Supporting Information

Water splitting over transition metal-doped $SrTiO_3$ photocatalysts with response to visible light up to 660 nm

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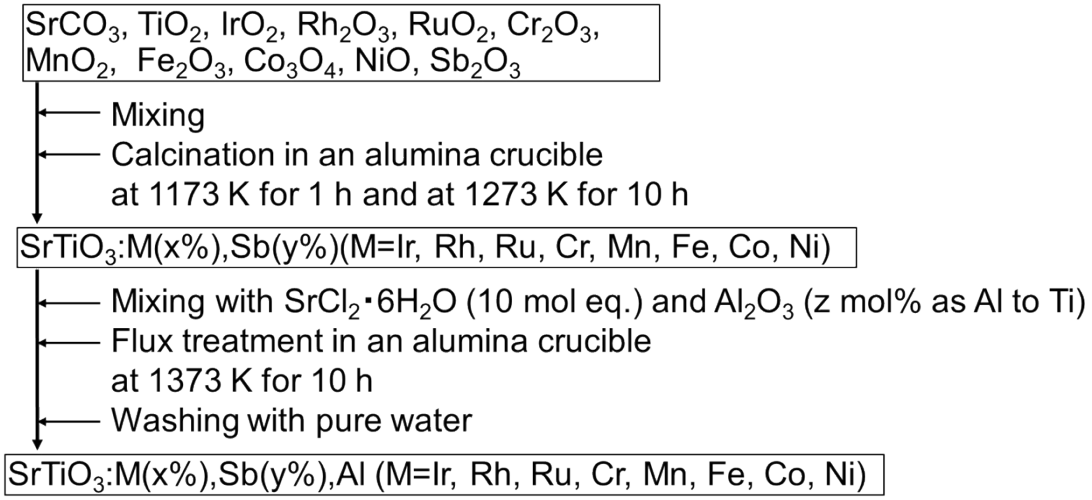

Figure S1. Flowchart of preparation of $SrTiO_3$:M(x%),Sb(y%),Al (M=Ir, Rh, Ru, Cr, Mn, Fe, Co, Ni) by the $SrCl_2$-flux treatment with/without Al_2O_3 addition after SSR.

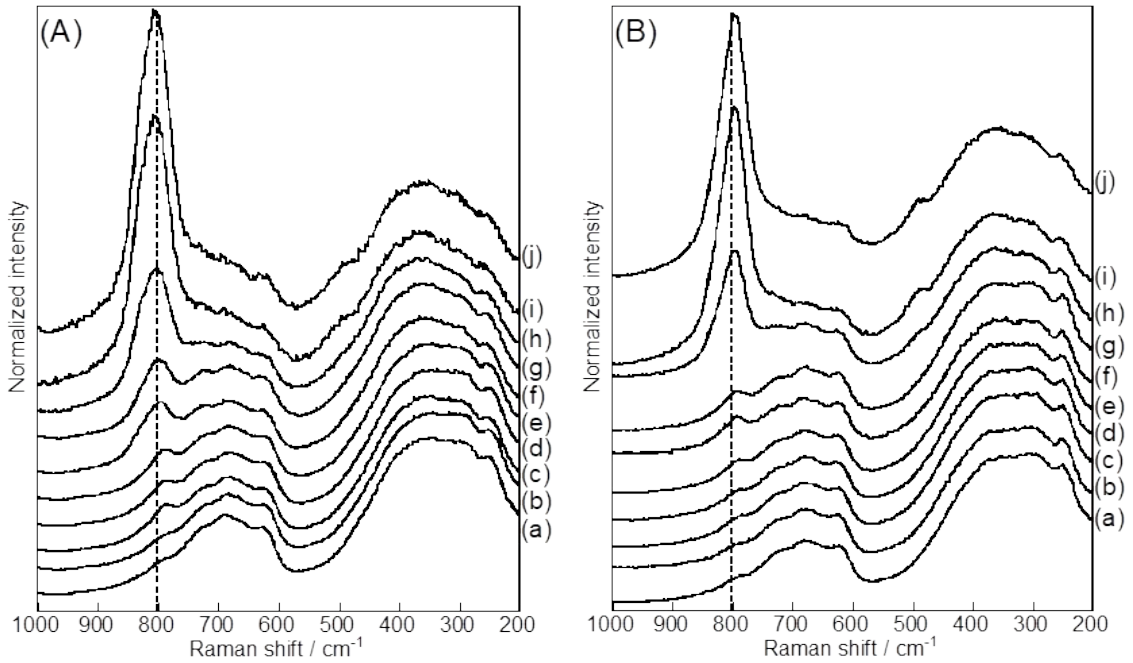

Figure S2. Raman spectra of SSR-prepared $SrTiO_3$:Ir(x%),Sb(2x%),Al with the flux treatment without Al_2O_3 addition. Samples were doped with (a) x=0, (b) x=0.001, (c) x=0.008, (d) x=0.009, (e) x=0.01, (f) x=0.025, (g) x=0.05, (h) x=0.1, (i) x=0.2, (j) x=0.3. Excitation wavelengths of (A) and (B) were 532 nm and 785 nm, respectively.

Table S1. Molar ratio of Ir and Sb in SSR-prepared $SrTiO_3$:Ir,Sb with or without the flux treatment without Al_2O_3 addition determined by XRF

Photocatalyst	$SrCl_2$-flux treatment	Molar ratio %	
		[Ir]/([Ti]+[Ir]+[Sb])	[Sb]/([Ti]+[Ir]+[Sb])
$SrTiO_3$	×	0	0
$SrTiO_3$:Ir(0.025%),Sb(0.05%)	×	Trace	Trace
$SrTiO_3$:Ir(0.05%),Sb(0.1%)	×	0.016	0.078
$SrTiO_3$:Ir(0.1%),Sb(0.2%)	×	0.046	0.16
$SrTiO_3$:Ir(0.2%),Sb(0.4%)	×	0.13	0.31
$SrTiO_3$:Ir(0.3%),Sb(0.6%)	×	0.26	0.52
$SrTiO_3$	○	0	0
$SrTiO_3$:Ir(0.025%),Sb(0.05%)	○	Trace	Trace
$SrTiO_3$:Ir(0.05%),Sb(0.1%)	○	0.015	0.081
$SrTiO_3$:Ir(0.1%),Sb(0.2%)	○	0.047	0.16
$SrTiO_3$:Ir(0.2%),Sb(0.4%)	○	0.14	0.37
$SrTiO_3$:Ir(0.3%),Sb(0.6%)	○	0.24	0.49

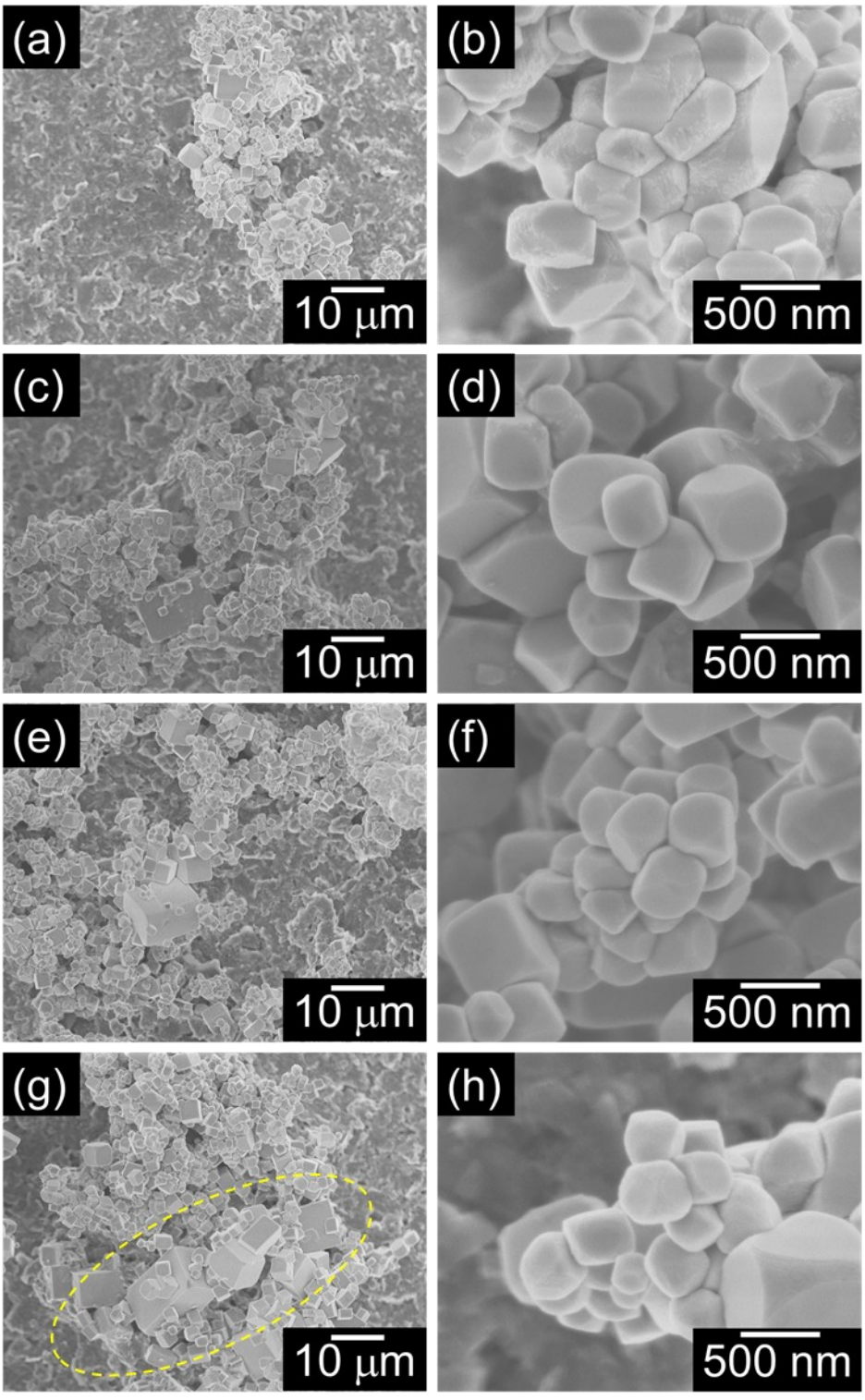

Figure S3. SEM images of SSR-prepared $SrTiO_3$:Ir(0.009%),Sb(y%) with the flux treatment without Al_2O_3 addition. Samples were doped with (a) y=0, (c) y=0.009, (e) x=0.018, (g) x=0.027. (b), (d), (f), (h) are the magnified images of (a), (c), (e), (g), respectively.

Table S2. Effect of an amount of Al_2O_3 addition in the flux on photocatalytic water splitting over $SrTiO_3$:Ir(0.009%),Sb(0.018%),Al prepared by the flux treatment after SSR under visible light irradiation.

Amount of Al_2O_3 addition (mol% as Al to Ti)	Water splitting activity/μmol h^{-1}	
	H_2	O_2
0	41	21
0.5	47	23
1	62	30
1.5	56	26
2	60	28
3	53	25
5	50	23

Photocatalyst: 0.2 g, cocat.: $RhCrO_x$(Rh 0.1 mol%, Cr 0.1 mol%) impregnation, reactant solution: Pure water (120 mL), light source: a 300 W Xe lamp (λ > 440 nm), cell: a top-irradiation cell with a Pyrex window, system: a gas-tight circulation system.

Table S3. Photocatalytic water splitting under visible light irradiation over various cocatalysts-loaded $SrTiO_3$:Ir(0.009%),Sb(0.018%),Al prepared by the flux treatment with Al_2O_3 (1%) addition after SSR

Entry	Cocatalyst (mol% as metal)	Water splitting activity/μmol h^{-1}	
		H_2	O_2
1	none	0.01	0.005
2	$RhCrO_x$ (0.05)	46	21
3	$RhCrO_x$ (0.08)	54	26
4	$RhCrO_x$ (0.1)	62	30
5	$RhCrO_x$ (0.2)	18	8.8
6	IrO_2 (0.2)	0.009	0
7	CoO_x (0.9)	0.05	0.02
8	RuO_2 (0.5)	0.5	0.2
9	Rh_2O_3 (0.1)	12	5.6
10	Cr_2O_3 (0.1)	0.13	0.07

Photocatalyst: 0.2 g, cocatalyst: $RhCrO_x$, Rh_2O_3, and Cr_2O_3 (Impregnation in air at 623 K for 1 h), IrO_2, CoO_x, and RuO_2 (Impregnation in air at 673 K for 2 h), reactant solution: pure water (120 mL), light source: a 300 W Xe lamp ($\lambda > 440$ nm), cell: a top-irradiation cell with a Pyrex window, system: a gas-tight circulation system.

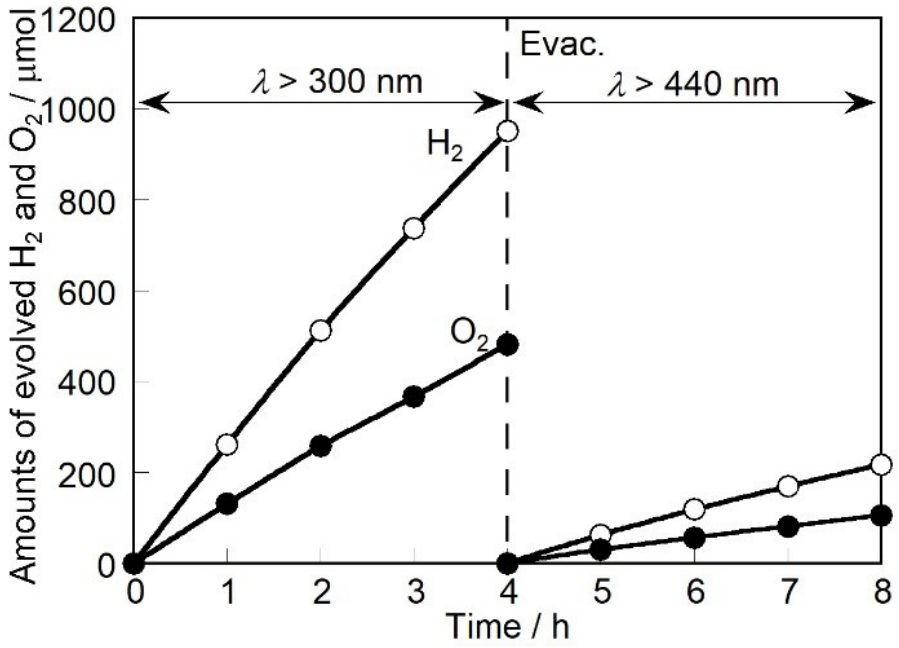

Figure S4. Photocatalytic water splitting over $RhCrO_x$-loaded $SrTiO_3$:Ir(0.009%),Sb(0.018%),Al prepared by the flux treatment with Al_2O_3 (1%) addition after SSR. Photocatalyst: 0.2 g, cocat.: $RhCrO_x$ (Rh 0.1 mol%, Cr 0.1 mol%) impregnation, reactant solution: pure water (120 mL), light source: a 300 W Xe lamp, system: a gas-tight circulation system.

Table S4. Responsive wavelength, AQY and STH of various transition metals-doped $SrTiO_3$:Al and representative visible-light-driven single particulate photocatalysts for overall water splitting.

Photocatalyst	Responsive wavelength/nm	AQY % at 420 nm	STH %	
			Full[a]	Vis[b] (λ>440 nm)
$SrTiO_3$:Ir(0.009%),Sb(0.018%)	660	0.73	0.33	0.030
$SrTiO_3$:Rh(0.02%),Sb(0.04%)	570	0.31	0.16	0.020
$SrTiO_3$:Ru(0.03%),Sb(0.06%)	600	0.33	0.16	0.019
$SrTiO_3$:Cr(0.01%)	660	—	0.31	0.0093

[a]STH (%) (Full)

= (100 × ([$\Delta G^o(H_2O)$/J mol^{-1}] × [rate of H_2 evolution(under full arc from a solar simulator)/mol h^{-1}]))/([3600 / s h^{-1}] × [solar energy/W cm^{-2}] × [irradiation area/cm^2])

[b]STH (%) (Vis(λ>440 nm))

= (100 × ([$\Delta G^o(H_2O)$/J mol^{-1}] × [rate of H_2 evolution(under visible light from a solar simulator)/ mol h^{-1}]))/([3600/s h^{-1}] × [solar energy/W cm^{-2}] × [irradiation area/cm^2])