Supporting Information for

Reaction between Strontium and steam in the primary circuit of HTR-PM: A theoretical investigation

Jingni Guo[1], Yu Wang[1], Wei Peng[1], Feng Xie[1,], Peng Li[2], Minghua Lyu[3]*

[1]Institute of Nuclear and New Energy Technology, Collaborative Innovation Center of Advanced Nuclear Energy Technology, Key Laboratory of Advanced Reactor Engineering and Safety of Ministry of Education, Tsinghua University, Beijing 100084, China

[2]School of Physics and Electronics Engineering, Shanxi University, Taiyuan 030006, China

[3]Dapartment of Nuclear Environmental Science Research, China Institute for Radiation Protection, Taiyuan 030006, China

**Corresponding author, Email: fxie@tsinghua.edu.cn*

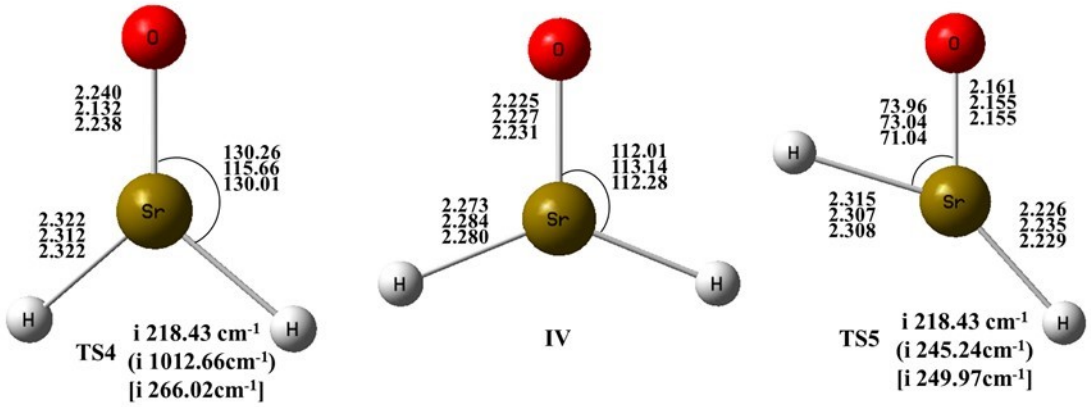

Fig. S 1 Structures and selected parameters of stationary points on the $SrO+H_2$ PES, which were optimized at different levels of theory. Bond distances are in Å, and angles are in degrees. (B3PW91/SDD/6-311G**, B3LYP/Def2QZVPP/6-311G**, B3LYP/Def2TZVPP/6-311G**, from top to bottom rows, respectively)

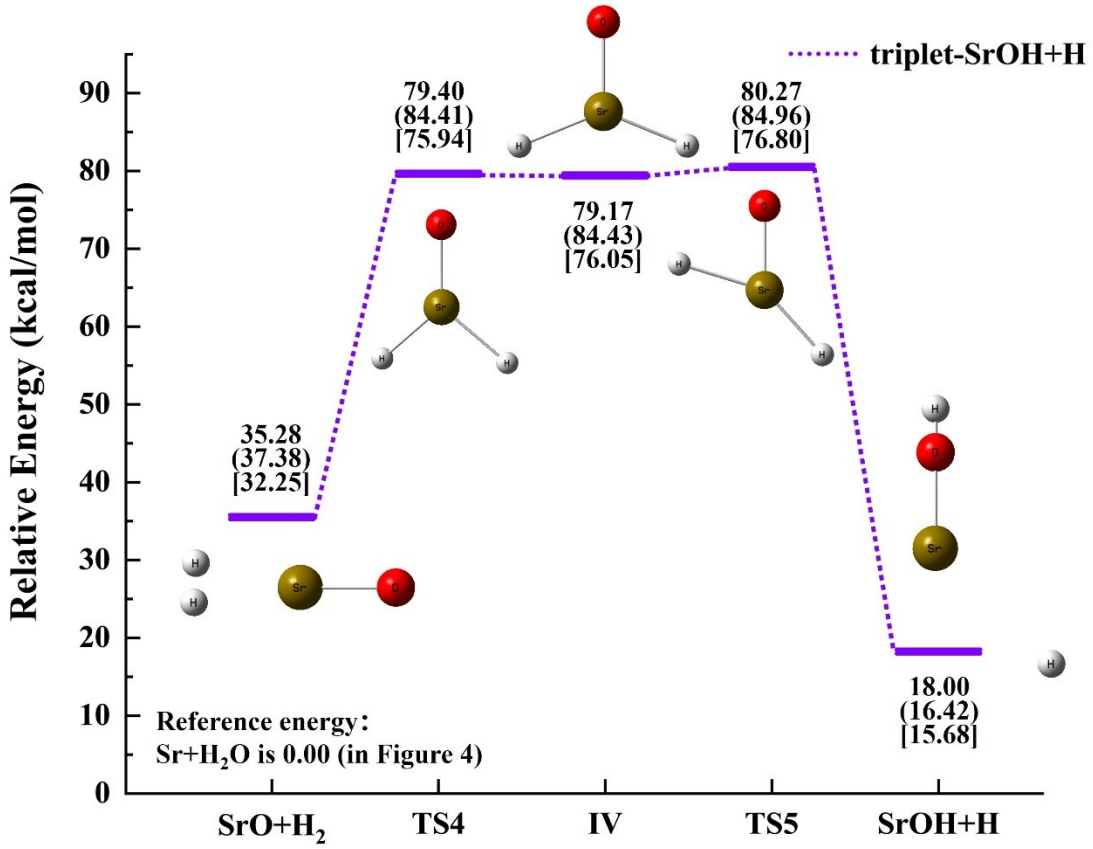

Fig. S 2 Potential energy profile for the reaction of SrO + H_2 computed at the B3LYP/Def2TZVPP/6-311G** levels of theory in triplet. B3PW91/SDD/6-311G** and B3LYP/Def2QZVPP/6-311G** energy parameters are reported in ( ) and [ ] respectively.

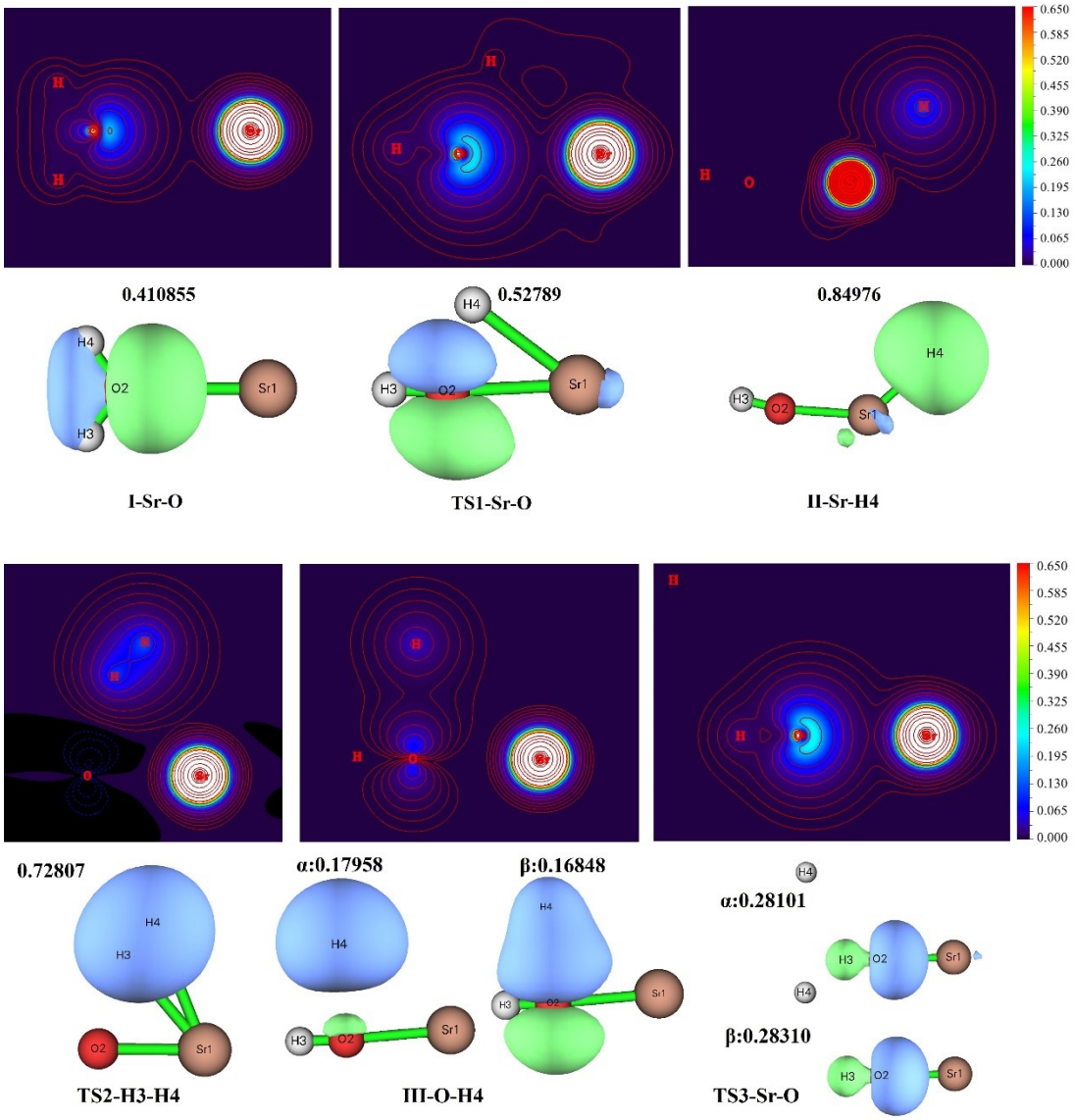

Fig. S 3 BOD map and NAdO of Sr+H_2O

Fig. S3 shows the bond order density (BOD) diagram and the natural adaptation orbital (NAdO) diagram for the Sr+H_2O reaction process. The numbers in the figure are the eigenvalues of the highest occupied NAdO orbitals, with higher values representing larger contributions. In TS1, Sr-H4 is not yet bonded, and in Compound II, Sr-H4 tends to form a single bond. In TS2, molecule H_2 is close to forming and H3-H4 shows strong covalent interactions.

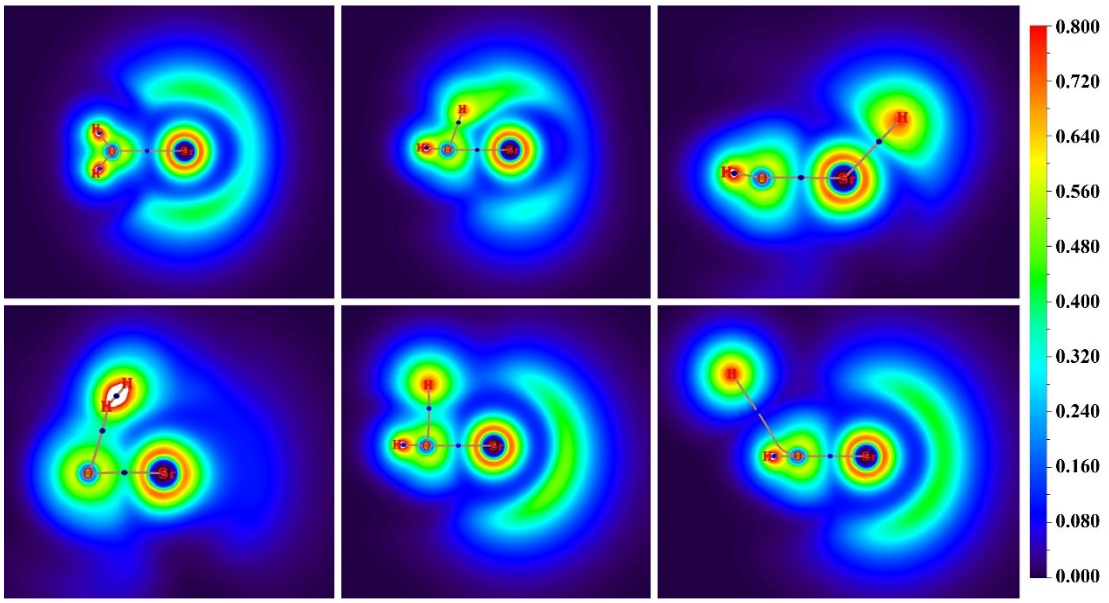

Fig. S 4 LOL color filled map of the stationary points on the Sr+H_2O reaction pathway computed at the B3PW91/SDD/6-311G** levels of theory (Dark blue balls are the (3, -1) BCPs, brown lines are the paths connecting (3, -3) and (3, -1))

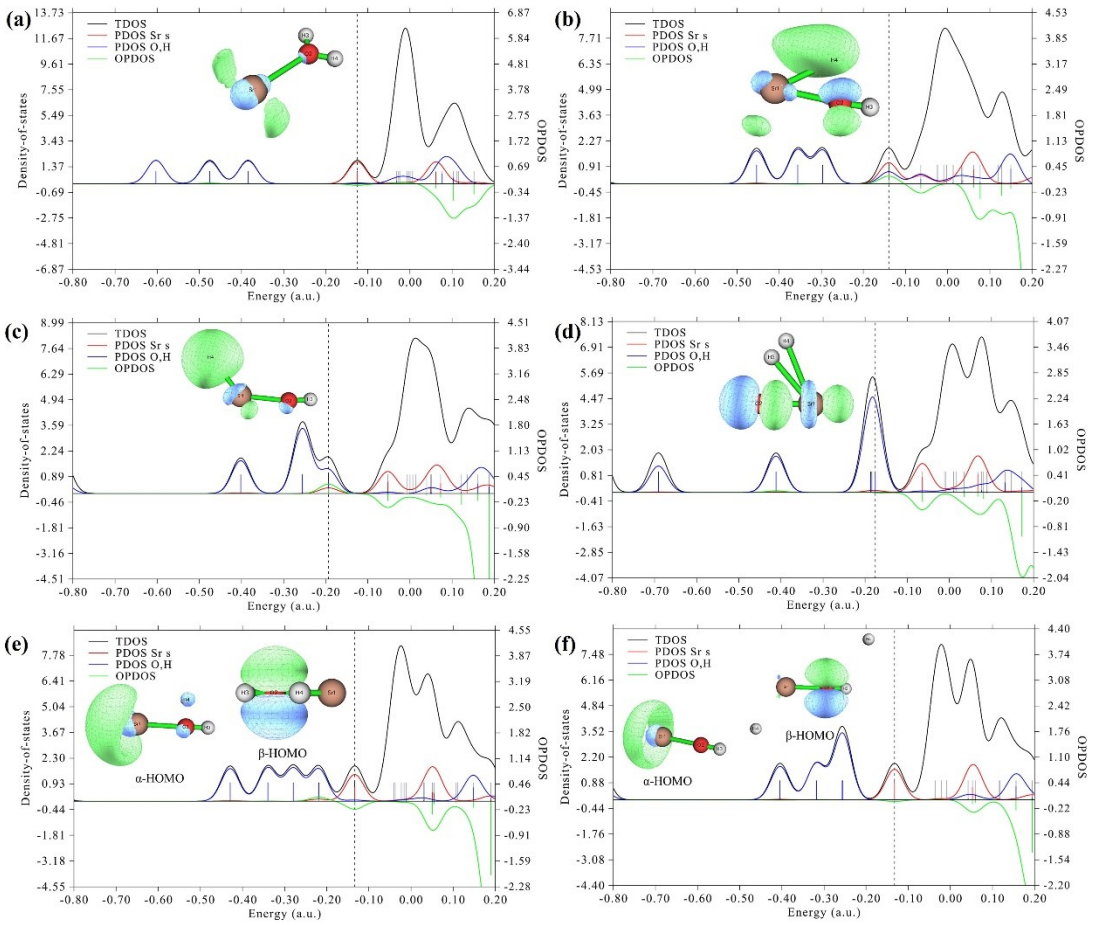

Fig. S 5 The TDOS, PDOS (s orbital and O, H atom), and OPDOS (s orbital with O, H atom) curves of the stationary points on the $Sr+H_2O$ reaction pathway at the B3PW91/SDD/6-311G** levels of theory (a) compound I (b) TS1 (c) compound II (d) TS2 (e) compound III (f) TS3

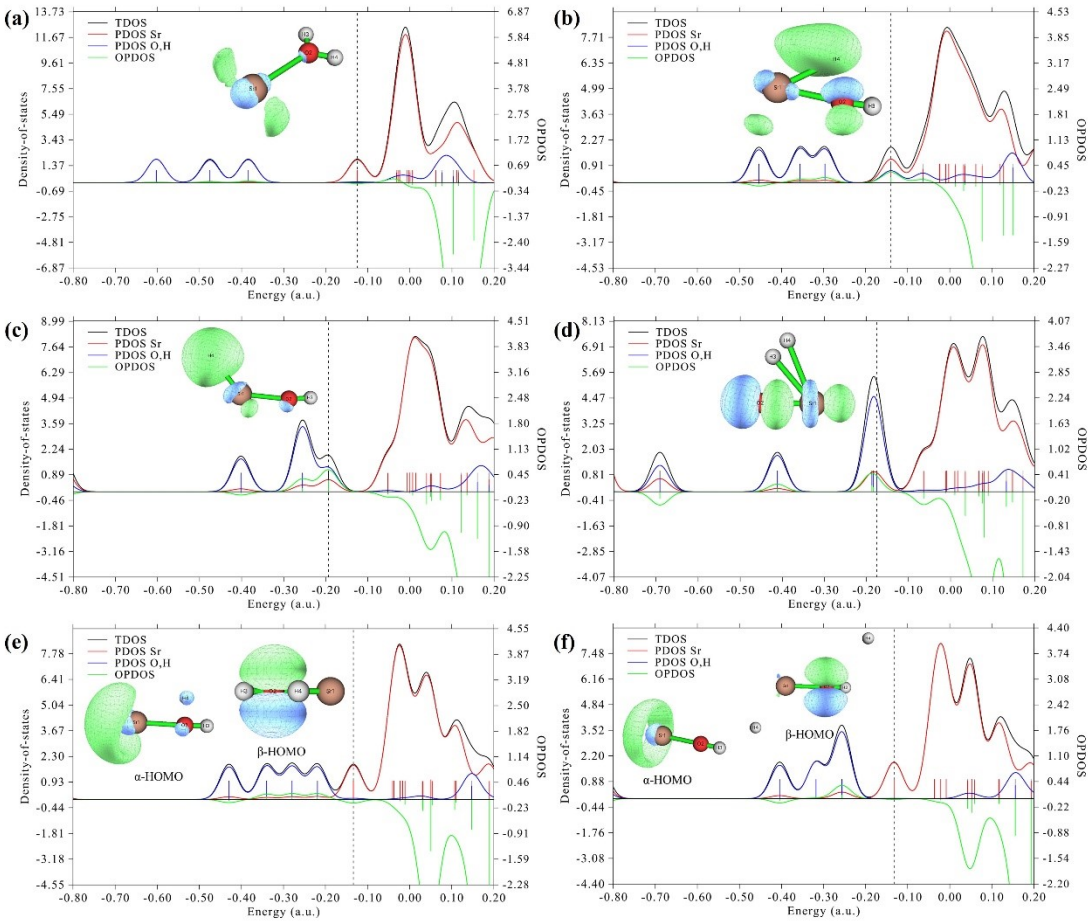

Fig. S 6 The TDOS, PDOS (Sr atom and O, H atom), and OPDOS (Sr atom with O, H atom) curves of the stationary points on the Sr+H_2O reaction pathway at the B3PW91/SDD/6-311G** levels of theory (a) compound I (b) TS1 (c) compound II (d) TS2 (e) compound III (f) TS3

Table S 1 Comparison of experiment values and calculated bond lengths and harmonic frequencies for SrO and SrOH.

	SrO		SrOH				
Method	r(Sr-O)	Frequencies	r(Sr-O)	Frequencies			
Expt	1.920	653.5	2.111	363.7	363.7	527.0	3766±10
B3LYP[a]	1.948	669.0	2.113	420.0	420.0	552.2	3941.8
B3LYP-D3[a]	1.948	669.1	2.113	419.6	419.6	553.6	3944.0
B3LYP[b]	1.923	675.8	2.101	358.9	358.9	549.9	3933.6
B3LYP-D3[b]	1.923	675.9	2.100	358.6	358.6	551.3	3935.8
B3LYP[c]	1.927	668.6	2.103	385.5	385.5	547.6	3939.4
B3LYP-D3[c]	1.927	668.7	2.103	385.1	385.1	549.0	3941.7
PBE[a]	1.955	649.6	2.104	396.3	396.3	550.3	3842.8
PBE-D3[a]	1.955	649.7	2.104	396.2	396.2	550.9	3843.7
PBE[b]	1.929	656.9	2.091	336.1	336.1	549.0	3833.9
PBE-D3[b]	1.929	656.9	2.090	336.0	336.0	549.6	3834.8
PBE[c]	1.934	648.8	2.093	363.8	363.8	546.2	3840.0
PBE-D3[c]	1.934	648.8	2.093	363.8	363.8	546.8	3840.8
PBE0[a]	1.925	695.5	2.100	430.7	430.7	562.0	4005.0
PBE0-D3[a]	1.925	695.5	2.100	430.9	430.9	562.4	4005.6
PBE0[b]	1.899	701.7	2.085	373.8	373.8	559.5	3996.0
PBE0-D3[b]	1.899	701.8	2.086	374.1	374.1	560.0	3996.6
PBE0[c]	1.904	694.3	2.088	396.8	396.8	557.8	4002.2
PBE0-D3[c]	1.904	694.3	2.088	397.0	397.0	558.3	4002.7
B3PW91[a]	1.929	691.8	2.103	425.4	425.4	558.1	3976.8
B3PW91-D3[a]	1.929	691.9	2.103	425.5	425.5	559.2	3978.4
B3PW91[b]	1.906	696.7	2.089	369.4	369.4	556.0	3967.9
B3PW91-D3[b]	1.906	696.7	2.089	369.6	369.6	557.2	3969.4
B3PW91[c]	1.910	689.8	2.091	392.3	392.3	554.2	3974.1
B3PW91-D3[c]	1.910	689.8	2.091	392.5	392.5	555.4	3975.7

[a] Calculations used the SDD for Sr and 6-311G** for H and O atoms.

[b] Calculations used the Def2QZVPP for Sr and 6-311G** for H and O atoms.

[c] Calculations used the Def2TZVPP for Sr and 6-311G** for H and O atoms.

Table S 2 Optimized cartesian x, y, z coordinates for the reaction of Sr+H_2O at the B3LYP/Def2TZVPP/6-311G** level of theory.

	atom	x	y	z
I（1A）	Sr	-0.00000400	-0.54558200	0.00000000
	O	-0.00000400	1.95855600	0.00000000
	H	-0.78292700	2.53196000	0.00000000
	H	0.78310900	2.53171000	0.00000000
TS1（1A）	Sr	-0.46372100	-0.00774000	-0.00000500
	O	1.70754800	-0.11865700	0.00007000
	H	2.66870000	-0.08885600	-0.00039800
	H	1.29232100	1.33221900	0.00002200
II（1A）	Sr	-0.38453700	-0.06386800	0.00002400
	O	1.73914800	0.07857900	-0.00027000
	H	2.66359300	0.32543500	0.00142300
	H	-1.96437200	1.47292500	-0.00016000
TS2（1A）	Sr	0.39084100	-0.01904300	-0.00001100
	O	-1.56118200	-0.32201200	0.00002900
	H	-1.40094300	1.28825400	0.00007900
	H	-0.96156100	2.01147700	0.00009200
I（3A）	Sr	0.00799400	-0.52858300	0.00000000
	O	0.00799400	1.89911000	0.00000000
	H	-0.93556500	2.20111300	0.00000000
	H	0.56786200	2.69214800	0.00000000
TS1（3A）	Sr	-0.48399900	-0.00414500	-0.00001300
	O	1.76007400	-0.11263700	0.00005000
	H	2.71907200	-0.15606100	0.00007700
	H	1.59228100	1.21467600	0.00000300

III(3A）	Sr	-0.46944800	0.00087500	-0.00003900
	O	1.67209400	-0.19595300	0.00015600
	H	2.62943500	-0.22027000	0.00023000
	H	1.83284500	1.75465000	0.00001900
TS2（3A）	Sr	-0.42533500	-0.03324200	-0.00000500
	O	1.69349900	-0.69052900	0.00002200
	H	1.67203100	3.31742000	0.00001400
	H	0.94269200	3.47002900	0.00001100
TS3（3A）*	Sr	-0.50143500	0.04838500	-0.00004800
	O	1.55957200	-0.37267500	0.00014800
	H	2.49374800	-0.56855900	0.00023700
	H	4.08421400	1.71134400	0.00040400

*Calculated results at B3PW91/SDD/6-311G**

Table S 3 Electric energy and G(T) of Sr atom, Compound I, Compound II and SrO with relativistic effects

		SR (Hartree)	SR + SO* (Hartree)	SO contribution (kcal/mol)
Sr	Electric energy	-30.6953641	-30.6971598	-1.13
	G(T)	-30.7116920	-30.7134880	-1.13
Compound I	Electric energy	-107.1651081	-107.1669033	-1.13
	G(T)	-107.1692800	-107.1710750	-1.13
Compound II	Electric energy	-107.1917527	-107.1935670	-1.14
	G(T)	-107.2026480	-107.2044620	-1.14
SrO	Electric energy	-105.9359248	-105.9377686	-1.16
	G(T)	-105.9570270	-105.9588710	-1.16

* The calculation was used B3LYP/dhf-TZVPP-2c/6-311G**

Table S 4 Structures of Compound I, Compound II and SrO with relativistic effects

		SR	SR + SO	SO contribution
Compound I	R(1,2)	2.5026	2.5026	0.00E+00
	R(2,3)	0.9709	0.9709	0.00E+00
	R(2,4)	0.9709	0.9709	0.00E+00
	A(1,2,3)	126.2099	126.2094	-5.00E-04
	A(1,2,4)	126.1573	126.1569	-4.00E-04
	A(3,2,4)	107.6327	107.6337	1.00E-03
Compound II	R(1,2)	2.125	2.1249	-1.00E-04
	R(1,4)	2.2059	2.2058	-1.00E-04
	R(2,3)	0.9573	0.9573	0.00E+00
	A(2,1,4)	132.8663	132.8432	-2.31E-02
	A(1,2,3)	168.521	168.5149	-6.10E-03
SrO	R(1,2)	1.9216	1.9215	-1.00E-04

* The calculation was used B3LYP/dhf-TZVPP-2c/6-311G**

Table S 5 Atomic charge analysis based Mulliken method and ADCH method of Sr + H_2O computed at the B3PW91/SDD/6-311G** levels of theory

Compound	Atom	ADCH	Mulliken				
		Atomic charge	Population of angular moment orbitals			Population	Net charge
			s	p	d		
Compound I	Sr1	-0.1779	3.9064	6.0502	0.1117	10.0683	-0.0683
	O2	-0.4946	3.7376	4.7498	0.0082	8.4956	-0.4956
	H3	0.3362	0.6544	0.0636	0.0000	0.7180	0.2820
	H4	0.3362	0.6544	0.0636	0.0000	0.7180	0.2820
TS1	Sr1	0.4454	3.2090	6.0505	0.3709	9.6305	0.3695
	O2	-0.4696	3.8091	4.8341	0.0050	8.6482	-0.6482
	H3	0.3138	0.6517	0.0640	0.0000	0.7157	0.2843
	H4	-0.2896	0.9885	0.0171	0.0000	1.0056	-0.0056
Compound II	Sr1	1.3622	2.3533	6.2538	0.5270	9.1340	0.8660
	O2	-1.0736	3.8169	4.9912	0.0042	8.8122	-0.8122
	H3	0.4191	0.6778	0.0657	0.0000	0.7435	0.2565
	H4	-0.7077	1.3083	0.002	0.0000	1.3103	-0.3103
TS2	Sr1	0.9328	2.1550	6.1738	0.8706	9.1993	0.8007
	O2	-0.8202	3.9526	4.8314	0.0033	8.7874	-0.7874
	H3	-0.0351	0.8010	0.0241	0.0000	0.8251	0.1749
	H4	-0.0775	1.1788	0.0094	0.0000	1.1882	-0.1882
Compound III	Sr1	0.2373	2.8496	6.2604	0.3261	9.4360	0.56397
	O2	-0.4873	3.8172	4.9335	0.0043	8.7550	-0.75501
	H3	0.3029	0.6528	0.0645	0.0000	0.7173	0.28271
	H4	-0.0529	1.0873	0.0044	0.0000	1.0917	-0.09168
TS3	Sr1	0.2539	2.8707	6.1960	0.3896	9.4563	0.5437
	O2	-0.5511	3.8127	4.9816	0.0041	8.7984	-0.7984
	H3	0.2840	0.6774	0.0663	0.0000	0.7437	0.2563
	H4	0.0133	1.0016	0.0001	0.0000	1.0017	-0.0017

For 2-parameters fit:

$$k_{Arrhenius} = A\exp\left(-\frac{E_a}{RT}\right)\#(1)$$

For 3-parameters fit:

$$k_{Arrhenius} = A\mathrm{T}^n \exp\left(-\frac{E_a}{RT}\right)\#(2)$$

Table S 6 Arrhenius fitting parameters of rate constants

		A	E_a(KJ/mol)	A	E_a(KJ/mol)	n
I→TS1→II	k_{VTST}	3.53E+11	99.17	4.08E+19	113.79	-2.39
	k_{VTST}-Wigner	3.07E+11	96.89	8.45E+18	110.38	-2.21
	k_{VTST}-Eckart	2.60E+11	95.16	1.01E+17	105.29	-1.66
II→TS2→P	k_{VTST}	2.10E+12	269.62	9.10E+13	272.59	-0.49
	k_{VTST}-Wigner	2.05E+12	269.29	6.29E+13	271.98	-0.44
	k_{VTST}-Eckart	1.98E+12	269.25	4.05E+13	271.63	-0.39