

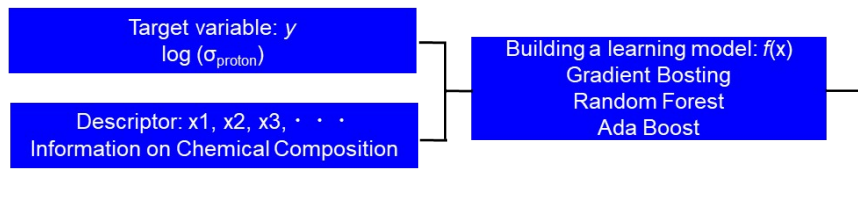
Prediction of proton conductivity using machine learning and exploration of proton-conductive ceramics

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Training



Prediction

Prediction of proton conductivity for virtual composition, $y_i=f(x_i)$

- Selection of matrix
- Selection of dopant

Verification

Experimental Verification
Proton conductivity measurement



Material exploration was conducted through the following steps:

1. Construction of a machine learning model to predict proton conductivity
2. Material selection based on predicted proton conductivity of hypothetical compositions
3. Synthesis of recommended materials and evaluation of their proton conductivity

Figure S1 Flow of learning model construction, prediction, and validation.

Dataset of proton conductivity and hole conductivity

In this study, the supervised machine learning model used experimental proton conductivities from 76 compositions and 726 samples (Table S1). Proton conductivity was determined by impedance measurements using the AC two-terminal method. All samples were dense discs and were measured under conditions where the water vapor pressure ranged from 0.01 to 0.019. The hole conductivity was primarily derived from reports of impedance measurements in air. The measurement and synthesis conditions for each sample are described in SI file: Supplementary Information 2.

Table S1 dataset of proton conductivity

Dataset 1 Dataset of II-IV type perovskite oxide						Dataset 2 Dataset of III-III type perovskite oxide					
composition	number	ref.	composition	number	ref.	composition	number	ref.	composition	number	ref.
BaCe _{0.45} Zr _{0.35} Y _{0.1} Yb _{0.1} O ₃	3		BaZr _{0.8} Lu _{0.2} O ₃	37	2	Ga _{0.9} Sr _{0.1} InO ₃	3		La _{0.84} Ba _{0.06} YbO ₃	10	
BaCe _{0.6} Zr _{0.2} Y _{0.2} O ₃	4	1	BaZr _{0.8} Sc _{0.2} O ₃	30	2	La _{0.9} Ba _{0.1} In _{0.9} Sc _{0.1} O ₃	3		La _{0.98} Ba _{0.02} YbO ₃	8	
BaCe _{0.7} Zr _{0.1} Y _{0.1} Yb _{0.1} O ₃	3		BaZr _{0.8} Tm _{0.2} O ₃	5	2	La _{0.9} Sr _{0.1} InO ₃	10	7	La _{0.9} Ba _{0.1} Yb _{0.9} In _{0.9} O ₃	7	
BaCe _{0.8} Y _{0.2} O ₃	5		BaZr _{0.8} Y _{0.2} O ₃	38	2	La _{0.9} Sr _{0.1} Y _{0.2} In _{0.8} O ₃	13		La _{0.9} Ba _{0.1} YbO ₃	10	10
Ba _{0.3} Sr _{0.7} Zr _{0.44} Ce _{0.38} Y _{0.22} O ₃	5		BaZr _{0.8} Yb _{0.2} O ₃	5	2	La _{0.9} Sr _{0.1} Yb _{0.2} In _{0.8} O ₃	11	8	La _{0.9} Ca _{0.1} YbO ₃	6	10
Ba _{0.5} Sr _{0.4} Zr _{0.8} Yb _{0.2} O ₃	4		BaZr _{0.9} Yb _{0.1} O ₃	7		La _{0.9} Sr _{0.1} Y _{0.5} In _{0.2} O ₃	11	7	La _{0.9} Mg _{0.1} YbO ₃	4	10
BaCe _{0.16} Zr _{0.84} Y _{0.1} Yb _{0.1} O ₃	3		CaZr _{0.95} Y _{0.05} O ₃	3	3	La _{0.9} Sr _{0.1} Y _{0.5} In _{0.8} O ₃	11		La _{0.9} Sr _{0.1} Y _{0.2} Yb _{0.8} O ₃	11	8
BaZr _{0.44} Ce _{0.38} Y _{0.2} O ₃	3		CaZr _{0.98} In _{0.02} O ₃	8		La _{0.9} Sr _{0.1} Y _{0.5} Yb _{0.2} O ₃	10	8	La _{0.9} Sr _{0.1} Yb _{0.9} In _{0.2} O ₃	11	8
BaZr _{0.55} Yb _{0.2} In _{0.25} O ₃	9		CaZr _{0.98} Yb _{0.02} O ₃	3	3	La _{0.9} Sr _{0.1} Y _{0.8} In _{0.2} O ₃	11		La _{0.9} Sr _{0.1} Yb _{0.9} In _{0.2} O ₃	11	9
BaZr _{0.5} Yb _{0.2} In _{0.3} O ₃	8		CaZr _{0.98} In _{0.02} O ₃	4		La _{0.9} Sr _{0.1} Y _{0.8} Yb _{0.2} O ₃	11	8	La _{0.9} Sr _{0.1} Yb _{0.9} In _{0.1} O ₃	11	8
BaZr _{0.9} Yb _{0.2} In _{0.3} O ₃	8		CaZr _{0.995} Al _{0.005} O ₃	12	4	La _{0.9} Sr _{0.1} Y _{0.3} O ₃	9	7	La _{0.9} Sr _{0.1} YbO ₃	11	7
BaZr _{0.8} Yb _{0.2} Lu _{0.2} O ₃	7		SrCe _{0.7} Yb _{0.3} O ₃	5		La _{0.9} Ba _{0.7} Yb _{0.9} In _{0.5} O ₃	3		LaBa _{0.02} Yb _{0.98} O ₃	9	
BaZr _{0.8} Yb _{0.2} Sc _{0.2} O ₃	8		SrCe _{0.85} Yb _{0.15} O ₃	11		La _{0.9} Ba _{0.5} Yb _{0.9} In _{0.5} O ₃	3		Nd _{0.9} Sr _{0.1} InO ₃	6	
BaZr _{0.7} Lu _{0.2} Sc _{0.1} O ₃	7		SrCe _{0.85} Yb _{0.15} O ₃	11		La _{0.7} Ba _{0.3} Yb _{0.9} In _{0.5} O ₃	3		Sm _{0.9} Sr _{0.1} InO ₃	6	
BaZr _{0.7} Sc _{0.3} O ₃	8		SrCe _{0.97} Yb _{0.03} O ₃	11							
BaZr _{0.7} Y _{0.2} Sc _{0.1} O ₃	7		SrCe _{0.98} Yb _{0.02} O ₃	11							
BaZr _{0.7} Yb _{0.2} Al _{0.1} O ₃	7		SrCe _{0.9} Sc _{0.05} Gd _{0.05} O ₃	3							
BaZr _{0.7} Yb _{0.2} In _{0.1} O ₃	8		SrCe _{0.9} Y _{0.1} O ₃	11	5						
BaZr _{0.7} Yb _{0.2} Lu _{0.1} O ₃	7		SrCe _{0.9} Yb _{0.1} O ₃	11							
BaZr _{0.7} Yb _{0.2} Sc _{0.1} O ₃	7		SrZr _{0.3} Ce _{0.6} Y _{0.1} O ₃	11	5						
BaZr _{0.7} Yb _{0.3} O ₃	8		SrZr _{0.4} Ce _{0.4} Y _{0.1} O ₃	11	6						
BaZr _{0.8} Er _{0.2} O ₃	32		SrZr _{0.7} Ce _{0.2} Y _{0.1} O ₃	11	5						
BaZr _{0.8} Gd _{0.2} O ₃	26		SrZr _{0.8} Sc _{0.2} O ₃	3							
BaZr _{0.8} In _{0.2} O ₃	33		SrZr _{0.8} Y _{0.1} O ₃	11							

Table S2 dataset of hole conductivity

composition	number	ref.	composition	number	ref.	composition	number	ref.
Ba _{0.3} Sr _{0.7} Co _{0.8} Fe _{0.2} O ₃	8	29	La _{0.3} Sr _{0.7} Co _{0.5} Ti _{0.5} O ₃	11	30	LaSc _{0.95} Mg _{0.05} O ₃	9	16
Ba _{0.3} Sr _{0.7} Zr _{0.44} Ce _{0.36} Y _{0.2} O ₃	4		La _{0.3} Sr _{0.7} Co _{0.7} Ti _{0.3} O ₃	11	30	LaSc _{0.9} Mg _{0.1} O ₃	9	16
Ba _{0.4} Sr _{0.6} Co _{0.8} Fe _{0.2} O ₃	8	29	La _{0.3} Sr _{0.7} TiO ₃	11	30	Nd _{0.4} Sr _{0.6} Co _{0.8} Fe _{0.2} O ₃	9	27
Ba _{0.5} Sr _{0.5} Co _{0.8} Fe _{0.2} O ₃	8	29	La _{0.4} Sr _{0.6} Co _{0.8} Fe _{0.2} O ₃	9	27	Nd _{0.9} Sr _{0.1} InO ₃	3	
Ba _{0.6} Sr _{0.4} Co _{0.8} Fe _{0.2} O ₃	8	29	La _{0.6} Sr _{0.4} Co _{0.2} Fe _{0.8} O ₃	5	23	Pr _{0.4} Sr _{0.6} Co _{0.8} Fe _{0.2} O ₃	9	27
Ba _{0.9} Sr _{0.1} Zr _{0.8} Yb _{0.2} O ₃	2		La _{0.6} Sr _{0.4} CoO ₃	7	22	Pr _{0.5} Sr _{0.5} Co _{0.2} Mn _{0.8} O ₃	9	11
BaCe _{0.16} Zr _{0.84} Y _{0.1} Yb _{0.1} O ₃	3		La _{0.75} Sr _{0.25} Cr _{0.5} Mn _{0.5} O ₃	8	28	Pr _{0.5} Sr _{0.5} Co _{0.4} Mn _{0.6} O ₃	9	11
BaCe _{0.45} Zr _{0.35} Y _{0.1} Yb _{0.1} O ₃	3		La _{0.75} Sr _{0.25} CrO ₃	8	28	Pr _{0.5} Sr _{0.5} Co _{0.6} Mn _{0.4} O ₃	9	11
BaCe _{0.6} Zr _{0.2} Y _{0.2} O ₃	3	1	La _{0.7} Ba _{0.3} CrO ₃	8	28	Pr _{0.5} Sr _{0.5} Co _{0.8} Mn _{0.2} O ₃	9	11
BaCe _{0.7} Zr _{0.1} Y _{0.1} Yb _{0.1} O ₃	3		La _{0.7} Ca _{0.3} CrO ₃	8	28	Pr _{0.5} Sr _{0.5} CoO ₃	9	11
BaCe _{0.8} Gd _{0.2} O ₃	5	14	La _{0.7} Sr _{0.3} CrO ₃	5	24	Pr _{0.5} Sr _{0.5} MnO ₃	9	11
BaCe _{0.8} Y _{0.2} O ₃	3		La _{0.7} Sr _{0.3} MnO ₃	7	18	Pr _{0.7} Sr _{0.3} Co _{0.2} Mn _{0.8} O ₃	9	11
BaCe _{0.9} Y _{0.1} O ₃	1		La _{0.85} Ca _{0.15} ScO ₃	9	16	Pr _{0.7} Sr _{0.3} Co _{0.4} Mn _{0.6} O ₃	9	11
BaPr _{0.6} Gd _{0.4} O ₃	5	14	La _{0.85} Sr _{0.15} CrO ₃	5	24	Pr _{0.7} Sr _{0.3} Co _{0.6} Mn _{0.4} O ₃	9	11
BaPr _{0.7} Gd _{0.3} O ₃	5	14	La _{0.8} Ca _{0.2} ScO ₃	9	16	Pr _{0.7} Sr _{0.3} Co _{0.8} Mn _{0.2} O ₃	9	11
BaPr _{0.8} Gd _{0.2} O ₃	5	14	La _{0.8} Sr _{0.2} CrO ₃	5	24	Pr _{0.7} Sr _{0.3} CoO ₃	9	11
BaZr _{0.44} Ce _{0.36} Y _{0.2} O ₃	2		La _{0.8} Sr _{0.2} Ga _{0.8} Mg _{0.15} Ni _{0.05} O ₃	6	15	Pr _{0.7} Sr _{0.3} MnO ₃	9	11
BaZr _{0.4} Pr _{0.4} Y _{0.2} O ₃	9	17	La _{0.8} Sr _{0.2} Ga _{0.8} Mg _{0.1} Ni _{0.1} O ₃	5	15	Pr _{0.85} Sr _{0.15} CoO ₃	5	25
BaZr _{0.5} Pr _{0.3} Y _{0.2} O ₃	9	17	La _{0.8} Sr _{0.2} Mn _{0.9} Cu _{0.1} O ₃	11	19	SrTi _{0.85} Fe _{0.15} O ₃	21	26
BaZr _{0.6} Pr _{0.2} Y _{0.2} O ₃	9	17	La _{0.8} Sr _{0.2} MnO ₃	12	21	SrTi _{0.95} Fe _{0.05} O ₃	21	26
BaZr _{0.7} Pr _{0.1} Y _{0.2} O ₃	9	17	La _{0.8} Sr _{0.2} Sc _{0.2} Mn _{0.8} O ₃	12	21	SrTi _{0.9} Fe _{0.1} O ₃	21	26
BaZr _{0.7} Yb _{0.2} In _{0.1} O ₃	3		La _{0.95} Ca _{0.05} ScO ₃	9	16	Sm _{0.4} Sr _{0.6} Co _{0.8} Fe _{0.2} O ₃	9	27
BaZr _{0.7} Yb _{0.2} Sc _{0.1} O ₃	1		La _{0.9} Ba _{0.1} In _{0.9} Sc _{0.1} O ₃	3		Sm _{0.9} Sr _{0.1} InO ₃	3	
BaZr _{0.8} In _{0.2} O ₃	3	2	La _{0.9} Ba _{0.1} YbO ₃	3	10	SrCe _{0.97} Yb _{0.03} O ₃	7	
BaZr _{0.8} Lu _{0.2} O ₃	3	2	La _{0.9} Ca _{0.1} ScO ₃	9	16	SrCe _{0.9} Sc _{0.05} Gd _{0.05} O ₃	3	
BaZr _{0.8} Sc _{0.2} O ₃	3	2	La _{0.9} Sr _{0.1} CoO ₃	7	22	SrCo _{0.85} Nb _{0.15} O ₃	9	12
BaZr _{0.8} Tm _{0.2} O ₃	3	2	La _{0.9} Sr _{0.1} CrO ₃	5	24	SrCo _{0.8} Nb _{0.2} O ₃	9	12
BaZr _{0.8} Y _{0.2} O ₃	3	2	La _{0.9} Sr _{0.1} ScO ₃	3	7	SrCo _{0.90} Nb _{0.1} O ₃	9	12
BaZr _{0.8} Yb _{0.2} O ₃	3	2	La _{0.9} Sr _{0.1} Yb _{0.8} In _{0.2} O ₃	1	9	SrCo _{0.95} Nb _{0.05} O ₃	9	12
Gd _{0.4} Sr _{0.6} Co _{0.8} Fe _{0.2} O ₃	9	27	LaFe _{0.6} Ni _{0.4} O ₃	11	20	SrZr _{0.3} Ce _{0.6} Y _{0.1} O ₃	1	5
Gd _{0.8} Ca _{0.2} CoO ₃	6	13	LaFe _{0.7} Ni _{0.3} O ₃	11	20	SrZr _{0.5} Ce _{0.4} Y _{0.1} O ₃	2	5
Gd _{0.9} Ca _{0.1} CoO ₃	6	13	LaFe _{0.8} Ni _{0.2} O ₃	11	20	SrZr _{0.8} Sc _{0.2} O ₃	3	
La _{0.1} Sr _{0.9} TiO ₃	11	30	LaFe _{0.9} Ni _{0.1} O ₃	7	20	SrZr _{0.9} Y _{0.1} O ₃	1	5
La _{0.2} Sr _{0.8} TiO ₃	11	30	LaSc _{0.85} Mg _{0.15} O ₃	9	16			
La _{0.3} Sr _{0.7} Co _{0.3} Ti _{0.7} O ₃	11	30	LaSc _{0.8} Mg _{0.2} O ₃	9	16			
						total	738	

Descriptors for Machine Learning

The 80 descriptors used in the machine learning model are listed below. They include the atomic weight, w , atomic density, ρ , melting temperature, T_m , first ionization energy, E_{1st} , electronegativity, χ , ionic radius, r , of the composed elements, where the values are divided or subtracted from one another and averaged over the A-site, B-site, and entire ABO_3 compound.

- 1) Temperature, T
- 2) Water partial pressure, p_{H_2O}
- 3) Sintering temperature, T_{sinter}
- 4) Sintering time, t_{sinter}
- 5) Product of descriptors 3) and 4)
- 6) Formula weight of perovskite oxides, M_w
- 7) Tolerance factor
- 8) Weighted average of w , ρ , T_m , E_{1st} , χ and r of A-site elements with its fraction in the A-site
- 9) Weighted average of w , ρ , T_m , E_{1st} , χ and r of B-site elements with its fraction in the B-site
- 10) Weighted average of w , ρ , T_m , E_{1st} , χ and r of dopant elements with its composition in perovskite
- 11) Weighted average of w , ρ , T_m , E_{1st} , χ and r of A-site host elements with its composition in perovskite
- 12) Weighted average of w , ρ , T_m , E_{1st} , χ and r of B-site host elements with its composition in perovskite
- 13) Sum of descriptors 11) and 12) for each w , ρ , T_m , E_{1st} , χ and r
- 14) Ratio of descriptors 8) to 9) for each w , ρ , T_m , E_{1st} , χ and r
- 15) Ratio of descriptors 12) to 11) for each w , ρ , T_m , E_{1st} , χ and r
- 16) Ratio of descriptors 10) to 11) for each w , ρ , T_m , E_{1st} , χ and r
- 17) Ratio of descriptors 10) to 12) for each w , ρ , T_m , E_{1st} , χ and r
- 18) Ratio of descriptors 10) to 13) for each w , ρ , T_m , E_{1st} , χ and r
- 19) Chemical composition of A-site dopant
- 20) Chemical composition of B-site dopant
- 21) Sum of descriptors 19) and 20)
- 22) Chemical composition of A-site host element
- 23) Chemical composition of B-site host element
- 24) Sum of descriptors 22) and 23)
- 25) $M_w (r_A r_B)^{-1/2}$

The “dopant” was defined as an acceptor whose valence is lower than that of the host element. In the case of $La_{0.9}^{3+}Sr_{0.1}^{2+}Yb_{0.5}^{3+}Y_{0.5}^{3+}O_{3-\delta}^{2-}$, for example, only Sr is considered as the dopant. We assumed the

valence of Nd (+2 or +3) and Yb (+2 or +3) cations to be +3. The Goldschmidt tolerance factor [31] was defined as $(r_A + r_O) / \{\sqrt{2}(r_B + r_O)\}$, where r_A , r_B , and r_O denote the Shannon ionic radii [32] of A-site cations, B-site cations, and oxygen ions, respectively. The descriptor 25) roughly expresses the theoretical density of perovskite oxides.

X-ray diffraction analysis

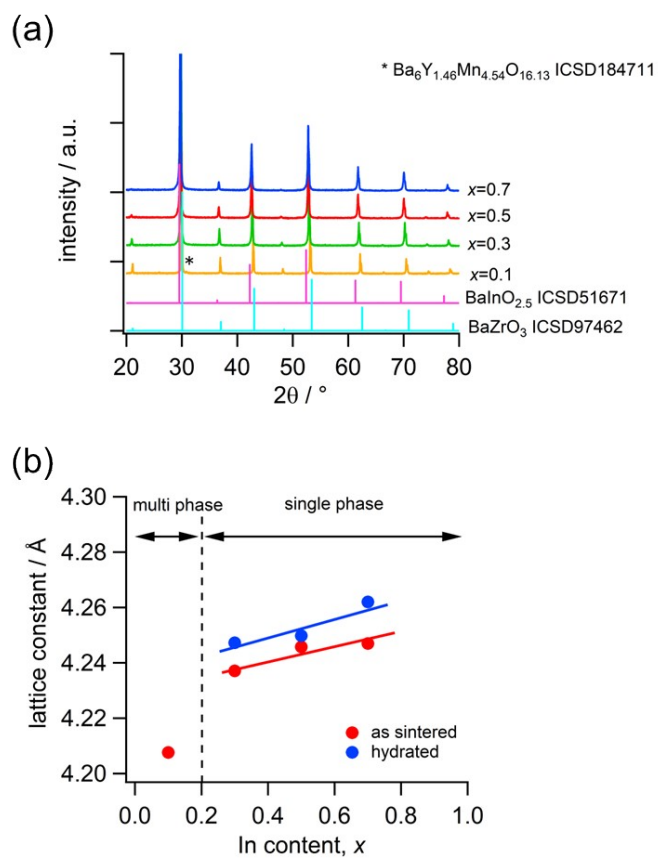


Figure S2 X-ray diffraction analysis. (a) X-ray diffraction pattern, (b) lattice constant.

X-ray Photoelectron Spectroscopy analysis

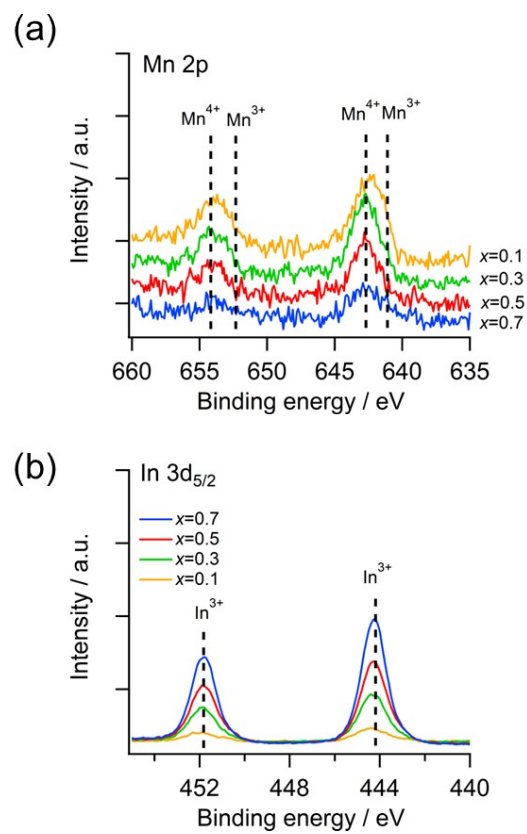


Figure S3 X-ray Photoelectron Spectroscopy analysis. (a) Mn 2p spectrum, (b) In 3d spectrum.

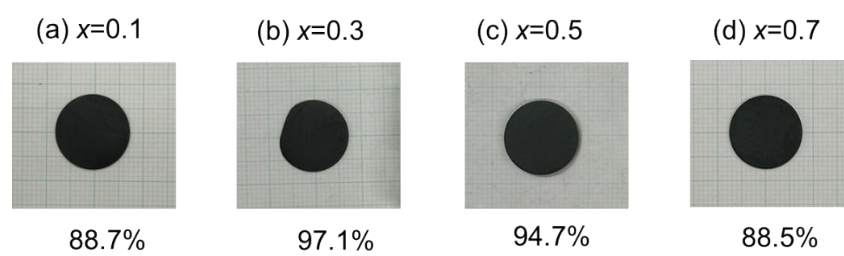


Figure S4 Photograph of samples. (a) $x=0.1$, (b) $x=0.3$, (c) $x=0.5$, (d) $x=0.7$.

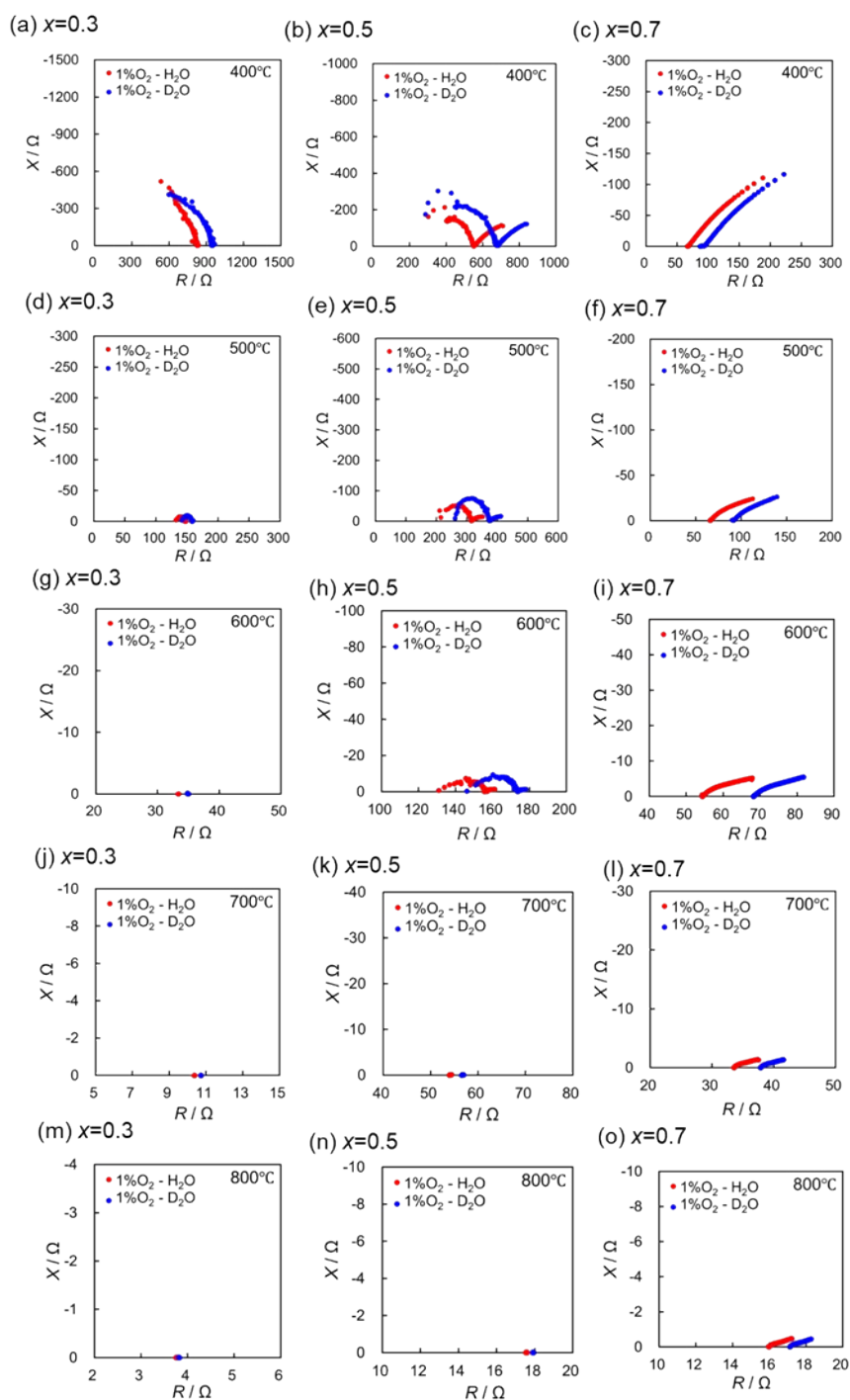


Figure S5 Nyquist plot of the sample for each temperature. (a)(b)(c)400°C, (d)(e)(f)500°C, (g)(h)(i)600°C, (j)(k)(l)700°C, (m)(n)(o)800°C.

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