$$

As a result, the Gibbs free energy changes for each step were calculated using the following equations (S1-S4):

$$\Delta G_1(U) = \Delta E_{OH} - 1.97 + U \quad (S1)$$

$$\Delta G_2(U) = \Delta E_O - \Delta E_{OH} - 2.94 + U \qquad (S2)$$

$$\Delta G_3(U) = \Delta E_{OH} - \Delta E_O - 0.26 + U \qquad (S3)$$

$$\Delta G_4(U) = U - \Delta E_{OH} + 0.25 \qquad (S4)$$

The activity (-overpotential) was calculated by means of the following equation:

$$\text{Activity} = - \max [\Delta G_1(U), \Delta G_2(U), \Delta G_3(U), \Delta G_4(U)]$$

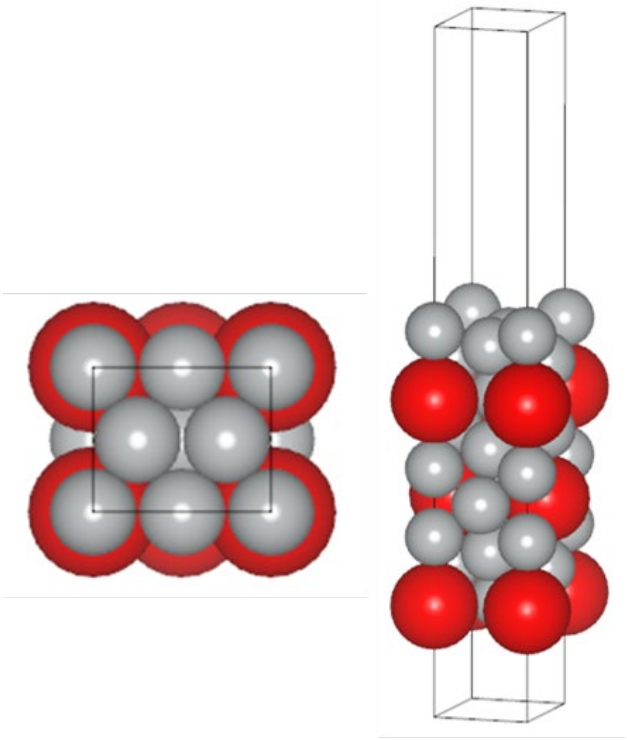

Figure S2. Structure of XAg_5 with P6/mmm (hexagonal) (X= Ba, Sr and Eu), which were the candidates proposed after high-throughput screening. The red atoms represent Ba, Sr or Eu, and the gray atoms represent Ag.