

Supporting information

-for-

Enhancing SO₃ Hydrolysis and Nucleation: The Role of Formic Sulfuric Anhydride

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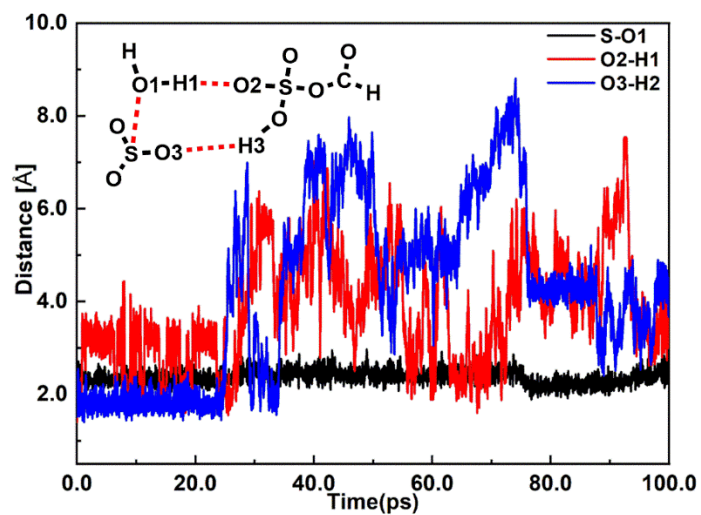
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S34	Fig. S13. (a) Cluster formation rate (J , cm ⁻³ s ⁻¹) and (b) enhancement factor (R) as a function of [A] with [FSA] = 10 ⁶ molecules·cm ⁻³ and five different [SA] concentrations (black: [SA] = 10 ⁴ molecules·cm ⁻³ ,

	red: $[SA] = 10^5 \text{ molecules}\cdot\text{cm}^{-3}$, blue: $[SA] = 10^6 \text{ molecules}\cdot\text{cm}^{-3}$, green: $[SA] = 10^7 \text{ molecules}\cdot\text{cm}^{-3}$, purple: $[SA] = 10^8 \text{ molecules}\cdot\text{cm}^{-3}$) at 298.15 K
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17 **Fig. S1.** BOMD simulation trajectories and snapshots of the FSA-assisted SO_3 hydrolysis reaction

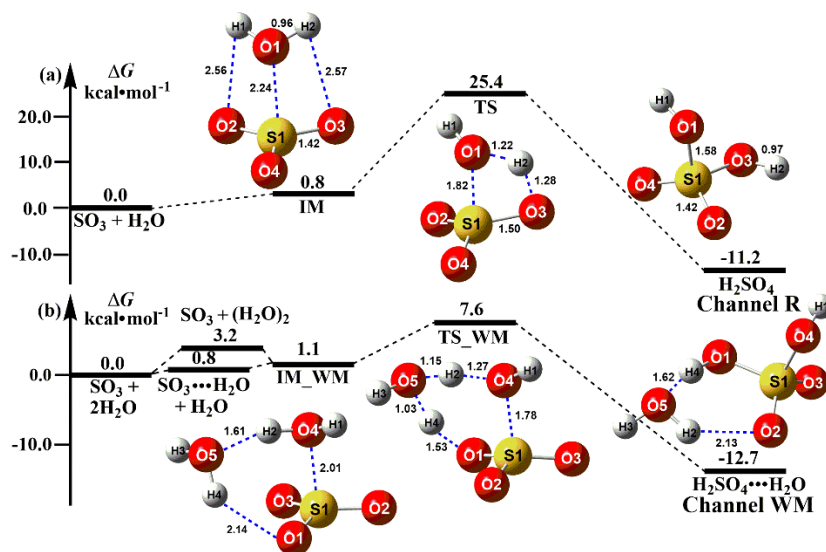


Fig. S2. Energy diagrams for H₂SO₄ formation from the SO₃ + H₂O reaction with and without H₂O at the CCSD(T)-F12/cc-pVDZ-F12//M06-2X/6-311++G(2df,2pd) level of theory

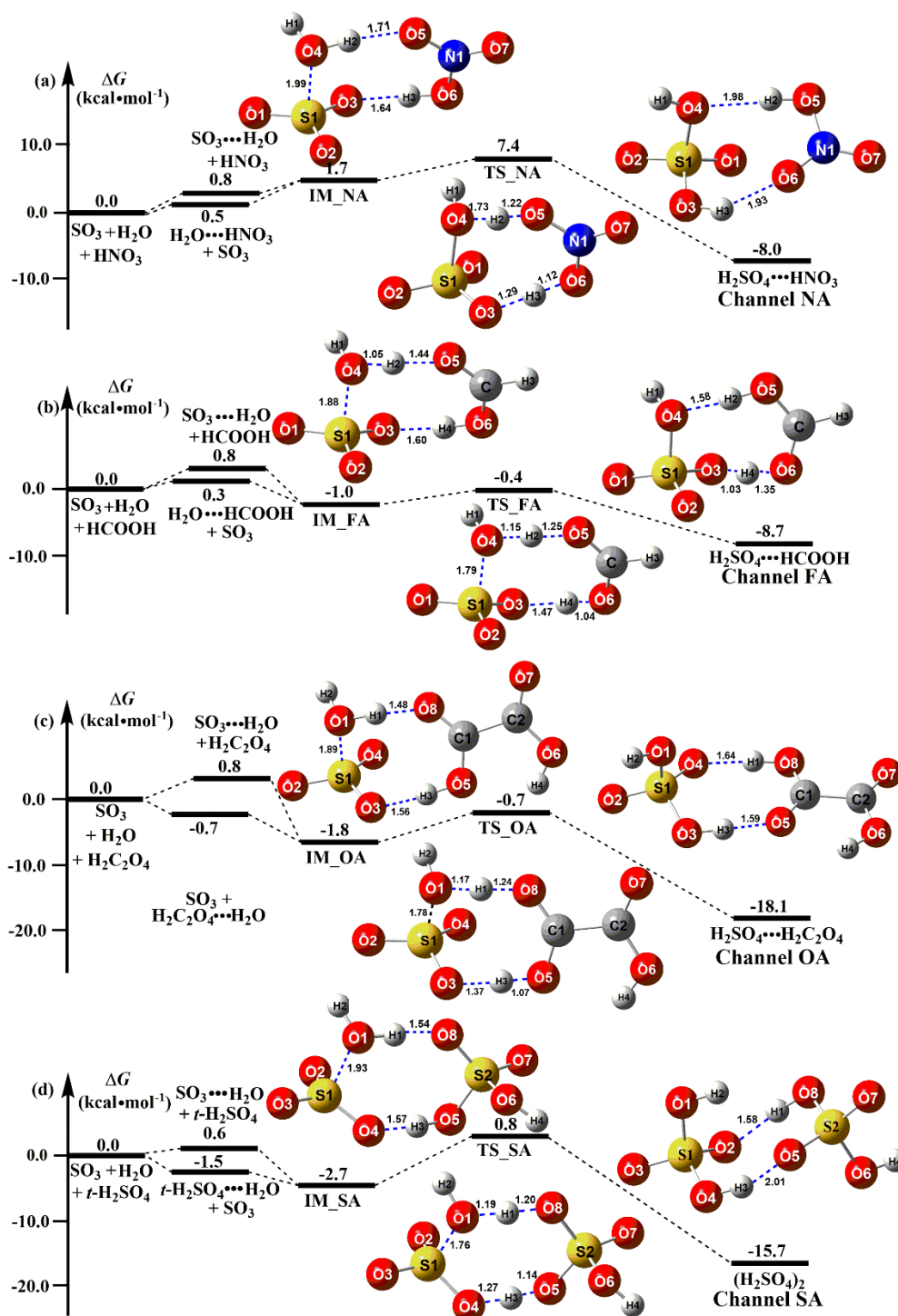


Fig. S3. Energy diagrams for SO $_3$ hydrolysis with HNO $_3$ (a), HCOOH (b) and H $_2$ SO $_4$ (c) at the CCSD(T)-F12/cc-pVDZ-F12//M06-2X/6-311+G(2*df*,2*pd*) level of theory

Table S1. Relative energies (ΔE and $\Delta(E + \text{ZPE})$), enthalpies (ΔH), entropy (S) and free energies (ΔG) for reactants, intermediates, transition states, and products in the $\text{SO}_3 + \text{H}_2\text{SO}_4$ reaction, with and without H_2O , and the SO_3 hydrolysis reaction, with and without H_2O and H_2SO_4 (units: $\text{kcal}\cdot\text{mol}^{-1}$ for ΔE , $\Delta(E + \text{ZPE})$, ΔH , and ΔG ; $\text{kcal}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$ for S)

<i>Species</i>	ZPE	ΔE	S	ΔG	$\Delta(E+\text{ZPE})$	ΔH
$\text{SO}_3 + \text{H}_2\text{O} + \text{FSA}$	52.9	0.0	188.3	0.0	0.0	0.0
$\text{SO}_3\cdots\text{H}_2\text{O} + \text{FSA}$	24.0	-9.4	77.9	0.8(0.33) ^a	-7.0	-7.6
$\text{SO}_3 + \text{FSA}\cdots\text{H}_2\text{O}$	47.1	-12.7	95.6	-1.8	-10.4	-11.2
IM	56.9	-26.4	115.3	-2.0	-22.4	-23.7
TS	53.7	-21.6	110.4	0.5	-20.7	-22.7
$\text{H}_2\text{SO}_4\cdots\text{FSA}$	57.4	-37.1	118.1	-12.9	-32.6	-33.8
$\text{SO}_3 + \text{H}_2\text{O}$	21.6	0.0	106.2	0.0	0.0	0.0
$\text{SO}_3\cdots\text{H}_2\text{O}$	24.0	-9.4	77.9	0.8(0.33) ^a (0.62) ^b	-7.0	-7.6
TS_{SA}	22.3	15.7	70.4	25.4 (25.7) ^a	16.4	14.7
H_2SO_4	25.2	-23.6	71.8	-11.2(-10.72) ^a	-20.0	-21.4
$\text{SO}_3 + \text{H}_2\text{O} + \text{H}_2\text{O}$	35.2	0.0	151.3	0.0	0.0	0.0
$\text{SO}_3\cdots\text{H}_2\text{O} + \text{H}_2\text{O}$	24.0	-9.4	77.9	0.8(0.33) ^a (0.62) ^b	-7.0	-7.6
$\text{SO}_3 + (\text{H}_2\text{O})_2$	29.5	-5.0	68.6	3.2(2.68) ^a	-2.7	-3.3
$\text{SO}_3\cdots(\text{H}_2\text{O})_2$	40.7	-21.6	87.4	1.1(0.8) ^c	-16.0	-17.9
$\text{TS}_{\text{SA_WM}}$	39.1	-14.5	80.1	7.6(6.7) ^c	-10.6	-13.6
$\text{H}_2\text{SO}_4\cdots\text{H}_2\text{O}$	41.0	-36.0	85.5	-12.7(-11.9) ^c	-30.2	-32.3
$\text{SO}_3 + \text{H}_2\text{O} + \text{HNO}_3$	38.9	0.0	169.5	0.0	0.0	0.0
$\text{SO}_3 + \text{HNO}_3\cdots\text{H}_2\text{O}$	41.0	-8.4	139.4	0.5	-7.9 (-8.1) ^c	-8.4
$\text{SO}_3\cdots\text{H}_2\text{O} + \text{HNO}_3$	41.2	-7.8	141.4	0.8	-7.2 (-7.4) ^c	-7.8
IM_NA	42.8	-18.1	103.2	1.7	-17.1 (-17.2) ^c	-18.1
TS_NA	39.9	-15.2	93.6	7.4	-13.2 (-13.5) ^c	-15.2
$\text{H}_2\text{SO}_4\cdots\text{HNO}_3$	43.4	-28.6	100.6	-8.0	-27.4 (-27.8) ^c	-28.6
$\text{SO}_3 + \text{H}_2\text{O} + \text{HCOOH}$	43.2	0.0	165.5	0.0	0.0	0.0
$\text{SO}_3 + \text{HCOOH}\cdots\text{H}_2\text{O}$	45.6	-10.2	134.8	0.3	-7.7	-8.5
$\text{SO}_3\cdots\text{H}_2\text{O} + \text{HCOOH}$	45.5	-9.5	137.4	0.8	-7.2	-7.8
IM_FA	47.4	-25.7	94.1	-1.8(-2.0) ^b	-21.5	-23.1
TS_FA	45.9	-23.9	89.4	-0.8(-1.0) ^b	-21.2	-23.5
$\text{H}_2\text{SO}_4\cdots\text{HCOOH}$	48.0	-40.9	130.8	-8.7(-8.3) ^b	-36.1	-37.7
$\text{SO}_3 + \text{H}_2\text{O} + \text{H}_2\text{C}_2\text{O}_4$	52.9	0.0	181.7	0.0	0.0	0.0
$\text{SO}_3 + \text{H}_2\text{C}_2\text{O}_4\cdots\text{H}_2\text{O}$	55.3	-11.8	150.1	-0.7	-10.6 (-11.1) ^d	-10.1
$\text{SO}_3\cdots\text{H}_2\text{O} + \text{H}_2\text{C}_2\text{O}_4$	55.3	-9.5	153.6	0.8	-7.2 (-7.4) ^d	-7.8
IM_OA	57.1	-26.3	109.1	-1.8	-23.3 (-23.5) ^d	-23.5
TS_OA	54.7	-23.5	104.5	-0.7	-23.0 (-23.5) ^d	-23.8
$\text{H}_2\text{SO}_4\cdots\text{H}_2\text{C}_2\text{O}_4$	57.6	-42.7	110.0	-18.1	-39.3 (-39.1) ^d	-39.5

$\text{SO}_3 + \text{H}_2\text{O} + t\text{-H}_2\text{SO}_4$	46.9	0.0	177.8	0.0	0.0	0.0
$\text{SO}_3 + \text{H}_2\text{SO}_4 \cdots \text{H}_2\text{O}$	49.1	-12.4	146.3	-1.5	-10.1 (-10.1) ^e	-10.9
$\text{SO}_3 \cdots \text{H}_2\text{O} + \text{H}_2\text{SO}_4$	49.2	-9.5	149.7	0.6	-7.2 (-7.0) ^e	-7.8
IM_SA	50.8	-26.2	108.4	-1.5	-22.2 (-21.3) ^e	-23.4
TS_SA	47.5	-20.5	102.1	2.1	-19.9 (-19.9) ^e	-21.8
$(\text{H}_2\text{SO}_4)_2$	51.6	-40.6	105.2	-14.3	-35.9 (-35.4) ^e	-37.4

^a The value was taken from reference (*Chem. Phys. Lett.*, **2013**, 581, 26-29.)

^b The value was taken from reference (*Chem. Phys. Chem.*, **2012**, 13, 323-329.)

^c The value was taken from reference (*J. Phys. Chem. A*, **2021**, 123, 3131-3141.)

^d The value was taken from reference (*Atmos. Chem. Phys.*, **2019**, 19, 2833-2844.)

^e The value was taken from reference (*J. Am. Chem. Soc.*, **2012**, 134, 20632-20644.)

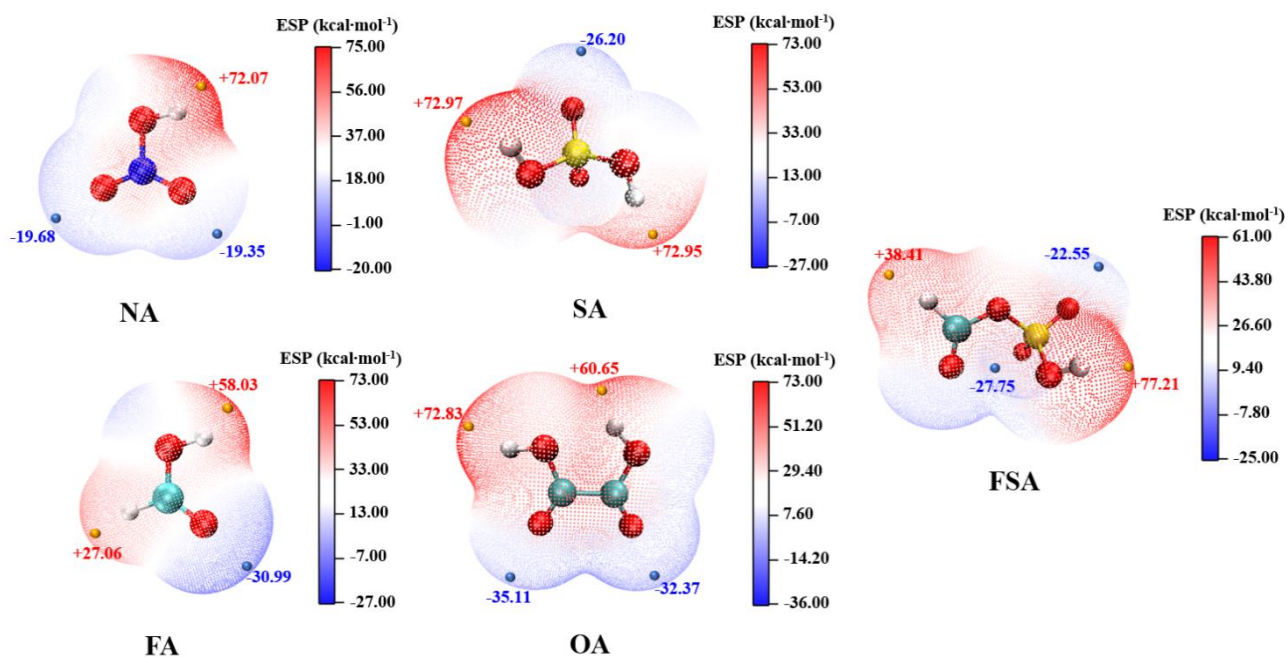


Fig. S4. ESP-mapped van der Waals surface of HNO_3 (NA), H_2SO_4 (SA), HCOOH (FA), $(\text{COOH})_2$ (OA) and HCOOSO_3H (FSA)

37 **Table S2.** Equilibrium constants (molecules·cm⁻³) for FSA···H₂O, SO₃···H₂O and (H₂O)₂ within the temperature
 38 range of 280-320 K

<i>T</i> /K	FSA···H ₂ O	SO ₃ ···H ₂ O	(H ₂ O) ₂	HCOOH···H ₂ O	HNO ₃ ···H ₂ O	H ₂ C ₂ O ₄ ···H ₂ O	H ₂ SO ₄ ···H ₂ O
280	2.63×10^{-18}	2.45×10^{-20}	2.86×10^{-22}	3.11×10^{-20} (2.69×10^{-20}) ^b	4.48×10^{-20}	3.07×10^{-18}	1.53×10^{-18}
290	1.37×10^{-18}	1.59×10^{-20}	2.42×10^{-22}	1.91×10^{-20}	2.75×10^{-20}	1.58×10^{-18}	8.12×10^{-19}
298	8.40×10^{-19}	1.14×10^{-20} (6.44×10^{-20}) ^a	2.14×10^{-22} (2.34×10^{-21}) ^a	1.33×10^{-20} (1.10×10^{-20}) ^b	1.91×10^{-20} (4.07×10^{-20}) ^c	9.65×10^{-19}	5.05×10^{-19} (3.88×10^{-19}) ^d
300	7.47×10^{-19}	1.06×10^{-20}	2.08×10^{-22}	1.22×10^{-20}	1.75×10^{-20}	8.56×10^{-19}	4.50×10^{-19}
310	4.23×10^{-19}	7.25×10^{-21}	1.80×10^{-22}	7.99×10^{-21}	1.14×10^{-20}	4.83×10^{-19}	2.60×10^{-19}
320	2.49×10^{-19}	5.10×10^{-21}	1.58×10^{-22}	5.40×10^{-21} (4.24×10^{-21}) ^b	7.71×10^{-21}	2.82×10^{-19}	1.55×10^{-19}

39 ^a The value was taken from reference (*J. Am. Chem. Soc.*, **2012**, *134*, 20632-20644)

40 ^b The value was taken from reference (*ChemPhysChem.*, **2012**, *13*, 323-329)

41 ^c The value was taken from reference (*RSC Adv.*, **2015**, *5*, 32941-32949)

42 ^d The value was taken from reference (*J. Phys. Chem. A*, **2021**, *125*, 2642-2652)

43 **Table S3.** Concentrations (molecules·cm⁻³) of H₂O, FSA···H₂O and SO₃···H₂O within the temperature range of 280-
 44 320 K^a

Catalysts	RH	280 K	290 K	298 K	300 K	310 K	320 K
H ₂ O	20%RH ^b	5.16×10^{16}	9.60×10^{16}	1.50×10^{17}	1.72×10^{17}	2.92×10^{17}	4.70×10^{17}
	40%RH ^b	1.03×10^{17}	1.91×10^{17}	3.10×10^{17}	3.43×10^{17}	5.84×10^{17}	9.40×10^{17}
	60%RH ^b	1.55×10^{17}	2.87×10^{17}	4.50×10^{17}	5.15×10^{17}	8.77×10^{17}	1.41×10^{18}
	80%RH ^b	2.07×10^{17}	3.82×10^{17}	6.20×10^{17}	6.86×10^{17}	1.17×10^{18}	1.88×10^{18}
	100%RH ^b	2.58×10^{17}	4.78×10^{17}	7.70×10^{17}	8.58×10^{17}	1.46×10^{18}	2.35×10^{18}
FSA···H ₂ O	20%RH ^b	1.36×10^6	1.32×10^6	1.26×10^6	1.28×10^6	1.24×10^6	1.17×10^6
	40%RH ^b	2.71×10^6	2.62×10^6	2.60×10^6	2.56×10^6	2.47×10^6	2.34×10^6
	60%RH ^b	4.08×10^6	3.94×10^6	3.78×10^6	3.85×10^6	3.71×10^6	3.51×10^6
	80%RH ^b	5.45×10^6	5.24×10^6	5.21×10^6	5.12×10^6	4.95×10^6	4.69×10^6
	100%RH ^b	6.80×10^6	6.55×10^6	6.47×10^6	6.41×10^6	6.18×10^6	5.86×10^6
SO ₃ ···H ₂ O	20%RH ^b	1.76	2.09	2.34	2.47	2.84	3.19
	40%RH ^b	3.52	4.17	4.83	4.93	5.69	6.38
	60%RH ^b	5.30	6.26	7.01	7.40	8.54	9.56
	80%RH ^b	7.08	8.33	9.66	9.86	11.4	12.8
	100%RH ^b	8.82	1.04	1.20	12.3	14.2	15.9

45 ^a All of the equilibrium constants were calculated by using energies computed at CCSD(T)-F12a/cc-pVDZ-F12//M06-2X/6-
 46 311++G(2df,2pd) level and partition functions obtained at the M06-2X/6-311++G(2df,2pd) level.

47 ^b The values are reported from reference (*J. Phys. Chem. A*, **2013**, *117*, 10381-10396)

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Part 1. Calculations details of high-pressure-limit (HPL) rate constants

The VRC-VTST calculations were carried out with the potential surface obtained by using CCSD(T)-F12/cc-pVDZ-F12//M06-2X/6-311+G(2df,2pd), and were performed by variationally minimizing the rate constant with respect to the distance s between pivot points and with respect to the location of the pivot points. Specifically, using two pivot points produces a single-faceted dividing surface for the hydrolysis of SO_3 with X ($X = \text{H}_2\text{O}$, HCOOH , HNO_3 and H_2SO_4). Such as, using two pivot points produces a single-faceted dividing surface for the reaction of $\text{FSA} \cdots \text{H}_2\text{O} + \text{SO}_3$. One pivot point is located at a distance d from the center of mass (COM) of $\text{FSA} \cdots \text{H}_2\text{O}$, where the vector connecting the pivot point with $\text{SO}_3 \cdots \text{H}_2\text{O}$'s COM is perpendicular to the SO_3 plane, and the other pivot point is located at a distance d from the COM of catalyst $\text{FSA} \cdots \text{H}_2\text{O}$, where the vector connecting the pivot point with catalyst FSA 's COM is perpendicular to catalyst $\text{FSA} \cdots \text{H}_2\text{O}$ plane. The lengths of these vectors are fixed successively at 0.2 Å. The reaction coordinate s is the distance between a pivot point on one reactant and a pivot point on the other reactant. The distance s between pivot points is varied from 2.5 to 8.0 Å for $\text{FSA} \cdots \text{H}_2\text{O} + \text{SO}_3$ in each case with a 0.2 Å grid increment. The details of the VRC-VTST calculations can be seen in the supporting information of reference.¹

References:

- (1) Bao, J. L.; Zhang, X.; Truhlar, D. G. Barrierless association of CF_2 and dissociation of C_2F_4 by variational transition-state theory and system-specific quantum Rice-Ramsperger-Kassel theory. *Proc. Natl. Acad. Sci. U. S. A.* **2016**, *113*, 13606-13611.

66 **Table S4.** High-pressure limiting rate constants ($\text{cm}^3 \cdot \text{molecules}^{-1} \cdot \text{s}^{-1}$) for the reactants to pre-reactive complex
67 process in SO_3 hydrolysis with and without X ($X = \text{H}_2\text{O}$, NH_3 , HCOOH , $(\text{COOH})_2$, HNO_3 , H_2SO_4 , $\text{H}_2\text{SO}_4 \cdots \text{H}_2\text{O}$
68 and $(\text{H}_2\text{SO}_4)_2$), calculated using the master equation within the temperature range of 280-320 K

$T(\text{K})$	$\text{SO}_3 + \text{H}_2\text{O} \rightarrow \text{IM}$	$\text{SO}_3 \cdots \text{H}_2\text{O} + \text{H}_2\text{O} \rightarrow \text{IM_WM}$	$\text{SO}_3 + \text{NH}_3 \cdots \text{H}_2\text{O} \rightarrow \text{IM_AM}$
280	1.07×10^{-10}	2.17×10^{-10}	1.88×10^{-10}
290	1.09×10^{-10}	2.21×10^{-10}	1.92×10^{-10}
298	1.10×10^{-10}	2.24×10^{-10}	1.94×10^{-10}
300	1.10×10^{-10}	2.25×10^{-10}	1.95×10^{-10}
310	1.12×10^{-10}	2.28×10^{-10}	1.98×10^{-10}
320	1.14×10^{-10}	2.32×10^{-10}	2.01×10^{-10}
$T(\text{K})$	$\text{SO}_3 + \text{HNO}_3 \cdots \text{H}_2\text{O} \rightarrow \text{IM_NA}$	$\text{SO}_3 + \text{HCOOH} \cdots \text{H}_2\text{O} \rightarrow \text{IM_FA}$	$\text{SO}_3 + \text{H}_2\text{C}_2\text{O}_4 \cdots \text{H}_2\text{O} \rightarrow \text{IM_OA}$
280	9.15×10^{-11}	1.25×10^{-10}	9.33×10^{-11}
290	9.32×10^{-11}	1.27×10^{-10}	9.50×10^{-11}
298	9.44×10^{-11}	1.29×10^{-10}	9.63×10^{-11}
300	9.47×10^{-11}	1.30×10^{-10}	9.66×10^{-11}
310	9.63×10^{-11}	1.32×10^{-10}	9.82×10^{-11}
320	9.79×10^{-11}	1.34×10^{-10}	9.98×10^{-11}
$T(\text{K})$	$\text{SO}_3 + \text{H}_2\text{SO}_4 \cdots \text{H}_2\text{O} \rightarrow \text{IM_SA}$	$\text{SO}_3 + \text{FSA} \cdots \text{H}_2\text{O} \rightarrow \text{IM_FSA}$	
280	1.06×10^{-10}	3.09×10^{-11}	
290	1.07×10^{-10}	3.14×10^{-11}	
298	1.09×10^{-10}	3.18×10^{-11}	
300	1.09×10^{-10}	3.19×10^{-11}	
310	1.11×10^{-10}	3.25×10^{-11}	
320	1.13×10^{-10}	3.30×10^{-11}	

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Part 2. Details of Master Equation Solver for Multi-Energy Well Reactions

The rate constants for $\text{SO}_3 + \text{H}_2\text{O} \cdots X$ ($X = \text{H}_2\text{O}$, NH_3 , HCOOH , HNO_3 , $\text{H}_2\text{C}_2\text{O}_4$, H_2SO_4 , $\text{H}_2\text{SO}_4 \cdots \text{H}_2\text{O}$ and $(\text{H}_2\text{SO}_4)_2$) and $\text{SO}_3 \cdots \text{H}_2\text{O} + Y$ reactions within the temperature range of 280-320 K and the pressure range of 10-760 Torr were calculated by using the Master Equation Solver for Multi-Energy Well Reactions (MESMER).¹ Specifically, as for $\text{SO}_3 + \text{H}_2\text{O} \cdots X$ and $\text{SO}_3 \cdots \text{H}_2\text{O} + X$ reaction, the barrierless bimolecular reaction steps were evaluated by using Inverse Laplace Transform (ILT) method, whereas, the rate determining steps were obtained by employing the RRKM theory. The ILT methods²⁻³ and RRKM theory⁴⁻⁵ can be respectively expressed in Eq. (S1)-Eq. (S2).

$$k(E) = \frac{W(E - E_0)}{h\rho(E)} \quad (\text{S1})$$

$$k^\infty(\beta) = \frac{1}{Q(\beta)} \int_0^\infty k(E)\rho(E)\exp(-\beta E)dE \quad (\text{S2})$$

In Eq. (S1) and Eq. (S2), where $W(E-E_0)$ is the rovibrational sum of states (SOS) at the optimized transition state (TS) geometry, E_0 is the reaction threshold energy, h is Planck's constant, $\rho(E)$ is the density of rovibrational states of the reactant, and $Q(\beta)$ is the corresponding canonical partition function. Moreover, the electronic geometries, vibrational frequencies, and rotational constants were calculated at the M06-2X/6-311+G(2df,2pd) level and single-point energy calculations were refined at the CCSD(T)-F12/cc-pVDZ-F12 level for the modeling. The one-dimensional asymmetric Eckart potential⁶ was used to treat the tunneling effect in the RRKM calculation. In addition, The Lennard-Jones (L-J) parameters $\epsilon/k_B = 82$ K and $\sigma = 3.798$ Å were used for N_2 ⁷⁻⁸ while $\epsilon/k_B = 420.08$ K and $\sigma = 2.89$ Å were estimated for H_2SO_4 and its isomer. It was noted that the hindered internal rotation.⁹⁻¹⁸

Reference:

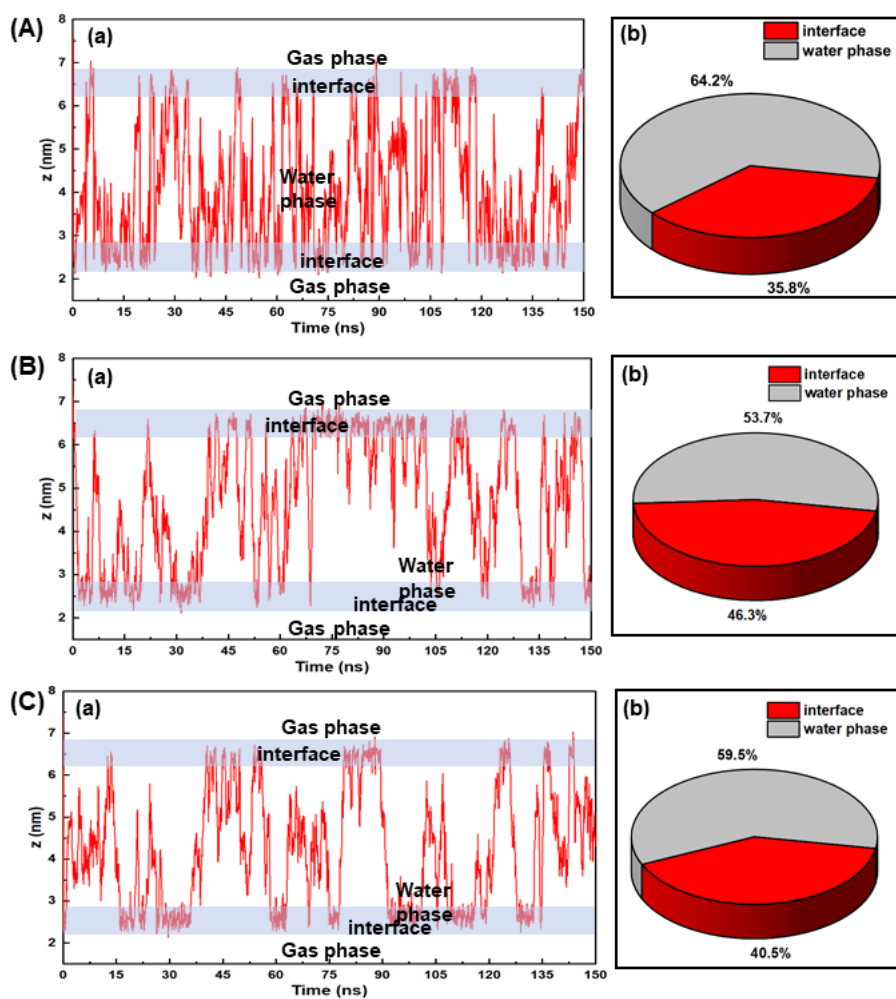
- (1) Glowacki, D. R.; Liang, C. H.; Morley, C.; Pilling, M. J.; Robertson, S. H. MESMER: an open-source master equation solver for multi-energy well reactions. *J. Phys. Chem. A* **2012**, *116*, 9545-9560.
- (2) Horváth, G.; Horváth, I.; Almousa, S. A.-D.; Telek, M. Numerical inverse Laplace transformation using concentrated matrix exponential distributions. *Perform. Evaluation* **2020**, *137*, 102067.
- (3) Kumar, A.; Mallick, S.; Kumar, P. Oxidation of HOSO^\cdot by Cl^\cdot : a new source of SO_2 in the atmosphere? *Phys. Chem. Chem. Phys.* **2021**, *23*, 18707-18711.
- (4) Mai, T. V.-T.; Duong, M. V.; Nguyen, H. T.; Huynh, L. K. J. P. C. C. P. Ab initio kinetics of the $\text{HOSO}_2 + {}^3\text{O}_2 \rightarrow \text{SO}_3 + \text{HO}_2$ reaction. *Phys. Chem. Chem. Phys.* **2018**, *20*, 6677-6687.
- (5) Mallick, S.; Kumar, A.; Kumar, P. Oxidation of HOSO by NH_2 : A new path for the formation of an acid rain precursor. *Chem. Phys. Lett.* **2021**, *773*, 138536.
- (6) Eckart, C. The penetration of a potential barrier by electrons. *Phys. Rev. B* **1930**, *35*, 1303.
- (7) Hippler, H.; Troe, J.; Wendelken, H. J. Collisional deactivation of vibrationally highly excited polyatomic molecules. II. Direct observations for excited toluene. *J. Chem. Phys.* **1983**, *78*, 6709-6717.
- (8) Reid, R. C.; Prausnitz, J. M.; Poling, B. E. The properties of gases and liquids. **1987**.

- 103 (9) Le, X. T.; Mai, T. V.-T.; Duong, M. v.; Huynh, L. K. Kinetics of hydrogen abstraction from desflurane by OH and Cl radicals-A
104 theoretical study. *Chem. Phys. Lett.* **2019**, 728, 142-147.
- 105 (10) Le, X. T.; Mai, T. V.-T.; Lin, K. C.; Huynh, L. K. Low temperature oxidation kinetics of biodiesel molecules: Rate rules for
106 concerted HO₂ elimination from alkyl ester peroxy radicals. *J. Phys. Chem. A* **2018**, 122, 8259-8273.
- 107 (11) Mai, T. V. T.; Duong, M. v.; Nguyen, H. T.; Lin, K. C.; Huynh, L. K. Kinetics of thermal unimolecular decomposition of acetic
108 anhydride: An integrated deterministic and stochastic model. *J. Phys. Chem. A* **2017**, 121, 3028-3036.
- 109 (12) Mai, T. V. T.; Duong, M. V.; Nguyen, H. T.; Lin, K. C.; Huynh, L. K. Ab initio chemical kinetics of the CH₂OO + C₂F₄ reaction.
110 *Chem. Phys. Lett.* **2018**, 706, 280-284.
- 111 (13) Mai, T. V. T.; Huynh, L. K. Ab initio kinetics of the C₂H₂ + NH₂ reaction: a revisited study. *Phys. Chem. Chem. Phys.* **2019**, 21,
112 17232-17239.
- 113 (14) Mai, T. V. T.; Raghunath, P.; Le, X. T.; Huynh, L. K.; Nam, P. C.; Lin, M. C. Ab initio chemical kinetics for the HCCO + OH
114 reaction. *Chem. Phys. Lett.* **2014**, 592, 175-181.
- 115 (15) Mai, T. V. T.; Ratkiewicz, A.; Duong, M. V.; Huynh, L. K. Direct ab initio study of the C₆H₆ + CH₃/C₂H₅ = C₆H₅ + CH₄/C₂H₆
116 reactions. *Chem. Phys. Lett.* **2016**, 646, 102-109.
- 117 (16) Mai, T. V. T.; Nguyen, H. T.; Huynh, L. K. Ab initio dynamics of hydrogen abstraction from N₂H₄ by OH radicals: an RRKM-
118 based master equation study. *Phys. Chem. Chem. Phys.* **2019**, 21, 23733-23741.
- 119 (17) Mai, T. V. T.; Nguyen, H. T.; Huynh, L. K. Kinetics of hydrogen abstraction from CH₃SH by OH radicals: An ab initio RRKM-
120 based master equation study. *Atmos. Environ.* **2020**, 242, 117833.
- 121 (18) Reiner, T.; Arnold, F. Laboratory investigations of gaseous sulfuric acid formation via SO₃ + H₂O + M → H₂SO₄ + M:
122 Measurement of the rate constant and product identification. *J. Chem. Phys.* **1994**, 101, 7399-7407.
- 123

124 **Table S5.** Rate ratios for SO₃ hydrolysis with FSA compared to H₂O and *X* (*X* = HCOOH, (COOH)₂, HNO₃ and
125 H₂SO₄) within the temperature range of 280-320 K

<i>T</i> /K	[<i>Y</i>]	280	290	298	300	310	320
<i>v</i> _{SA_FSA} / <i>v</i> _{SA_WM}	20%RH	7.04×10^{-7}	3.55×10^{-7}	2.16×10^{-7}	1.87×10^{-7}	1.04×10^{-7}	6.11×10^{-8}
	40%RH	3.53×10^{-7}	1.78×10^{-7}	1.05×10^{-7}	9.36×10^{-8}	5.19×10^{-8}	3.06×10^{-8}
	60%RH	2.34×10^{-7}	1.19×10^{-7}	7.22×10^{-8}	6.23×10^{-8}	3.46×10^{-8}	2.04×10^{-8}
	80%RH	1.75×10^{-7}	8.92×10^{-8}	5.24×10^{-8}	4.68×10^{-8}	2.59×10^{-8}	1.53×10^{-8}
	100%RH	1.41×10^{-7}	7.13×10^{-8}	4.22×10^{-8}	3.74×10^{-8}	2.08×10^{-8}	1.22×10^{-8}
<i>v</i> _{SA_FSA} / <i>v</i> _{SA_VFA}	10 ⁸	2.89×10^0	2.72×10^0	2.61×10^0	2.58×10^0	2.47×10^0	2.38×10^0
	10 ¹¹	3.79×10^{-2}	3.62×10^{-2}	3.38×10^{-2}	3.32×10^{-2}	3.24×10^{-2}	3.02×10^{-2}
	10 ¹¹	2.89×10^{-3}	2.72×10^{-3}	2.61×10^{-3}	2.58×10^{-3}	2.47×10^{-3}	2.38×10^{-3}
<i>v</i> _{SA_OA} / <i>v</i> _{SA_OA}	10 ⁷	1.27×10^{-1}	1.31×10^{-1}	1.34×10^{-1}	1.35×10^{-1}	1.38×10^{-1}	1.27×10^{-1}
	10 ⁹	1.27×10^{-3}	1.31×10^{-3}	1.34×10^{-3}	1.35×10^{-3}	1.38×10^{-3}	1.27×10^{-3}
<i>v</i> _{SA_FSA} / <i>v</i> _{SA_VNA}	10 ⁹	3.71×10^1	3.72×10^1	3.73×10^1	3.73×10^1	3.70×10^1	3.61×10^1
	10 ¹¹	3.71×10^{-1}	3.72×10^{-1}	3.73×10^{-1}	3.73×10^{-1}	3.70×10^{-1}	3.61×10^{-1}
<i>v</i> _{SA_FSA} / <i>v</i> _{SA_SA}	10 ⁶	6.26×10^0	6.85×10^0	7.40×10^0	7.70×10^0	8.42×10^0	9.20×10^0
	10 ⁷	4.88×10^{-1}	5.21×10^{-1}	5.48×10^{-1}	5.54×10^{-1}	5.89×10^{-1}	6.26×10^{-1}
	10 ⁸	6.26×10^{-2}	6.85×10^{-2}	7.40×10^{-2}	7.70×10^{-2}	8.42×10^{-2}	9.20×10^{-2}

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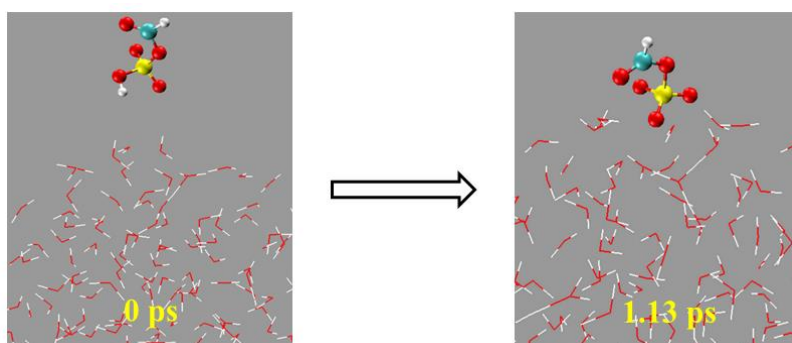
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Fig. S5. Z coordinates of SO_3 (A), FSA (B) and SO_3 -FSA complex (C) as the function of simulation time, along with (a) the water density profile and (b) a pie chart showing occurrence percentages (c) at the air-water interface and in the water phase

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134 **Fig. S6.** Snapshot structures from BOMD simulations of the FSA reaction at the air-water interface. White, red,
135 yellow and blue spheres represent H, O, S, and C atoms, respectively

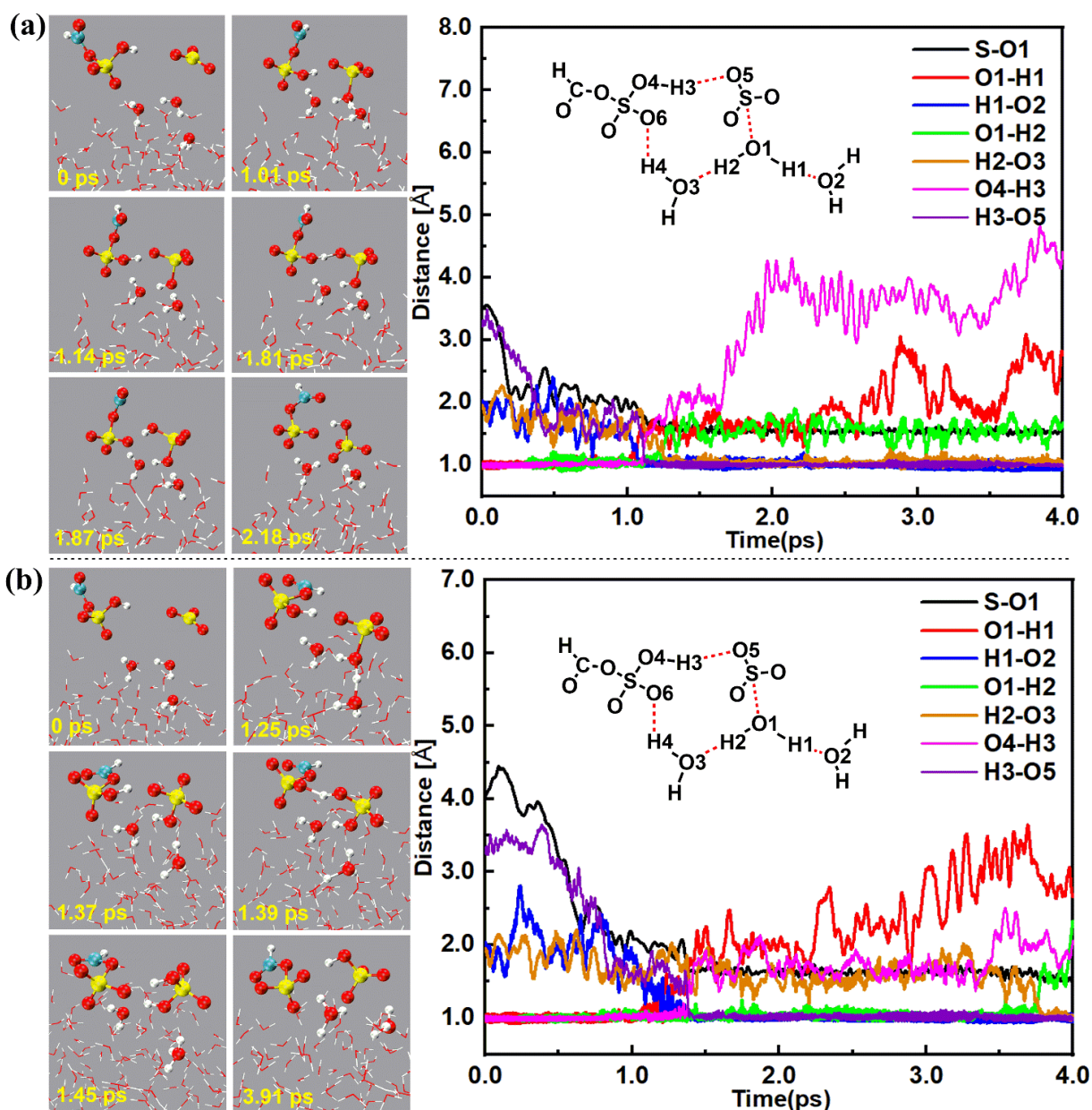
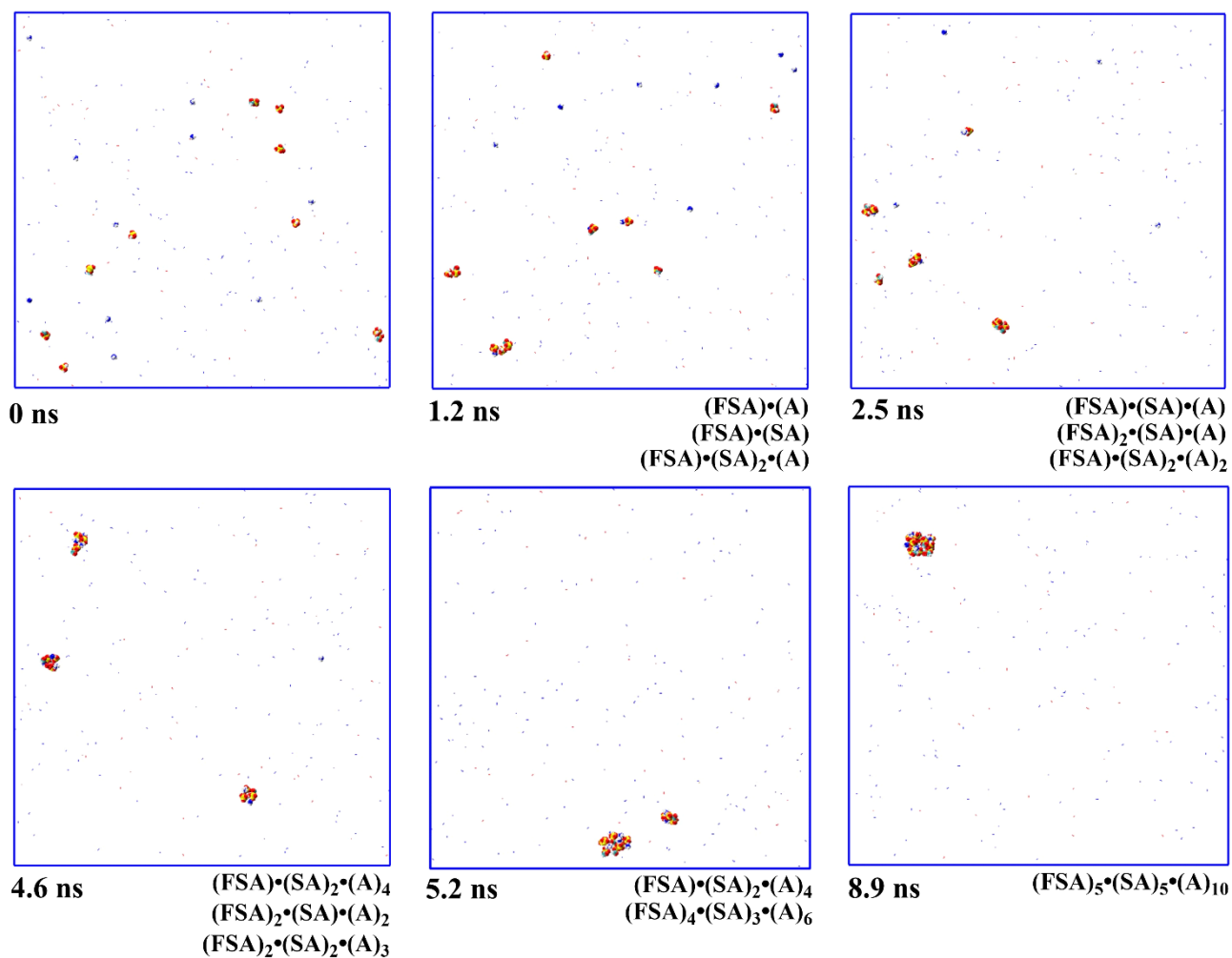


Fig. S7. BOMD simulations of $\text{HSO}_4^- \cdots \text{FSA} \cdots \text{H}_3\text{O}^+$ ion pair formation from SO_3 hydrolysis with FSA at the air-water interface (Top panel: Snapshot structures showing the formation of $\text{HSO}_4^- \cdots \text{FSA} \cdots \text{H}_3\text{O}^+$ ion pair. Lower panel: Time evolution of key bond distances (S-O1, O5-H3, and O1-H2) involved in the mechanism.)



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Fig. S8. Snapshots of nucleation simulation of FSA, SA and A using the VDW representation, with N₂ and O₂ shown using the line drawing method at 278.15 K

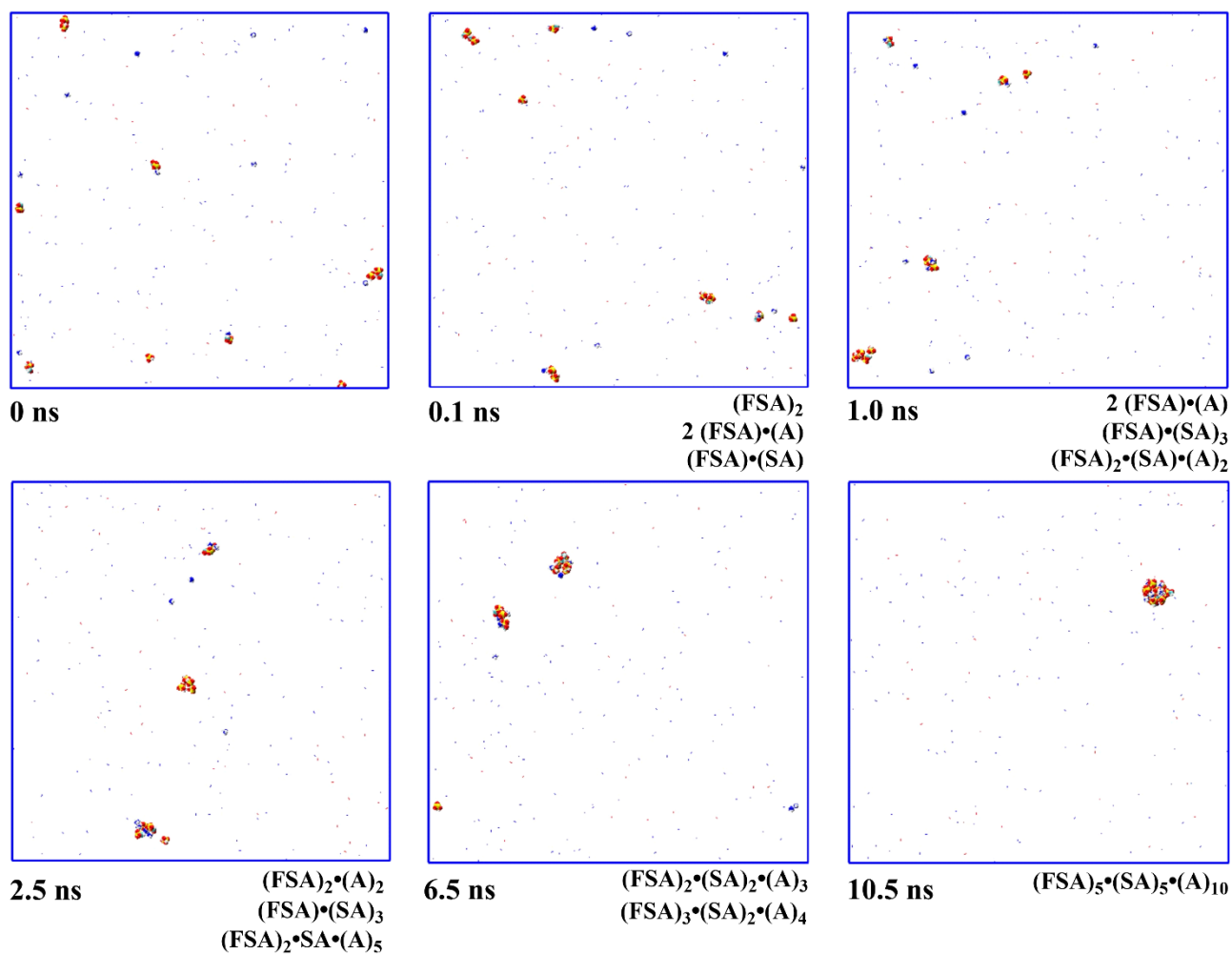


Fig. S9. Snapshots of nucleation simulation of FSA, SA and A using the VDW representation, with N₂ and O₂ shown using the line drawing method at 298.15 K

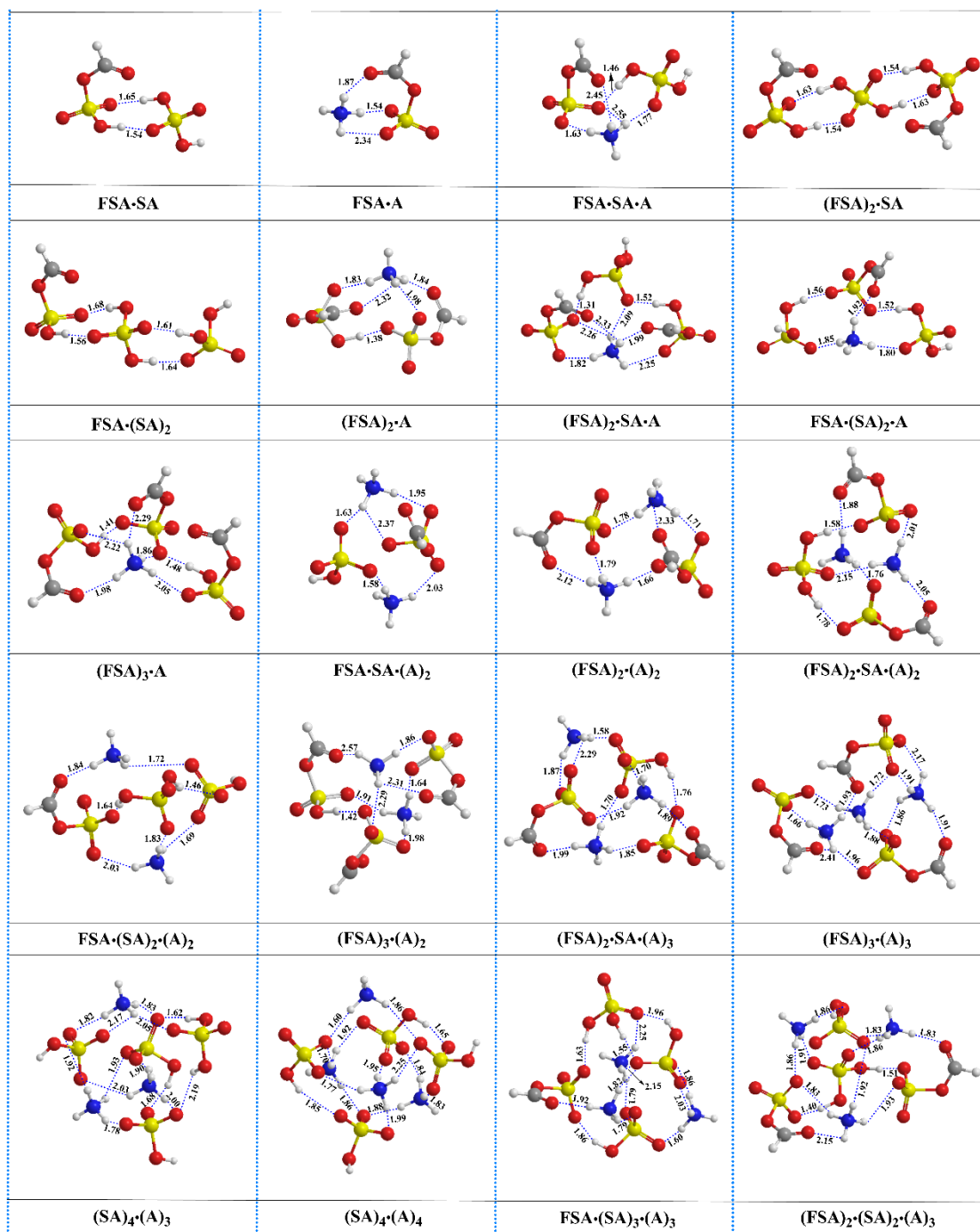


Fig. S10. Most stable configurations of FSA-SA-A-based clusters identified at the M06-2X/6-311++G(2df,2pd) level of theory. Hydrogen bond lengths are given in Å.

150 **Table S6.** Gibbs free energy change ΔG (kcal·mol⁻¹) for the formation of all clusters at 1 atm and
151 temperatures of 298.15, 278.15, and 258.15K

Clusters	$T = 298.15$ K	$T = 278.15$ K	$T = 258.15$ K
SA·A	-7.0 (-7.3) ^a	-7.6 (-7.9) ^a	-8.3 (-8.5) ^a
(SA) ₂	-8.0 (-8.4) ^a	-8.7 (-9.1) ^a	-9.4 (-9.77) ^a
(SA) ₂ ·A	-21.0 (-20.8) ^a	-22.5 (-22.4) ^a	-24.0 (-24.0) ^a
(SA) ₂ ·(A) ₂	-26.5 (-26.5) ^a	-28.7 (-28.8) ^a	-30.9 (-31.0) ^a
(SA) ₃	-12.9 (-13.9) ^a	-14.4 (-15.5) ^a	-16.0 (-17.0) ^a
(SA) ₃ ·A	-29.8 (-30.2) ^a	-32.1 (-32.5) ^a	-34.3 (-34.9) ^a
(SA) ₃ ·(A) ₂	-41.6 (-41.8) ^a	-44.6 (-44.9) ^a	-47.6 (-47.9) ^a
(SA) ₃ ·(A) ₃	-52.0 (-52.8) ^a	-55.8 (-56.6) ^a	-59.7 (-60.5) ^a
(FSA) ₂	-1.8	-2.5	-3.3
FSA·A	-9.1	-9.8	-10.5
(FSA) ₂ ·(A) ₂	-31.1	-33.3	-35.5
(FSA) ₂ ·A	-19.9	-21.5	-23.0
SA·FSA	-5.5	-6.2	-7.0
SA·FSA·A	-18.4	-20.0	-21.5
SA·FSA·(A) ₂	-21.5	-23.8	-26.2
(FSA) ₃	2.2	0.6	-1.0
(SA) ₂ ·FSA·A	-19.3	-21.8	-24.3
(SA) ₂ ·FSA·(A) ₂	-42.9	-46.0	-49.1
(SA) ₂ ·FSA·(A) ₃	-54.1	-57.9	-61.7
SA·(FSA) ₂	-9.8	-11.4	-13.1
(FSA) ₃ ·A	-27.7	-30.1	-32.4
SA·(FSA) ₂ ·(A) ₂	-38.9	-42.1	-45.3
(SA) ₂ ·FSA	-11.7	-13.2	-14.7
SA·(FSA) ₂ ·A	-24.3	-26.5	-28.8
(FSA) ₃ ·(A) ₂	-37.4	-40.4	-43.5

152	Continued table			
	SA·(FSA) ₂ ·(A) ₃	-56.7	-60.6	-64.4
	(FSA) ₃ ·(A) ₃	-51.6	-55.4	-59.2
153	*Calculated at M06-2X/6-311++G(2df,2pd) level of theory.			
154	^a The values in parentheses taken from reference (<i>J. Phys. Chem. A</i> 2020, 124, 3261-3268.)			

Table S7. Evaporation rates (γ , s^{-1}) of the studied clusters at 298.15, 278.15, and 258.15K

Evaporation pathways	298.15 K	278.15 K	258.15 K
$(\text{SA})_2 \rightarrow \text{SA} + \text{SA}$	4.11×10^4	4.33×10^3	3.23×10^2
$(\text{SA})_3 \rightarrow (\text{SA})_2 + \text{SA}$	1.18×10^5	1.37×10^4	1.13×10^3
$(\text{SA})_1 \cdot (\text{A})_1 \rightarrow \text{SA} + \text{A}$	2.96×10^4	4.01×10^3	3.98×10^2
$(\text{SA})_2 \cdot (\text{A})_1 \rightarrow (\text{SA})_1 \cdot (\text{A})_1 + \text{SA}$	1.29×10^{-1}	4.80×10^{-3}	1.07×10^{-4}
$(\text{SA})_2 \cdot (\text{A})_1 \rightarrow \text{A} + (\text{SA})_2$	1.21×10^{-1}	5.76×10^{-3}	1.71×10^{-4}
$(\text{SA})_3 \cdot (\text{A})_1 \rightarrow (\text{SA})_2 \cdot (\text{A})_1 + \text{SA}$	7.65×10^2	6.47×10^1	3.70×10^0
$(\text{SA})_3 \cdot (\text{A})_1 \rightarrow \text{A} + (\text{SA})_3$	1.09×10^{-3}	3.80×10^{-5}	7.83×10^{-7}
$(\text{SA})_2 \cdot (\text{A})_2 \rightarrow (\text{SA})_2 \cdot (\text{A})_1 + \text{A}$	6.13×10^5	8.86×10^4	9.51×10^3
$(\text{SA})_3 \cdot (\text{A})_2 \rightarrow (\text{SA})_2 \cdot (\text{A})_2 + \text{SA}$	2.06×10^{-2}	7.91×10^{-4}	1.82×10^{-5}
$(\text{SA})_3 \cdot (\text{A})_2 \rightarrow (\text{SA})_3 \cdot (\text{A})_1 + \text{A}$	1.66×10^1	1.09×10^0	4.70×10^{-2}
$(\text{SA})_3 \cdot (\text{A})_3 \rightarrow (\text{SA})_3 \cdot (\text{A})_2 + \text{A}$	2.25×10^2	1.48×10^1	6.32×10^{-1}
$(\text{SA})_1 \cdot (\text{FSA})_1 \rightarrow \text{SA} + \text{FSA}$	1.82×10^5	2.38×10^4	2.26×10^3
$(\text{SA})_2 \cdot (\text{FSA})_1 \rightarrow (\text{SA})_1 \cdot (\text{FSA})_1 + \text{SA}$	5.78×10^4	7.03×10^3	6.15×10^2
$(\text{SA})_2 \cdot (\text{FSA})_1 \rightarrow \text{FSA} + (\text{SA})_2$	2.29×10^5	3.46×10^4	3.86×10^3
$(\text{FSA})_1 \cdot (\text{A})_1 \rightarrow \text{FSA} + \text{A}$	1.16×10^3	1.01×10^2	6.00×10^0
$(\text{SA})_1 \cdot (\text{FSA})_1 \cdot (\text{A})_1 \rightarrow \text{SA} + (\text{FSA})_1 \cdot (\text{A})_1$	2.59×10^2	2.04×10^1	1.07×10^0
$(\text{SA})_1 \cdot (\text{FSA})_1 \cdot (\text{A})_1 \rightarrow \text{A} + (\text{SA})_1 \cdot (\text{FSA})_1$	2.27×10^0	1.19×10^{-1}	1.56×10^{-2}
$(\text{SA})_1 \cdot (\text{FSA})_1 \cdot (\text{A})_1 \rightarrow \text{FSA} + (\text{SA})_1 \cdot (\text{A})_1$	9.73×10^0	4.92×10^{-1}	1.45×10^{-5}
$(\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_1 \rightarrow \text{SA} + (\text{SA})_1 \cdot (\text{A})_1 \cdot (\text{FSA})_1$	4.93×10^8	7.62×10^7	8.78×10^6
$(\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_1 \rightarrow \text{A} + (\text{SA})_2 \cdot (\text{FSA})_1$	3.08×10^4	2.05×10^3	8.94×10^1
$(\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_1 \rightarrow \text{FSA} + (\text{SA})_2 \cdot (\text{A})_1$	3.67×10^{10}	7.73×10^9	1.26×10^9
$(\text{SA})_1 \cdot (\text{FSA})_1 \cdot (\text{A})_2 \rightarrow \text{A} + (\text{SA})_1 \cdot (\text{FSA})_1 \cdot (\text{A})_1$	3.83×10^7	6.25×10^6	7.69×10^5
$(\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_2 \rightarrow \text{SA} + (\text{SA})_1 \cdot (\text{FSA})_1 \cdot (\text{A})_2$	4.31×10^{-7}	7.70×10^{-9}	7.33×10^{-11}
$(\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_2 \rightarrow \text{A} + (\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_1$	5.33×10^{-8}	1.00×10^{-9}	1.02×10^{-11}
$(\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_2 \rightarrow \text{FSA} + (\text{SA})_2 \cdot (\text{A})_2$	2.28×10^{-3}	6.23×10^{-5}	9.64×10^{-7}
$(\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_3 \rightarrow \text{A} + (\text{SA})_2 \cdot (\text{FSA})_1 \cdot (\text{A})_2$	5.75×10^1	4.01×10^0	1.85×10^{-1}
$(\text{FSA})_2 \rightarrow \text{FSA} + \text{FSA}$	9.93×10^7	2.01×10^7	3.15×10^6
$(\text{SA})_1 \cdot (\text{FSA})_2 \rightarrow \text{SA} + (\text{FSA})_2$	2.64×10^3	2.13×10^2	1.16×10^1

$(SA)_1 \cdot (FSA)_2 \rightarrow FSA + (SA)_1 \cdot (FSA)_1$	1.35×10^6	1.69×10^5	1.52×10^4
$(FSA)_2 \cdot (A)_1 \rightarrow A + (FSA)_2$	3.88×10^{-4}	1.01×10^{-5}	1.50×10^{-7}
$(FSA)_2 \cdot (A)_1 \rightarrow FSA + (FSA)_1 \cdot (A)_1$	2.31×10^1	1.40×10^0	5.47×10^{-2}
$(SA)_1 \cdot (FSA)_2 \cdot (A)_1 \rightarrow SA + (FSA)_2 \cdot (A)_1$	1.40×10^6	2.47×10^5	3.30×10^4
$(SA)_1 \cdot (FSA)_2 \cdot (A)_1 \rightarrow A + (SA)_1 \cdot (FSA)_2$	3.03×10^{-1}	1.73×10^{-2}	6.28×10^{-4}
$(SA)_1 \cdot (FSA)_2 \cdot (A)_1 \rightarrow FSA + (SA)_1 \cdot (FSA)_1 \cdot (A)_1$	1.03×10^5	1.41×10^4	1.39×10^3
$(FSA)_2 \cdot (A)_2 \rightarrow A + (FSA)_2 \cdot (A)_1$	5.34×10^1	4.34×10^0	2.37×10^{-1}
$(SA)_1 \cdot (FSA)_2 \cdot (A)_2 \rightarrow SA + (FSA)_2 \cdot (A)_2$	4.47×10^3	3.05×10^2	1.37×10^1
$(SA)_1 \cdot (FSA)_2 \cdot (A)_2 \rightarrow A + (SA)_1 \cdot (FSA)_2 \cdot (A)_1$	1.91×10^{-1}	6.00×10^{-3}	1.11×10^{-4}
$(SA)_1 \cdot (FSA)_2 \cdot (A)_2 \rightarrow FSA + (SA)_1 \cdot (FSA)_1 \cdot (A)_2$	3.36×10^{-4}	8.82×10^{-6}	1.31×10^{-7}
$(SA)_1 \cdot (FSA)_2 \cdot (A)_3 \rightarrow A + (SA)_1 \cdot (FSA)_2 \cdot (A)_2$	8.49×10^{-4}	2.90×10^{-5}	5.89×10^{-7}
$(FSA)_3 \rightarrow (FSA)_2 + FSA$	1.62×10^{12}	5.98×10^{11}	1.87×10^{11}
$(FSA)_3 \cdot (A)_1 \rightarrow A + (FSA)_3$	1.19×10^{-12}	8.05×10^{-15}	2.52×10^{-17}
$(FSA)_3 \cdot (A)_1 \rightarrow FSA + (FSA)_2 \cdot (A)_1$	4.26×10^3	4.08×10^2	2.69×10^1
$(FSA)_3 \cdot (A)_2 \rightarrow A + (FSA)_3 \cdot (A)_1$	8.51×10^2	7.09×10^1	4.01×10^0
$(FSA)_3 \cdot (A)_2 \rightarrow FSA + (FSA)_2 \cdot (A)_2$	6.26×10^4	6.16×10^3	4.19×10^2
$(FSA)_3 \cdot (A)_3 \rightarrow A + (FSA)_3 \cdot (A)_2$	3.30×10^{-1}	1.60×10^{-2}	4.82×10^{-4}

Table S8. Collision coefficients (β , $\text{cm}^3 \text{s}^{-1}$) for each cluster

Collisions	$\beta \text{ (cm}^3\text{s}^{-1}\text{)}$		
	298.15 K	278.15 K	258.15 K
SA + A	1.65×10^{-10}	1.54×10^{-10}	1.42×10^{-10}
SA + FSA	7.51×10^{-11}	7.00×10^{-11}	6.50×10^{-11}
FSA + A	2.06×10^{-10}	1.92×10^{-10}	1.78×10^{-10}
SA + SA	6.77×10^{-11}	6.32×10^{-11}	5.87×10^{-11}
FSA + FSA	8.01×10^{-11}	7.47×10^{-11}	6.94×10^{-11}
(SA) ₂ + A	2.82×10^{-10}	2.63×10^{-10}	2.45×10^{-10}
SA·A + SA	8.95×10^{-11}	8.35×10^{-11}	7.75×10^{-11}
(SA) ₂ ·A + A	2.95×10^{-10}	2.75×10^{-10}	2.55×10^{-10}
(FSA) ₂ + A	3.07×10^{-10}	2.86×10^{-10}	2.65×10^{-10}
FSA·A + FSA	8.28×10^{-11}	7.72×10^{-11}	7.17×10^{-11}
(FSA) ₂ ·A + A	3.47×10^{-10}	3.24×10^{-10}	3.00×10^{-10}
(SA) ₂ + FSA	8.77×10^{-11}	8.18×10^{-11}	7.59×10^{-11}
SA·FSA + SA	8.83×10^{-11}	8.24×10^{-11}	7.65×10^{-11}
(FSA) ₂ + SA	8.77×10^{-11}	8.19×10^{-11}	7.60×10^{-11}
SA·FSA + FSA	8.81×10^{-11}	8.22×10^{-11}	7.63×10^{-11}
(SA) ₂ + SA	8.67×10^{-11}	8.09×10^{-11}	7.51×10^{-11}
(FSA) ₂ + FSA	8.68×10^{-11}	8.10×10^{-11}	7.52×10^{-11}
SA + (SA) ₂ ·A	8.82×10^{-11}	8.23×10^{-11}	7.63×10^{-11}
(SA) ₃ + A	4.00×10^{-10}	3.73×10^{-10}	3.46×10^{-10}
(SA) ₂ ·(A) ₂ + SA	9.40×10^{-11}	8.77×10^{-11}	8.14×10^{-11}
(SA) ₃ ·A + A	3.16×10^{-10}	2.95×10^{-10}	2.73×10^{-10}
(SA) ₃ ·(A) ₂ + A	4.15×10^{-10}	3.87×10^{-10}	3.59×10^{-10}
(FSA) ₂ ·A + FSA	9.44×10^{-11}	8.81×10^{-11}	8.17×10^{-11}
(FSA) ₃ + A	3.88×10^{-10}	3.62×10^{-10}	3.36×10^{-10}
(FSA) ₂ ·(A) ₂ + FSA	1.01×10^{-10}	9.44×10^{-11}	8.76×10^{-11}
(FSA) ₃ ·A + A	4.03×10^{-10}	3.76×10^{-10}	3.49×10^{-10}
(FSA) ₃ ·(A) ₂ + A	3.92×10^{-10}	3.65×10^{-10}	3.39×10^{-10}
FSA·A + SA	7.89×10^{-11}	7.36×10^{-11}	6.83×10^{-11}
SA·A + FSA	9.49×10^{-11}	8.85×10^{-11}	8.22×10^{-11}
SA·FSA + A	2.99×10^{-10}	2.79×10^{-10}	2.59×10^{-10}
SA·FSA·A + A	2.90×10^{-10}	2.70×10^{-10}	2.51×10^{-10}
SA·FSA·A + SA	8.43×10^{-11}	7.86×10^{-11}	7.30×10^{-11}

$(SA)_2 \cdot A + FSA$	8.84×10^{-11}	8.24×10^{-11}	7.65×10^{-11}
$(SA)_2 \cdot FSA + A$	4.54×10^{-10}	4.24×10^{-10}	3.93×10^{-10}
$SA \cdot FSA \cdot (A)_2 + SA$	8.00×10^{-11}	7.46×10^{-11}	6.92×10^{-11}
$(SA)_2 \cdot (A)_2 + FSA$	9.32×10^{-11}	8.69×10^{-11}	8.07×10^{-11}
$(SA)_2 \cdot FSA \cdot A + A$	4.37×10^{-10}	4.07×10^{-10}	3.78×10^{-10}
$(SA)_2 \cdot FSA \cdot (A)_2 + A$	3.66×10^{-10}	3.42×10^{-10}	3.17×10^{-10}
$(FSA)_2 \cdot A + SA$	9.66×10^{-11}	9.01×10^{-11}	8.36×10^{-11}
$SA \cdot FSA \cdot A + FSA$	8.39×10^{-11}	7.83×10^{-11}	7.27×10^{-11}
$SA \cdot (FSA)_2 + A$	4.96×10^{-10}	4.63×10^{-10}	4.30×10^{-10}
$(FSA)_2 \cdot (A)_2 + SA$	1.05×10^{-10}	9.75×10^{-11}	9.05×10^{-11}
$SA \cdot FSA \cdot (A)_2 + FSA$	7.96×10^{-11}	7.42×10^{-11}	6.89×10^{-11}
$SA \cdot (FSA)_2 \cdot A + A$	4.20×10^{-10}	3.92×10^{-10}	3.64×10^{-10}
$SA \cdot (FSA)_2 \cdot (A)_2 + A$	3.95×10^{-10}	3.68×10^{-10}	3.42×10^{-10}

161 **Table S9.** Total evaporation coefficients ($\sum\gamma$, s-1) for each cluster

Clusters	$\sum\gamma$, (s ⁻¹)		
	298.15 K	278.15 K	258.15 K
SA·A	2.96×10^4	4.01×10^3	3.98×10^2
SA·FSA	1.82×10^5	2.38×10^4	2.26×10^3
A·FSA	1.16×10^3	1.01×10^2	6.00×10^0
(SA) ₂	4.11×10^4	4.33×10^3	3.23×10^2
(FSA) ₂	9.93×10^7	2.01×10^7	3.15×10^6
(SA) ₂ ·A	2.50×10^{-1}	1.06×10^{-2}	2.78×10^{-4}
(SA) ₂ ·(A) ₂	6.13×10^5	8.86×10^4	9.51×10^3
A·(FSA) ₂	2.31×10^1	1.40×10^0	5.47×10^{-2}
(A) ₂ ·(FSA) ₂	5.34×10^1	4.34×10^0	2.37×10^{-1}
(SA) ₂ ·FSA	2.87×10^5	4.17×10^4	4.47×10^3
SA·(FSA) ₂	1.35×10^6	1.69×10^5	1.52×10^4
(SA) ₃	1.18×10^5	1.37×10^4	1.13×10^3
(FSA) ₃	1.62×10^{12}	5.98×10^{11}	1.87×10^{11}
(SA) ₃ ·A	7.65×10^2	6.47×10^1	3.70×10^0
(SA) ₃ ·(A) ₂	1.66×10^1	1.09×10^0	4.70×10^{-2}
(SA) ₃ ·(A) ₃	2.25×10^2	1.48×10^1	6.32×10^{-1}
A·(FSA) ₃	4.26×10^3	4.08×10^2	2.69×10^1
(A) ₂ ·(FSA) ₃	6.35×10^4	6.23×10^3	4.23×10^2
(A) ₃ ·(FSA) ₃	3.30×10^{-1}	1.60×10^{-2}	4.82×10^{-4}
SA·A·FSA	2.71×10^2	2.10×10^1	1.09×10^0
SA·(A) ₂ ·FSA	3.83×10^7	6.25×10^6	7.69×10^5
SA ₂ ·A·FSA	3.72×10^{10}	7.80×10^9	1.27×10^9
(SA) ₂ ·(A) ₂ ·FSA	2.28×10^{-3}	6.23×10^{-5}	9.64×10^{-7}
(SA) ₂ ·(A) ₃ ·FSA	5.75×10^1	4.01×10^0	1.85×10^{-1}
SA·A·(FSA) ₂	1.50×10^6	2.61×10^5	3.44×10^4
SA·(A) ₂ ·(FSA) ₂	4.47×10^3	3.05×10^2	1.37×10^1
SA·(A) ₃ ·(FSA) ₂	8.49×10^{-4}	2.90×10^{-5}	5.89×10^{-7}

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Table S10. Ratios ($\beta C/\Sigma\gamma$) of monomer collisions to evaporation coefficients for each FSA-involved cluster in this study. $[SA] = 1.0 \times 10^7$ molecules \cdot cm $^{-3}$, $[A] = 1.0 \times 10^{10}$ molecules \cdot cm $^{-3}$, $[FSA] = 1.0 \times 10^6$ molecules \cdot cm $^{-3}$

Clusters	$\beta C/\Sigma\gamma$		
	298.15 K	278.15 K	258.15 K
Collision with SA monomer: $C = [SA]$			
SA \cdot A	3.02×10^{-14}	2.08×10^{-13}	1.95×10^{-12}
SA \cdot FSA	1.15×10^{-18}	4.32×10^{-18}	2.01×10^{-17}
A \cdot FSA	1.03×10^{-21}	1.72×10^{-21}	3.11×10^{-21}
(SA) $_2$	2.11×10^{-14}	1.87×10^{-13}	2.33×10^{-12}
(FSA) $_2$	2.65×10^{-19}	1.00×10^{-18}	4.72×10^{-18}
(SA) $_2\cdot$ A	3.53×10^{-9}	7.79×10^{-8}	2.74×10^{-6}
(SA) $_2\cdot$ (A) $_2$	1.54×10^{-15}	9.91×10^{-15}	8.56×10^{-14}
A \cdot (FSA) $_2$	1.30×10^{-18}	4.89×10^{-18}	2.26×10^{-17}
(A) $_2\cdot$ (FSA) $_2$	1.27×10^{-19}	3.81×10^{-19}	1.36×10^{-18}
SA \cdot A \cdot FSA	1.30×10^{-15}	1.04×10^{-14}	1.16×10^{-13}
SA \cdot (A) $_2\cdot$ FSA	2.61×10^{-22}	5.24×10^{-22}	1.18×10^{-21}
Collision with A monomer: $C = [A]$			
SA \cdot FSA	3.53×10^{-17}	1.33×10^{-16}	6.18×10^{-16}
(SA) $_2$	6.86×10^{-13}	6.08×10^{-12}	7.57×10^{-11}
(FSA) $_2$	7.49×10^{-18}	2.84×10^{-17}	1.33×10^{-16}
(SA) $_2\cdot$ A	1.18×10^{-7}	2.60×10^{-6}	9.17×10^{-5}
A \cdot (FSA) $_2$	3.85×10^{-17}	1.45×10^{-16}	6.69×10^{-16}
(SA) $_2\cdot$ FSA	5.81×10^{-14}	3.92×10^{-13}	3.60×10^{-12}
SA \cdot (FSA) $_2$	2.02×10^{-16}	1.34×10^{-15}	1.20×10^{-14}
(SA) $_3$	3.38×10^{-13}	2.73×10^{-12}	3.07×10^{-11}
(FSA) $_3$	2.28×10^{-14}	1.75×10^{-13}	1.86×10^{-12}
(SA) $_3\cdot$ A	4.13×10^{-11}	4.56×10^{-10}	7.39×10^{-9}
(SA) $_3\cdot$ (A) $_2$	2.50×10^{-9}	3.55×10^{-8}	7.65×10^{-7}
A \cdot (FSA) $_3$	5.89×10^{-19}	1.73×10^{-18}	5.97×10^{-18}
(A) $_2\cdot$ (FSA) $_3$	2.51×10^{-17}	8.68×10^{-17}	3.67×10^{-16}
SA \cdot A \cdot FSA	4.10×10^{-14}	3.28×10^{-13}	3.67×10^{-12}
(SA) $_2\cdot$ A \cdot FSA	3.11×10^{-15}	2.03×10^{-14}	1.79×10^{-13}

$(SA)_2 \cdot (A)_2 \cdot FSA$	2.45×10^{-12}	2.82×10^{-11}	4.82×10^{-10}
$SA \cdot A \cdot (FSA)_2$	4.52×10^{-14}	3.88×10^{-13}	4.70×10^{-12}
$SA \cdot (A)_2 \cdot (FSA)_2$	3.12×10^{-21}	5.14×10^{-21}	9.27×10^{-21}
Collision with FSA monomer: $C=[FSA]$			
$SA \cdot A$	3.94×10^{-15}	2.71×10^{-14}	2.54×10^{-13}
$SA \cdot FSA$	1.55×10^{-19}	5.82×10^{-19}	2.71×10^{-18}
$A \cdot FSA$	1.30×10^{-22}	2.15×10^{-22}	3.90×10^{-22}
$(SA)_2$	2.89×10^{-15}	2.56×10^{-14}	3.18×10^{-13}
$(FSA)_2$	3.48×10^{-20}	1.32×10^{-19}	6.21×10^{-19}
$(SA)_2 \cdot A$	4.87×10^{-10}	1.07×10^{-8}	3.78×10^{-7}
$(SA)_2 \cdot (A)_2$	2.13×10^{-16}	1.37×10^{-15}	1.19×10^{-14}
$A \cdot (FSA)_2$	1.73×10^{-19}	6.52×10^{-19}	3.02×10^{-18}
$(A)_2 \cdot (FSA)_2$	1.71×10^{-20}	5.12×10^{-20}	1.84×10^{-19}
$SA \cdot A \cdot FSA$	1.76×10^{-16}	1.41×10^{-15}	1.58×10^{-14}
$SA \cdot (A)_2 \cdot FSA$	3.57×10^{-23}	7.17×10^{-23}	1.62×10^{-22}

168 **Table S11.** Formation rate (J , s^{-1}) of FSA at $T = 298.15$ K with $[SA] = 10^4 - 10^8$ molecules \cdot cm $^{-3}$, $[A]$
169 $= 10^7 - 10^{11}$ molecules \cdot cm $^{-3}$, and $[FSA] = 10^3 - 10^7$ molecules \cdot cm $^{-3}$.

[SA]	[A]	[FSA] = 10^3	[FSA] = 10^4	[FSA] = 10^5	[FSA] = 10^6	[FSA] = 10^7
[SA] = 10^4	[A] = 10^7	6.44×10^{-29}	1.24×10^{-27}	7.22×10^{-26}	6.58×10^{-24}	5.53×10^{-22}
[SA] = 10^4	[A] = 10^8	6.44×10^{-26}	1.24×10^{-24}	7.22×10^{-23}	6.58×10^{-21}	5.53×10^{-19}
[SA] = 10^4	[A] = 10^9	6.42×10^{-23}	1.24×10^{-21}	7.20×10^{-20}	6.56×10^{-18}	5.51×10^{-16}
[SA] = 10^4	[A] = 10^{10}	6.22×10^{-20}	1.20×10^{-18}	7.00×10^{-17}	6.38×10^{-15}	5.34×10^{-13}
[SA] = 10^4	[A] = 10^{11}	4.80×10^{-17}	9.42×10^{-16}	5.56×10^{-14}	5.06×10^{-12}	4.09×10^{-10}
[SA] = 10^5	[A] = 10^7	5.94×10^{-26}	6.44×10^{-25}	1.24×10^{-23}	7.09×10^{-22}	5.57×10^{-20}
[SA] = 10^5	[A] = 10^8	5.94×10^{-23}	6.43×10^{-22}	1.24×10^{-20}	7.08×10^{-19}	5.57×10^{-17}
[SA] = 10^5	[A] = 10^9	5.92×10^{-20}	6.41×10^{-19}	1.24×10^{-17}	7.06×10^{-16}	5.55×10^{-14}
[SA] = 10^5	[A] = 10^{10}	5.74×10^{-17}	6.22×10^{-16}	1.20×10^{-14}	6.87×10^{-13}	5.38×10^{-11}
[SA] = 10^5	[A] = 10^{11}	4.43×10^{-14}	4.80×10^{-13}	9.39×10^{-12}	5.43×10^{-10}	4.11×10^{-8}
[SA] = 10^6	[A] = 10^7	6.86×10^{-23}	5.94×10^{-22}	6.42×10^{-21}	1.22×10^{-19}	6.00×10^{-18}
[SA] = 10^6	[A] = 10^8	6.86×10^{-20}	5.93×10^{-19}	6.42×10^{-18}	1.22×10^{-16}	6.00×10^{-15}
[SA] = 10^6	[A] = 10^9	6.84×10^{-17}	5.92×10^{-16}	6.40×10^{-15}	1.21×10^{-13}	5.98×10^{-12}
[SA] = 10^6	[A] = 10^{10}	6.66×10^{-14}	5.73×10^{-13}	6.20×10^{-12}	1.18×10^{-10}	5.78×10^{-9}
[SA] = 10^6	[A] = 10^{11}	5.33×10^{-11}	4.39×10^{-10}	4.74×10^{-9}	9.09×10^{-8}	4.39×10^{-6}
[SA] = 10^7	[A] = 10^7	1.67×10^{-19}	6.86×10^{-19}	5.93×10^{-18}	6.30×10^{-17}	1.03×10^{-15}
[SA] = 10^7	[A] = 10^8	1.67×10^{-16}	6.86×10^{-16}	5.92×10^{-15}	6.30×10^{-14}	1.03×10^{-12}
[SA] = 10^7	[A] = 10^9	1.67×10^{-13}	6.83×10^{-13}	5.90×10^{-12}	6.28×10^{-11}	1.02×10^{-9}
[SA] = 10^7	[A] = 10^{10}	1.64×10^{-10}	6.60×10^{-10}	5.66×10^{-9}	6.02×10^{-8}	9.80×10^{-7}
[SA] = 10^7	[A] = 10^{11}	1.48×10^{-7}	4.99×10^{-7}	4.04×10^{-6}	4.27×10^{-5}	6.92×10^{-4}
[SA] = 10^8	[A] = 10^7	1.15×10^{-15}	1.67×10^{-15}	6.86×10^{-15}	5.83×10^{-14}	5.37×10^{-13}
[SA] = 10^8	[A] = 10^8	1.15×10^{-12}	1.67×10^{-12}	6.85×10^{-12}	5.82×10^{-11}	5.36×10^{-10}
[SA] = 10^8	[A] = 10^9	1.15×10^{-9}	1.66×10^{-9}	6.76×10^{-9}	5.74×10^{-8}	5.29×10^{-7}
[SA] = 10^8	[A] = 10^{10}	1.14×10^{-6}	1.59×10^{-6}	6.03×10^{-6}	5.02×10^{-5}	4.67×10^{-4}
[SA] = 10^8	[A] = 10^{11}	1.12×10^{-3}	1.31×10^{-3}	3.26×10^{-3}	2.27×10^{-2}	2.15×10^{-1}

170 SA, A and FSA represent sulfuric acid, ammonia and formic sulfuric anhydride, respectively.

171 **Table S12.** Formation rate J (s^{-1}) of FSA at $T = 278.15$ K with $[\text{SA}] = 10^4 - 10^8$ molecules $\cdot\text{cm}^{-3}$, $[\text{A}]$
 172 $= 10^7 - 10^{11}$ molecules $\cdot\text{cm}^{-3}$, and $[\text{FSA}] = 10^3 - 10^7$ molecules $\cdot\text{cm}^{-3}$

[SA]	[A]	[FSA] = 10^3	[FSA] = 10^4	[FSA] = 10^5	[FSA] = 10^6	[FSA] = 10^7
[SA] = 10^4	[A] = 10^7	1.92×10^{-24}	3.63×10^{-23}	2.09×10^{-21}	1.88×10^{-19}	1.44×10^{-17}
[SA] = 10^4	[A] = 10^8	1.90×10^{-21}	3.60×10^{-20}	2.08×10^{-18}	1.87×10^{-16}	1.43×10^{-14}
[SA] = 10^4	[A] = 10^9	1.78×10^{-18}	3.36×10^{-17}	1.95×10^{-15}	1.75×10^{-13}	1.34×10^{-11}
[SA] = 10^4	[A] = 10^{10}	1.07×10^{-15}	2.06×10^{-14}	1.22×10^{-12}	1.10×10^{-10}	8.28×10^{-9}
[SA] = 10^4	[A] = 10^{11}	2.52×10^{-13}	5.50×10^{-12}	3.77×10^{-10}	3.46×10^{-8}	2.46×10^{-6}
[SA] = 10^5	[A] = 10^7	2.00×10^{-21}	1.92×10^{-20}	3.61×10^{-19}	2.03×10^{-17}	1.45×10^{-15}
[SA] = 10^5	[A] = 10^8	1.99×10^{-18}	1.90×10^{-17}	3.59×10^{-16}	2.02×10^{-14}	1.44×10^{-12}
[SA] = 10^5	[A] = 10^9	1.87×10^{-15}	1.77×10^{-14}	3.35×10^{-13}	1.89×10^{-11}	1.35×10^{-9}
[SA] = 10^5	[A] = 10^{10}	1.23×10^{-12}	1.07×10^{-11}	2.05×10^{-10}	1.18×10^{-8}	8.33×10^{-7}
[SA] = 10^5	[A] = 10^{11}	4.73×10^{-10}	2.51×10^{-9}	5.46×10^{-8}	3.61×10^{-6}	2.46×10^{-4}
[SA] = 10^6	[A] = 10^7	4.55×10^{-18}	2.00×10^{-17}	1.91×10^{-16}	3.51×10^{-15}	1.57×10^{-13}
[SA] = 10^6	[A] = 10^8	4.54×10^{-15}	1.99×10^{-14}	1.90×10^{-13}	3.48×10^{-12}	1.56×10^{-10}
[SA] = 10^6	[A] = 10^9	4.42×10^{-12}	1.87×10^{-11}	1.76×10^{-10}	3.24×10^{-9}	1.45×10^{-7}
[SA] = 10^6	[A] = 10^{10}	3.77×10^{-9}	1.21×10^{-8}	1.05×10^{-7}	1.96×10^{-6}	8.84×10^{-5}
[SA] = 10^6	[A] = 10^{11}	3.01×10^{-6}	4.68×10^{-6}	2.44×10^{-5}	5.10×10^{-4}	2.53×10^{-2}
[SA] = 10^7	[A] = 10^7	3.03×10^{-14}	4.56×10^{-14}	2.00×10^{-13}	1.86×10^{-12}	2.70×10^{-11}
[SA] = 10^7	[A] = 10^8	3.03×10^{-11}	4.54×10^{-11}	1.98×10^{-10}	1.84×10^{-9}	2.68×10^{-8}
[SA] = 10^7	[A] = 10^9	3.01×10^{-8}	4.39×10^{-8}	1.83×10^{-7}	1.67×10^{-6}	2.45×10^{-5}
[SA] = 10^7	[A] = 10^{10}	2.94×10^{-5}	3.66×10^{-5}	1.09×10^{-4}	8.97×10^{-4}	1.34×10^{-2}
[SA] = 10^7	[A] = 10^{11}	2.83×10^{-2}	2.96×10^{-2}	4.26×10^{-2}	1.92×10^{-1}	3.08×10^0
[SA] = 10^8	[A] = 10^7	2.91×10^{-10}	3.07×10^{-10}	4.61×10^{-10}	1.97×10^{-9}	1.46×10^{-8}
[SA] = 10^8	[A] = 10^8	2.91×10^{-7}	3.06×10^{-7}	4.55×10^{-7}	1.92×10^{-6}	1.42×10^{-5}
[SA] = 10^8	[A] = 10^9	2.91×10^{-4}	3.02×10^{-4}	4.14×10^{-4}	1.52×10^{-3}	1.12×10^{-2}
[SA] = 10^8	[A] = 10^{10}	2.88×10^{-1}	2.91×10^{-1}	3.24×10^{-1}	6.47×10^{-1}	3.82×10^0
[SA] = 10^8	[A] = 10^{11}	2.69×10^2	2.69×10^2	2.74×10^2	3.15×10^2	7.56×10^2

173 SA, A and FSA represent sulfuric acid, ammonia and formic sulfuric anhydride, respectively

174 **Table S13.** Formation rate J (s^{-1}) of FSA at $T = 258.15$ K with $[\text{SA}] = 10^4 - 10^8$ molecules $\cdot\text{cm}^{-3}$, $[\text{A}]$
175 $= 10^7 - 10^{11}$ molecules $\cdot\text{cm}^{-3}$, and $[\text{FSA}] = 10^3 - 10^7$ molecules $\cdot\text{cm}^{-3}$

[SA]	[A]	[FSA] = 10^3	[FSA] = 10^4	[FSA] = 10^5	[FSA] = 10^6	[FSA] = 10^7
[SA] = 10^4	[A] = 10^7	2.49×10^{-20}	4.64×10^{-19}	3.13×10^{-17}	2.89×10^{-15}	2.24×10^{-13}
[SA] = 10^4	[A] = 10^8	2.27×10^{-17}	4.23×10^{-16}	2.90×10^{-14}	2.69×10^{-12}	2.07×10^{-10}
[SA] = 10^4	[A] = 10^9	1.39×10^{-14}	2.62×10^{-13}	1.97×10^{-11}	1.85×10^{-9}	1.41×10^{-7}
[SA] = 10^4	[A] = 10^{10}	5.48×10^{-12}	1.16×10^{-10}	1.00×10^{-8}	9.48×10^{-7}	6.48×10^{-5}
[SA] = 10^4	[A] = 10^{11}	6.87×10^{-10}	3.11×10^{-8}	2.87×10^{-6}	2.35×10^{-4}	4.75×10^{-3}
[SA] = 10^5	[A] = 10^7	7.46×10^{-17}	2.49×10^{-16}	4.62×10^{-15}	3.03×10^{-13}	2.25×10^{-11}
[SA] = 10^5	[A] = 10^8	7.23×10^{-14}	2.27×10^{-13}	4.22×10^{-12}	2.81×10^{-10}	2.08×10^{-8}
[SA] = 10^5	[A] = 10^9	6.14×10^{-11}	1.39×10^{-10}	2.61×10^{-9}	1.91×10^{-7}	1.41×10^{-5}
[SA] = 10^5	[A] = 10^{10}	3.28×10^{-8}	5.47×10^{-8}	1.15×10^{-6}	9.57×10^{-5}	6.48×10^{-3}
[SA] = 10^5	[A] = 10^{11}	2.32×10^{-6}	6.82×10^{-6}	3.04×10^{-4}	2.36×10^{-2}	4.74×10^{-1}
[SA] = 10^6	[A] = 10^7	5.98×10^{-13}	7.46×10^{-13}	2.48×10^{-12}	4.47×10^{-11}	2.35×10^{-9}
[SA] = 10^6	[A] = 10^8	5.93×10^{-10}	7.22×10^{-10}	2.26×10^{-9}	4.06×10^{-8}	2.16×10^{-6}
[SA] = 10^6	[A] = 10^9	5.52×10^{-7}	6.12×10^{-7}	1.37×10^{-6}	2.48×10^{-5}	1.43×10^{-3}
[SA] = 10^6	[A] = 10^{10}	3.10×10^{-4}	3.23×10^{-4}	5.34×10^{-4}	1.07×10^{-2}	6.42×10^{-1}
[SA] = 10^6	[A] = 10^{11}	2.04×10^{-2}	2.22×10^{-2}	6.40×10^{-2}	2.41×10^0	4.69×10^1
[SA] = 10^7	[A] = 10^7	5.84×10^{-9}	5.98×10^{-9}	7.49×10^{-9}	2.45×10^{-8}	3.42×10^{-7}
[SA] = 10^7	[A] = 10^8	5.79×10^{-6}	5.91×10^{-6}	7.19×10^{-6}	2.16×10^{-5}	3.02×10^{-4}
[SA] = 10^7	[A] = 10^9	5.36×10^{-3}	5.41×10^{-3}	5.93×10^{-3}	1.23×10^{-2}	1.68×10^{-1}
[SA] = 10^7	[A] = 10^{10}	2.65×10^0	2.66×10^0	2.75×10^0	4.29×10^0	6.18×10^1
[SA] = 10^7	[A] = 10^{11}	1.35×10^2	1.36×10^2	1.46×10^2	3.68×10^2	4.30×10^3
[SA] = 10^8	[A] = 10^7	5.80×10^{-5}	5.82×10^{-5}	6.00×10^{-5}	7.76×10^{-5}	2.31×10^{-4}
[SA] = 10^8	[A] = 10^8	5.66×10^{-2}	5.68×10^{-2}	5.79×10^{-2}	6.93×10^{-2}	1.76×10^{-1}
[SA] = 10^8	[A] = 10^9	4.47×10^1	4.47×10^1	4.50×10^1	4.75×10^1	7.52×10^1
[SA] = 10^8	[A] = 10^{10}	8.29×10^3	8.29×10^3	8.30×10^3	8.46×10^3	1.13×10^4
[SA] = 10^8	[A] = 10^{11}	1.57×10^5	1.58×10^5	1.58×10^5	1.63×10^5	2.94×10^5

176 SA, A and FSA represent sulfuric acid, ammonia and formic sulfuric anhydride, respectively

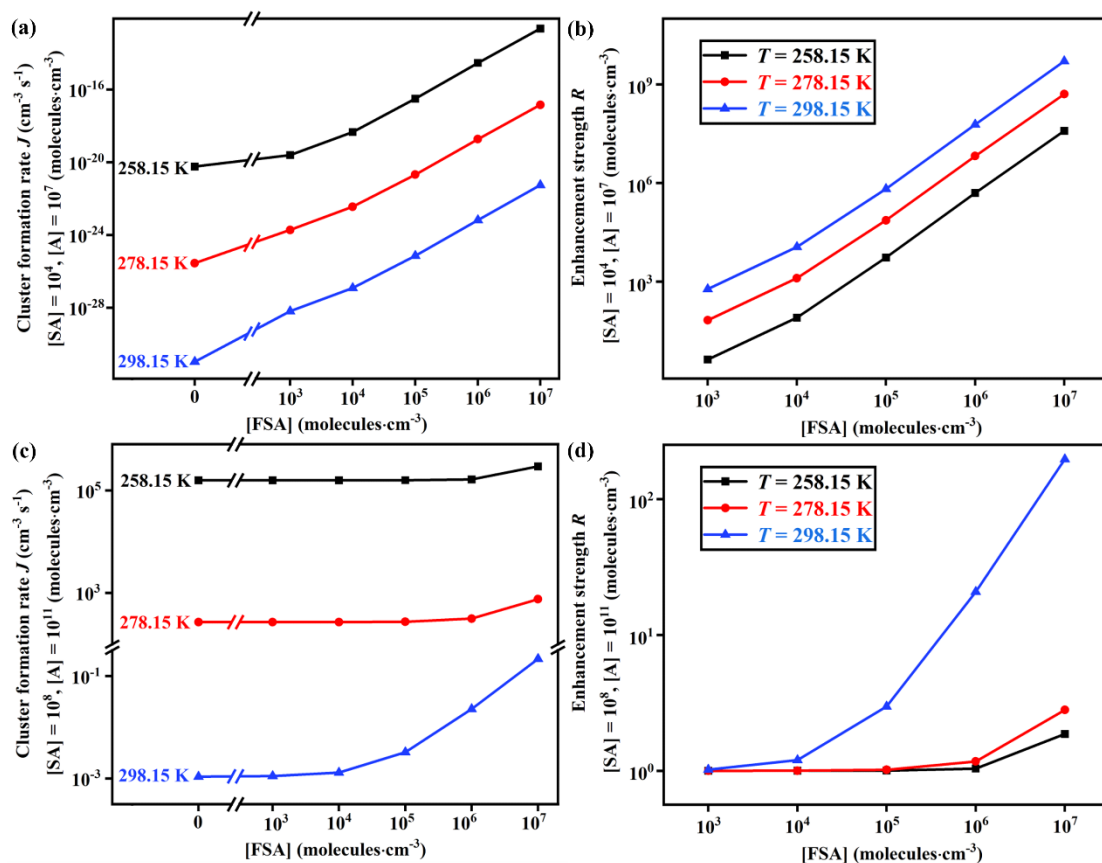


Fig. S11. (a) Cluster formation rate (J , cm³ s⁻¹) and (b) enhancement factor (R) with $[SA] = 10^4$ molecules·cm⁻³, $[A] = 10^7$ molecules·cm⁻³ at three temperatures, (c) Cluster formation rate (J , cm³ s⁻¹) and (d) enhancement factor (R) with $[SA] = 10^8$ molecules·cm⁻³, $[A] = 10^{11}$ molecules·cm⁻³ at three temperatures (black: 258.15 K, red: 278.15 K, blue: 298.15 K)

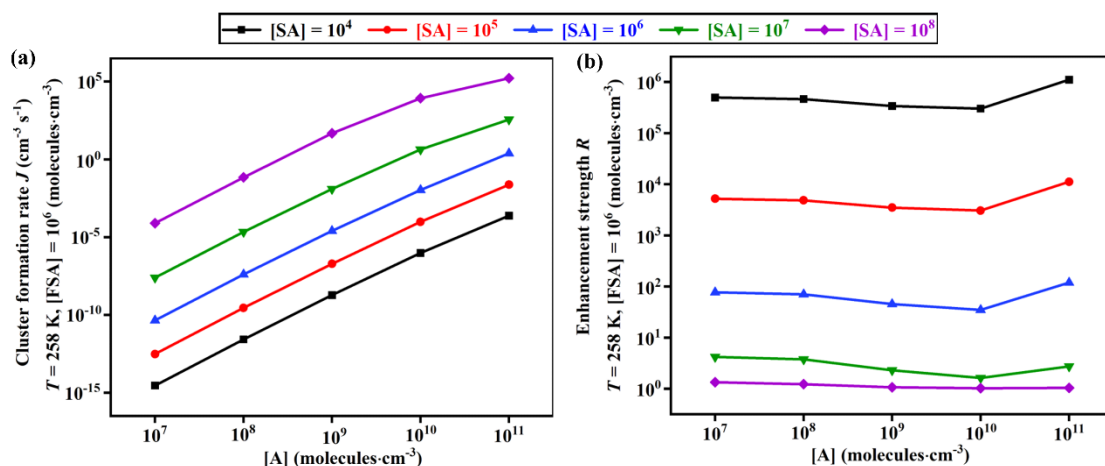


Fig. S12. (a) Cluster formation rate (J , cm³ s⁻¹) and (b) enhancement factor (R) as a function of $[A]$ with $[FSA] = 10^6$ molecules·cm⁻³ and five different $[SA]$ concentrations (black: $[SA] = 10^4$ molecules·cm⁻³, red: $[SA] = 10^5$ molecules·cm⁻³, blue: $[SA] = 10^6$ molecules·cm⁻³, green: $[SA] = 10^7$ molecules·cm⁻³, purple: $[SA] = 10^8$ molecules·cm⁻³) at 258.15 K

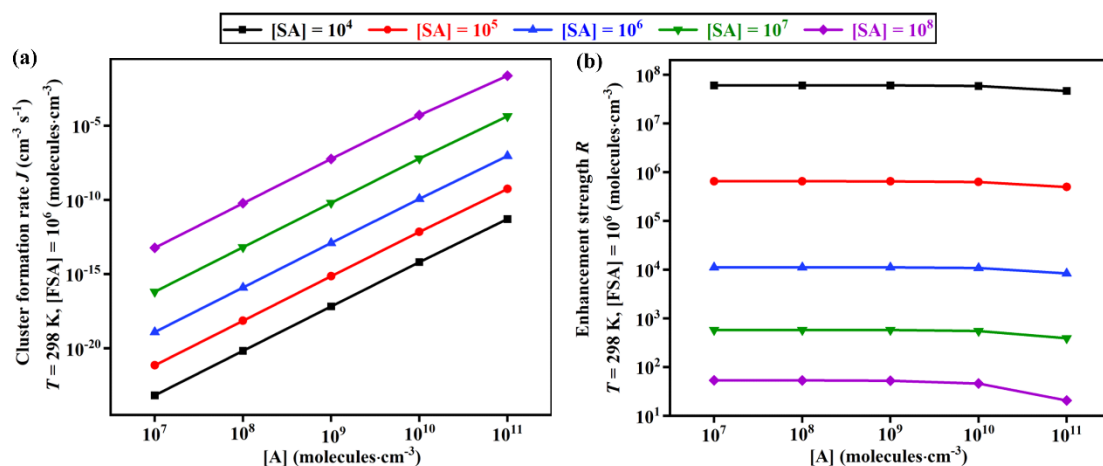


Fig. S13. (a) Cluster formation rate (J , cm³ s⁻¹) and (b) enhancement factor (R) as a function of $[A]$ with $[FSA] = 10^6$ molecules·cm⁻³ and five different $[SA]$ concentrations (black: $[SA] = 10^4$ molecules·cm⁻³, red: $[SA] = 10^5$ molecules·cm⁻³, blue: $[SA] = 10^6$ molecules·cm⁻³, green: $[SA] = 10^7$ molecules·cm⁻³, purple: $[SA] = 10^8$ molecules·cm⁻³) at 298.15 K

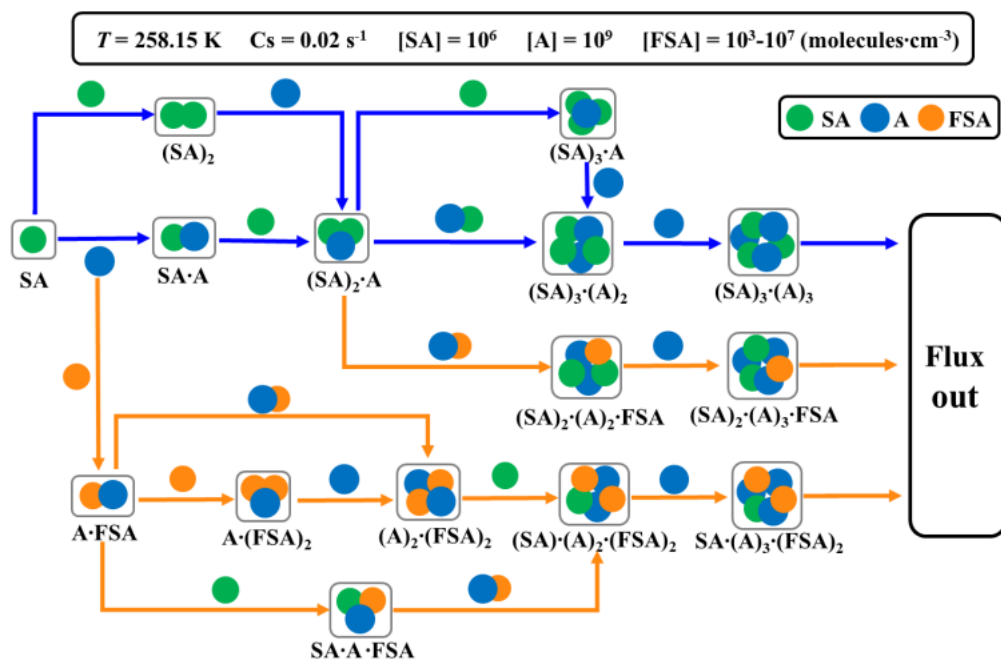


Fig. S14. Main pathways of clusters growth under conditions of $T = 258.15 \text{ K}$, $[\text{SA}] = 10^6 \text{ molecules}\cdot\text{cm}^{-3}$, $[\text{A}] = 10^9 \text{ molecules}\cdot\text{cm}^{-3}$, and $[\text{FSA}] = 10^3\text{-}10^7 \text{ molecules}\cdot\text{cm}^{-3}$. Blue and orange fluxes represent the pathways of the SA-A-based and SA-A-FSA-based cluster, respectively

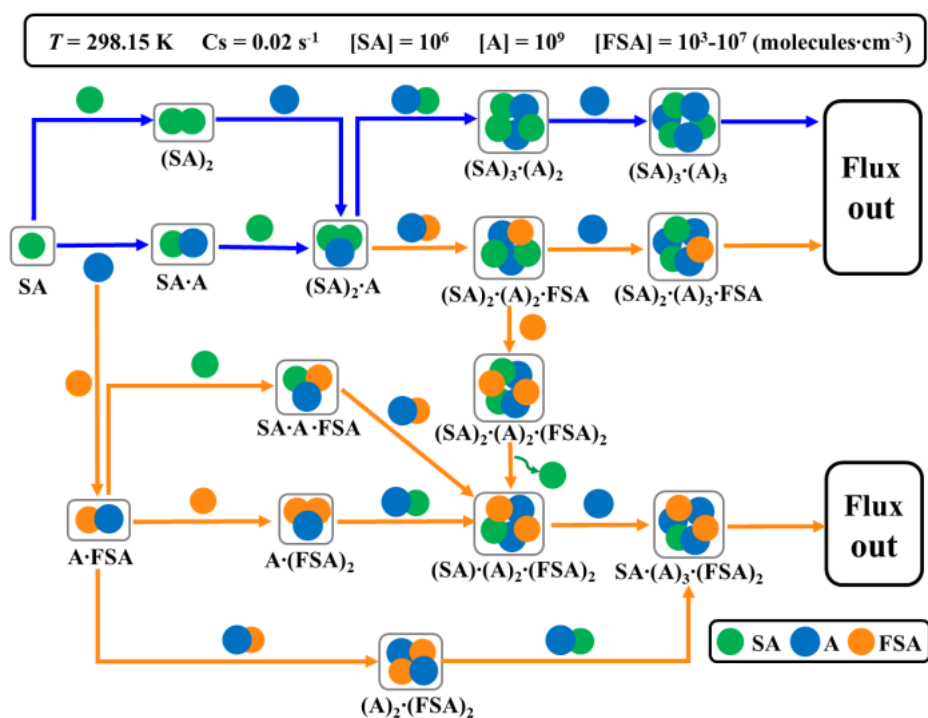


Fig. S15. Main pathways of clusters growth under conditions of $T = 298.15 \text{ K}$, $[\text{SA}] = 10^6$ molecules·cm⁻³, $[\text{A}] = 10^9$ molecules·cm⁻³, and $[\text{FSA}] = 10^3\text{-}10^7$ molecules·cm⁻³. The blue and orange fluxes represent the pathways of the SA-A-based and SA-A-FSA-based cluster, respectively

Table S14. Cartesian coordinates of all molecules and clusters in this study

SA

Atom	X	Y	Z
S	-0.00000100	0.00000200	-0.15545600
O	0.65765000	-1.06508500	-0.82224200
O	-0.65766000	1.06510800	-0.82220400
O	1.02424100	0.67764400	0.84020100
O	-1.02423000	-0.67766800	0.84019400
H	1.69311900	0.03016000	1.09984500
H	-1.69310700	-0.03019100	1.09986300

A

Atom	X	Y	Z
N	0.00000000	0.11213000	0.00000000
H	-0.94020500	-0.26169700	0.00000000
H	0.47010300	-0.26160800	0.81428100
H	0.47010300	-0.26160800	-0.81428100

FSA

Atom	X	Y	Z
S	-0.73245600	-0.00003100	0.084092000
O	-1.86438700	-0.67719800	-0.41027000
O	-0.37942100	1.13847800	-0.92486400
O	-0.61218900	0.46342500	1.41730000
O	1.78817900	-0.59587800	-0.05562900
H	0.51177600	-1.04252700	-0.16626600
C	2.47070900	-1.44826000	-0.09353000
H	2.10711400	0.54561400	0.04517600
O	0.41637300	1.60168100	-0.60677600

SA·A

Atom	X	Y	Z
S	-0.59827600	-0.11336800	0.08617500
O	0.10893200	-0.09214100	1.32834700
O	-1.74906500	-0.92894600	-0.10565200
O	0.39576600	-0.37422800	-1.06128100
O	-1.03681100	1.39710200	-0.16132600
H	1.37463300	-0.19089300	-0.73462700
H	-1.78503200	1.40349300	-0.77106700
N	2.73855300	0.04357600	-0.05140800
H	3.20053200	0.91724700	-0.27070200
H	3.40981800	-0.70549900	-0.16620600

209		H	2.45200800	0.07021600	0.92295000
210	FSA·A				
		Atom	X	Y	Z
		S	-2.06986300	-1.43720000	-0.02487300
		O	-2.28947300	-0.45708400	0.19881300
		O	-1.74977800	-1.89068700	0.82786100
		O	-2.84681600	-1.92440600	-0.45419300
		O	0.87085200	-0.31194100	0.02282000
		H	2.28806200	-0.35459200	-0.03984100
		H	0.16258200	-0.81847000	-1.16009200
		N	0.21548900	-0.69441000	1.25156800
		H	-0.61216000	1.88395400	-0.03391400
		H	0.60096100	1.37010700	-0.07332800
		C	-0.55396200	2.97630800	-0.08844400
		H	-1.65868000	1.29398800	0.04496400
		O	-1.19891900	-1.32936600	-0.65773400
211					
212	(SA) ₂				
		Atom	X	Y	Z
		S	2.03944100	0.07517400	0.11329500
		O	1.06831400	-0.09670000	1.16086600
		O	3.31172000	0.62626400	0.39691600
		O	2.20680700	-1.37335100	-0.48979500
		H	2.98774000	-1.40489100	-1.05933100
		O	1.41198000	0.87974800	-1.04535300
		H	0.44673500	0.64256800	-1.15033500
		O	-1.06831700	0.09668400	-1.16087000
		S	-2.03944300	-0.07517400	-0.11329500
		O	-1.41198700	-0.87975200	1.04535400
		O	-3.31172900	-0.62625000	-0.39691000
		O	-2.20678900	1.37335500	0.48979100
		H	-0.44674000	-0.64257900	1.15033500
		H	-2.98771900	1.40490700	1.05933100
213					
214	(FSA) ₂				
		Atom	X	Y	Z
		S	-2.10673800	-0.64032400	-0.24218500
		O	-3.16703900	-1.51334400	-0.55398700
		O	-1.52529400	-0.99650900	1.13623900
		O	-1.04397200	-0.39901200	-1.17532800
		C	-2.13419400	1.83567000	0.56745300

O	-2.82560300	0.78565000	0.01405000
H	-2.78175100	2.71571600	0.54331000
O	-1.03853400	1.77228500	1.00367700
H	-0.58548000	-0.67711000	1.22411800
S	2.10673800	-0.64032400	0.24218400
O	3.16703900	-1.51334400	0.55398700
O	1.52529400	-0.99650900	-1.13623900
O	1.04397200	-0.39901200	1.17532900
C	2.13419400	1.83567000	-0.56745300
O	2.82560300	0.78565000	-0.01405100
H	2.78175100	2.71571600	-0.54331100
O	1.03853300	1.77228500	-1.00367600
H	0.58547900	-0.67711100	-1.22411600

215

216 SA·FSA

Atom	X	Y	Z
S	2.22843800	-0.00423900	0.07863900
O	3.23791900	0.97524400	0.24010300
O	1.27077300	-0.25369000	1.12244200
O	2.90768300	-1.40834000	-0.15214600
O	1.49401400	0.26141300	-1.25288200
H	3.74994700	-1.28862800	-0.61125600
H	0.56250200	-0.08538900	-1.23312600
S	-1.93817600	-0.60901300	-0.05970300
O	-3.06674300	-1.45127200	-0.05756800
O	-1.24859400	-0.62326500	1.30624500
O	-0.95965800	-0.70137600	-1.10956400
C	-1.73682200	1.97561700	-0.03610200
O	-2.56398900	0.88882400	-0.12069700
H	-2.34435400	2.88083400	-0.11348900
O	-0.56291500	1.92177700	0.10376500
H	-0.25926600	-0.42300400	1.23392100

217

218 (SA)₂·A

Atom	X	Y	Z
S	-1.77231300	-0.34077900	-0.04909000
O	-1.07131700	0.16352900	1.14296700
O	-0.99847000	-1.34675900	-0.75741300
O	-3.07876600	-1.07528900	0.45986500
H	-2.85067000	-1.96938800	0.74413600
O	-2.28848200	0.74869000	-0.85700500
H	-1.40689900	2.09302200	-0.44210900
O	1.42804700	1.08270500	-0.53209900

S	2.06372500	-0.11895200	-0.01742600
O	1.43416000	-0.42809800	1.36683400
O	3.47167000	-0.14405700	0.10310200
O	1.62893900	-1.30669600	-0.91181400
H	0.44446700	-0.24695300	1.34915800
H	0.63395000	-1.35519900	-0.95819300
N	-0.64527900	2.68400200	-0.01766800
H	0.24045900	2.13587500	-0.15608200
H	-0.82981700	2.75595300	0.97931500
H	-0.58337400	3.60217400	-0.44379900

219

220 SA·A·FSA

Atom	X	Y	Z
S	0.55397800	2.48299400	-0.70439700
O	0.45073600	3.03570100	0.14224400
O	-0.24090200	1.81899900	-0.72850000
O	0.55144200	3.07525400	-1.52784700
O	2.13527700	-0.46863900	-0.40785800
H	3.00805500	-1.52978200	-0.75383900
H	2.68481300	0.87791700	-0.40902500
N	0.80219000	-0.48805700	-1.02270300
H	1.05345300	0.02665000	1.93691500
H	1.81162200	-0.79790800	1.20606800
S	0.96935400	-0.36474900	2.95539900
O	0.53492900	1.03427100	1.55561300
O	1.44609600	1.92109200	-0.62581400
O	-2.37916900	-0.14053600	-0.01737800
O	-1.84312000	1.07847500	-0.55578700
H	-3.21751400	-0.11977900	1.12909100
H	-1.25121700	-1.15558600	0.21694100
C	-3.19108800	-0.76047000	-1.22541400
H	-0.40838300	-0.93672100	-0.33532800
O	-3.77401000	-1.45628600	-0.89463100

221

222 (FSA)₂·A

Atom	X	Y	Z
N	0.53599000	1.62901700	1.53538100
H	1.33593800	1.86879000	0.92730000
H	0.73156800	0.67470000	1.86455300
H	0.44707100	2.27841600	2.30898800
S	1.82396600	-0.88183100	-0.13231500
O	1.80719000	-2.19493100	-0.66933400
O	0.78405400	0.02834200	-0.65823700

O	2.03005200	-0.70863300	1.28211100
C	3.54095800	1.04184400	-0.61837800
O	3.20543000	-0.22448100	-0.83384200
H	4.47964800	1.26606200	-1.13446400
O	2.93793700	1.85135900	0.03095800
H	-0.33133500	1.59116000	0.97715900
S	-2.52000400	0.36564800	-0.62012600
O	-3.66487200	0.56839600	-1.41984700
O	-1.50362100	-0.51119700	-1.31541400
O	-1.89506900	1.47643000	0.04285400
C	-2.24247400	-1.05306800	1.54702500
O	-3.07199100	-0.63666600	0.56714800
H	-2.78840400	-1.72915800	2.21135200
O	-1.10228300	-0.73486600	1.67210600
H	-0.49533300	-0.33683700	-1.02340400

223

224 (SA)₃

Atom	X	Y	Z
S	0.17750700	1.30695000	-0.05944900
O	-0.09390200	0.06719200	-0.78009000
O	1.37463700	1.98332600	-0.44312400
H	3.10843900	1.28610800	-0.37359300
O	0.17205700	0.99707200	1.45713200
O	-1.00451000	2.25666600	-0.18455500
O	3.82087700	0.61891100	-0.31555500
S	3.23113400	-0.76663900	0.09084400
O	4.31169100	-1.65664100	0.25254200
O	2.42122500	-1.20817600	-1.17323700
O	-2.41639300	-1.20536900	-1.04774400
H	1.51320800	-0.84728200	-1.14877900
H	-1.52390100	-0.77676800	-0.99149200
O	2.28428500	-0.56330900	1.16270100
O	-3.14100400	0.86502300	0.01296900
H	0.93006700	0.38422300	1.63170700
H	-1.88155500	1.76156800	-0.09917000
O	-4.69818200	-1.03136200	-0.32527500
S	-3.39889000	-0.55000200	-0.04775200
O	-2.91048600	-1.08369200	1.35077700
H	-3.26462400	-1.96992300	1.51070800

225

226 (FSA)₃

Atom	X	Y	Z
S	-3.23194400	-0.59637700	-0.25851000

O	-4.51319100	-0.86236400	-0.79376300
O	-2.16008800	-1.00223500	-1.29416100
O	-2.89865700	0.66721800	0.37672000
C	-1.85890000	-1.89976900	1.47204800
O	-3.06403200	-1.70774200	0.87655800
H	-1.98901700	-2.60927700	2.25876100
O	-0.84291000	-1.38849500	1.16683800
H	-1.29933600	-0.54007000	-1.10837700
S	0.10009300	2.06276100	-0.68190000
O	0.68944600	2.80847500	-1.72092700
O	-1.24487000	2.69328800	-0.27988000
O	-0.06913400	0.63672600	-0.77376700
C	0.63470800	1.90232600	1.85577600
O	1.02955900	2.34576900	0.60712100
H	1.44694200	2.12286300	2.55616800
O	-0.41216400	1.38226800	2.09994300
H	-1.84632800	2.00376500	0.11677500
S	3.07446000	-0.57195000	0.10911000
O	4.22086400	-0.50151200	0.92477800
O	1.84116100	-0.60394300	1.05586400
O	2.81545200	0.29037500	-0.98445400
C	2.06960000	-2.60418700	-1.11004100
O	3.15002600	-2.10354300	-0.48818400
H	2.36905000	-3.56437900	-1.57315100
O	0.97890500	-2.08765900	-1.12358800
H	1.00155600	-0.64709400	0.55113200

227

228 (SA)₂·FSA

Atom	X	Y	Z
S	0.31393300	0.27128100	0.80272400
O	1.41010300	1.14724900	1.09984700
O	-0.86402600	0.38006700	1.61620000
O	0.76621500	0.97877800	-0.09423400
O	-0.03223000	-1.06706500	-0.95553900
H	1.68969000	-1.50955800	-0.84556300
H	-0.90520200	-1.19916600	0.89051400
S	-3.54538300	0.50006100	-0.68317400
O	-4.49435400	-1.30305700	0.52329200
O	-3.27097300	0.08072500	-0.90098400
O	-2.31119600	-0.76729700	-0.24322500
C	-3.72422400	-1.80240900	-0.34368500
O	-4.34249300	-0.42685700	1.22451000
H	-4.41327900	-0.82986600	-0.97754600

O	-2.62724100	1.77061100	-0.73336400
H	-2.33757500	0.55060600	-0.76049900
S	4.17910500	2.51668200	-1.13704700
O	5.55665000	1.96342400	-0.33226800
O	3.19718500	-0.06954300	1.36773100
O	3.73034400	-0.24299300	-0.32449600
O	3.99097900	-0.56491400	-0.36294300
H	4.45057900	-1.25396600	-0.04119100
H	3.02693100	0.34216500	-1.71845100

229

230

SA·(FSA)₂

Atom	X	Y	Z
S	0.00000300	-0.00002800	0.64830500
O	-0.75790100	0.97127900	-0.09591600
O	0.75794500	-0.97141300	-0.09578700
O	-0.93532600	-0.74113000	1.61626600
O	0.93529600	0.74117800	1.61622700
H	-1.89125600	-0.51059200	1.45471200
H	1.89122100	0.51056900	1.45478500
S	4.12652700	-0.41133700	0.05570600
O	5.38190600	-1.04011800	0.15954000
O	3.18315600	-1.22394000	-0.83248000
O	3.43564500	0.04122300	1.23295100
C	3.38169200	1.71437500	-1.22313300
O	4.41750700	0.90281500	-0.85685100
H	3.78099100	2.54795700	-1.80636700
O	2.24469900	1.52681200	-0.94873200
H	2.21091700	-1.07916900	-0.58578700
S	-4.12653200	0.41133400	0.05572300
O	-5.38190500	1.04011300	0.15962200
O	-3.18316800	1.22400700	-0.83241000
O	-3.43564800	-0.04131800	1.23293200
C	-3.38168200	-1.71429500	-1.22323100
O	-4.41750600	-0.90274600	-0.85692800
H	-3.78097400	-2.54781300	-1.80656000
O	-2.24470000	-1.52677500	-0.94875300
H	-2.21093000	1.07916500	-0.58578600

231

232

(SA)₂·(A)₂

Atom	X	Y	Z
S	-2.06184100	0.03024000	-0.13465200
O	-2.04117600	1.44224200	0.23783700
O	-3.44180400	-0.18999500	-0.89528300

O	-1.07447700	-0.35111100	-1.12711700
O	-2.07527000	-0.85956100	1.02215600
H	-0.62565200	2.17366500	0.71126100
H	-4.16339900	0.11236500	-0.33044200
S	2.14824600	-0.06917100	-0.20933500
O	1.98617900	-1.44915800	-0.67010100
O	1.31072900	0.84270300	-1.21025200
O	1.44496800	0.08726200	1.09667700
O	3.46837600	0.47040600	-0.23135200
H	0.82649800	-2.09927000	0.18550400
H	0.42274300	0.42956900	-1.33621000
N	0.16605000	-2.42220400	0.96179200
H	-0.03648600	-3.41297000	0.89015000
H	0.64646900	-2.21664600	1.83389300
H	-0.71778100	-1.86109400	0.94374900
N	0.35020200	2.43893700	0.98896700
H	0.35301500	3.05370700	1.79434900
H	0.86196200	1.50602200	1.17669400
H	0.81620300	2.87810800	0.19901900

233

234 SA·(A)₂·FSA

Atom	X	Y	Z
N	-0.56014400	-2.50162300	-1.18465600
H	-0.51312700	-3.31715500	-0.58087000
H	0.24189900	-1.86322200	-0.91499800
H	-0.49089300	-2.76590600	-2.16129700
S	-2.01210800	0.36785000	-0.31238200
O	-2.62217700	1.67359200	-0.33240400
O	-2.86486200	-0.78410300	-0.47660100
O	-0.76692500	0.28066100	-1.05249400
C	-0.91367900	-0.71721100	1.84228300
O	-1.51723600	0.35112600	1.30152900
H	-0.57047200	-0.46665600	2.84999400
O	-0.78402900	-1.78897100	1.33362300
H	-1.44645400	-2.00919800	-0.99711800
S	2.27254000	-0.09072400	0.06125900
O	1.71174100	-1.31696200	-0.48662400
O	3.65606800	-0.17762300	0.42225600
O	1.39806600	0.55074600	1.04932200
O	2.21153500	0.99868500	-1.15471900
H	0.58368300	1.79927500	0.53551600
H	3.04161700	0.92588300	-1.63930400
N	0.14155800	2.61144100	-0.00672300

H	0.55313300	3.49885700	0.25954500
H	-0.88044200	2.61495600	0.10152100
H	0.34885200	2.38648600	-0.98017800

235

236 (A)₂·(FSA)₂

Atom	X	Y	Z
N	0.51148200	2.21605000	0.26359000
H	1.25940400	2.30709700	-0.42793300
H	0.90111700	1.54792200	0.95568500
H	0.28671200	3.11166600	0.68183300
S	2.00305500	-0.65065500	0.17540100
O	1.96884000	-2.07899400	0.23914100
O	1.28396600	-0.07317100	-0.95522100
O	1.80990200	0.07670500	1.41292700
C	4.01436400	0.89889400	-0.42278200
O	3.61354300	-0.35473100	-0.20718100
H	5.08904000	0.90567600	-0.63275600
O	3.33419100	1.88415700	-0.40110200
H	-0.32876000	1.77240300	-0.18012500
S	-2.85590100	0.42548300	-0.36412200
O	-3.93486700	1.34816000	-0.43110200
O	-3.12220300	-0.94470500	-0.76286000
O	-1.56411400	0.89502700	-0.85432000
C	-1.73688800	-0.53841800	1.81611900
O	-2.61719500	0.32824700	1.31127800
H	-1.77253100	-0.50324200	2.91060600
O	-1.00327200	-1.24862300	1.19472600
H	-1.64200600	-1.71722000	-1.14245700
N	-0.65735500	-1.99590300	-1.35904500
H	-0.07627600	-1.13464800	-1.37477700
H	-0.58601000	-2.49305000	-2.23965100
H	-0.27922600	-2.55431300	-0.59300400

237

238 (SA)₃·A

Atom	X	Y	Z
N	-0.08306900	0.43034300	2.52884500
H	-0.96686500	0.21892000	2.01889600
H	0.62338000	-0.24748900	2.20270400
H	-0.22034100	0.36070000	3.53139300
S	1.14064300	1.97416100	-0.15831000
O	0.06648000	0.98458200	-0.29616000
O	1.25367800	2.50665600	1.17335800
H	0.26824900	1.36202900	2.25930200

O	2.38467600	1.45066900	-0.74121700
S	-2.89899900	-0.31614900	-0.11437500
O	-2.20892100	-1.35592600	-1.04413200
O	-2.49532900	1.05527200	-0.72151700
O	-4.29806300	-0.45733500	-0.25192500
O	-2.29521200	-0.43001100	1.19720400
H	-1.28439900	-1.55011900	-0.76684000
H	-1.51456700	1.18329100	-0.61120100
S	1.62500000	-1.74705900	-0.06495200
O	2.49474900	-3.05585900	-0.03919200
O	2.25892200	-0.95907300	-1.19405200
O	0.28429300	-2.14902000	-0.39499600
O	1.85113600	-1.10561700	1.19884400
H	2.13547100	-3.69945900	-0.66426200
H	2.33436100	0.08623200	-0.99483800
O	0.75413400	3.22380900	-1.04837100
H	0.93553700	3.03305100	-1.97763000

239

240 (SA)₂·A·FSA

Atom	X	Y	Z
N	-0.37923500	-1.61848700	0.51393000
H	-0.72962600	-2.40970400	1.04954400
H	0.61354800	-1.73754800	0.24637300
H	-0.97316100	-1.47534100	-0.31706500
S	-0.23533500	1.63051300	-0.41786400
O	-0.59344800	2.73170700	-1.26140300
O	-1.27494000	1.13832600	0.51765900
O	0.48704200	0.49589900	-1.04079700
C	1.47787000	1.52350300	1.56386900
O	0.92242100	2.30063300	0.61194800
H	2.24428000	2.09621900	2.09896000
O	1.18910400	0.37975000	1.79207600
H	-0.43577500	-0.75476800	1.07033400
S	3.46041000	-0.86448500	-0.13695200
O	2.35495100	-1.80366900	-0.11698500
O	4.25092200	-0.61065900	1.03805700
O	2.97998300	0.49720400	-0.72930600
O	4.43249900	-1.41597100	-1.27920500
H	1.98448300	0.47621100	-0.98085100
H	5.32608200	-1.07672400	-1.11191400
S	-3.72981600	-0.75109400	-0.18716200
O	-2.66648800	-1.11059000	-1.10242900
O	-5.07856600	-1.19843800	-0.37289200

O	-3.75240800	0.78833300	0.05631900
O	-3.18410400	-1.31740600	1.23325100
H	-2.79817900	1.10930800	0.18164000
H	-3.90410900	-1.27914400	1.88357000

241

242 SA·A·(FSA)₂

Atom	X	Y	Z
N	-0.12726200	-1.84684600	-0.42365600
H	0.30849300	-1.60355300	0.47046200
H	-0.28726300	-0.95610800	-0.90261100
H	0.51118100	-2.42022500	-0.96687400
S	-3.40300200	-0.97176800	-0.54312600
O	-4.70177800	-0.85341600	-1.09550800
O	-2.88019700	-2.29154100	-0.28317200
O	-2.36887300	-0.11436700	-1.18227100
C	-2.55236400	-0.17279100	1.80472900
O	-3.58880700	-0.26086200	0.95576800
H	-2.88968400	0.28121100	2.74123200
O	-1.43280000	-0.52042400	1.58134300
H	-1.05776800	-2.27053000	-0.27560300
S	-0.25221100	2.13698400	0.04135500
O	0.43061200	0.96247800	-0.48801800
O	0.04848200	2.56374800	1.36310400
O	-1.73585800	1.98059400	-0.17501200
O	0.16874700	3.28151800	-0.96129200
H	-2.04361000	1.05180300	-0.67223200
H	0.08108400	4.14035300	-0.52685300
S	3.56124100	-0.57648400	-0.61780300
O	4.82862700	-0.50502300	-1.23130200
O	2.96657000	0.82490200	-0.47074200
O	2.56900100	-1.51049300	-1.04742500
C	2.94634400	-1.06786900	1.86862000
O	3.92467600	-0.93558800	0.94565400
H	3.40228000	-1.26938700	2.84190000
O	1.77876500	-0.97724300	1.66017700
H	1.94844500	0.84432700	-0.44116800

243

244 A·(FSA)₃

Atom	X	Y	Z
N	-0.31846600	-1.49948400	0.67567200
H	-1.00334800	-2.06692100	0.16388500
H	-0.77180200	-1.15287300	1.52238100
H	0.54405300	-2.00898400	0.86222200

S	-3.42516100	-0.19026100	0.29706900
O	-4.27466000	0.93609500	0.38980700
O	-2.28213200	0.00913100	0.69515100
O	-2.97552600	-0.87595500	1.46499600
C	-3.74718800	-2.48517100	0.88445500
O	-4.29436800	-1.29629000	0.56047200
H	-4.49879900	-3.09560300	1.39193400
O	-2.62659800	-2.82814200	0.66717500
H	-0.06789300	-0.67510200	0.10923100
S	0.26628700	2.09574900	0.91638700
O	1.03928000	2.96583600	1.72091300
O	-1.18402100	2.17778300	1.04326400
O	0.70654400	0.68857800	0.88803900
C	0.00065800	2.05210700	1.67455700
O	0.58458700	2.63920000	0.61320300
H	0.33227000	2.55245000	2.58850500
O	-0.75973100	1.13591700	1.63079200
H	-1.89658100	0.98841500	0.80336800
S	3.48657100	-1.27416200	0.43973400
O	4.62554500	-1.84089600	1.04710800
O	3.11859600	0.03917700	1.11501600
O	2.32543100	-2.06055300	0.15648600
C	3.25522000	-0.10370000	1.87784200
O	4.06376600	-0.74995200	1.01519600
H	3.83056200	0.17029600	2.76677800
O	2.10010100	0.13305200	1.70383000
H	2.13097500	0.32021800	-1.00381200

245

246 (SA)₃·(A)₂

Atom	X	Y	Z
N	0.00518100	-2.07016200	0.13235300
H	-0.75853200	-2.16079900	-0.55462500
H	0.93474300	-1.88806300	-0.30515600
H	0.03719300	-2.89913000	0.71738900
N	-2.34720800	1.90536900	1.66490700
H	-3.28300000	2.15582800	1.35682500
H	-2.15626000	2.28748100	2.58494300
H	-2.28784300	0.86629500	1.66909200
S	-3.05154400	-0.82064200	-0.13076300
O	-2.54077100	-1.86675900	-0.99529000
O	-2.97918700	0.51386700	-0.72804800
O	-2.49768700	-0.83783600	1.22061400
H	-1.36657100	0.64835200	-1.39460500

S	0.28837400	1.51641400	-0.40036800
O	-0.38382400	0.61467600	-1.49818000
O	1.58674500	1.85904500	-0.92585500
O	-0.60465300	2.64712900	-0.17390700
O	0.36418400	0.67575900	0.81966400
H	-0.22309800	-1.24829000	0.70426700
H	-1.64624200	2.26948800	0.94588000
O	-4.59477800	-1.12522500	0.10609000
H	-5.01675300	-1.29530900	-0.74583900
S	3.45174800	-0.69237600	0.13959600
O	3.87133700	0.63659300	-0.53293900
O	2.71916700	-0.25160100	1.44404300
O	4.61983900	-1.39864900	0.50930300
O	2.47134100	-1.33631400	-0.71283100
H	3.05627700	1.14908100	-0.79026500
H	1.83331600	0.16880500	1.23453400

247

248 (SA)₂· (A)₂·FSA

Atom	X	Y	Z
S	2.40391200	-1.07942500	-0.36129600
O	1.71053700	-1.25313700	-1.62432800
O	3.93485000	-1.39284600	-0.64312600
O	2.45422600	0.30895400	0.10438500
O	1.97479500	-1.98921100	0.68846800
H	0.16014500	-0.66201200	-2.06959300
H	4.02118800	-2.30963400	-0.93347400
S	0.13090500	2.77099300	0.23948700
O	0.52718100	2.22941500	1.51602800
O	0.95491200	2.04616800	-0.86784700
O	-1.32140800	2.28625400	-0.08580200
O	0.15960200	4.16954900	0.02567800
H	0.71912300	0.51593200	2.14015500
H	1.59722700	1.33912600	-0.47051200
N	0.82421100	-0.40948600	2.59019800
H	1.30829900	-0.31796500	3.47661200
H	-0.09695100	-0.84199200	2.70844700
H	1.35056700	-1.03668200	1.93661900
N	-0.54682000	0.00432200	-2.44844900
H	-1.48253800	-0.21180200	-2.07199100
H	-0.26303100	0.93496800	-2.12694200
H	-0.55070900	-0.03008600	-3.46270000
S	-1.51508900	-1.52449700	0.46380600
O	-1.31946500	-2.13805600	1.74464700

O	-0.84361300	-0.22355000	0.34583800
O	-1.43521900	-2.32475700	-0.71761100
C	-3.73537000	-0.45623000	-0.42866100
O	-3.15087300	-1.09347000	0.57740100
H	-4.80124800	-0.32047400	-0.22130800
O	-3.20123700	-0.05980200	-1.42839300
H	-1.36154100	1.31689700	0.11975700

249

250 SA·(A)₂·(FSA)₂

Atom	X	Y	Z
N	0.88963900	0.53209400	2.15145900
H	1.84966900	0.41081100	1.79133900
H	0.83420700	0.13754200	3.08575900
H	0.62595400	1.51890800	2.14764400
S	2.05308100	-1.52499800	-0.28033400
O	1.86208800	-2.52535200	-1.29162100
O	1.56829600	-0.21036900	-0.69826200
O	1.74324300	-1.87586600	1.07550400
C	4.24923600	-0.40930500	0.52002400
O	3.72193000	-1.32633400	-0.28834800
H	5.33771900	-0.39884700	0.41336200
O	3.64710000	0.31648900	1.26178000
H	0.22502200	0.02316400	1.53083500
S	-2.47028500	-0.12945100	0.31801600
O	-2.80915800	1.05221100	1.08298300
O	-2.93186600	-0.17716900	-1.03705100
O	-1.09183800	-0.56319500	0.51862600
C	-3.25017700	-2.56141200	0.62325600
O	-3.35078200	-1.31108700	1.10186400
H	-3.89023600	-3.22717300	1.20973100
O	-2.57184200	-2.91559000	-0.29470100
H	-1.31172900	-1.82703100	-1.49415100
N	-0.68616600	-1.32213500	-2.13810800
H	-0.53976600	-0.38179800	-1.74321300
H	-1.13732600	-1.23961000	-3.04422900
H	0.21823700	-1.80568400	-2.19540900
S	-0.06455200	2.72561200	-0.53080200
O	-0.04785500	4.08602500	-0.92213700
O	-0.64874400	1.72545900	-1.38120900
O	-0.72154700	2.61421300	0.89639100
O	1.39733200	2.30480600	-0.18118300
H	-1.58974300	2.11178800	0.89308600
H	1.53655800	1.33998700	-0.39906800

251

252

(A)₂·(FSA)₃

Atom	X	Y	Z
N	-0.11133900	0.24657900	-2.28896400
H	0.82895800	-0.08629700	-2.04773100
H	-0.18294700	1.22685200	-1.99200100
H	-0.28175800	0.14911400	-3.28384500
S	0.08139700	2.06744100	0.56332000
O	-0.39544000	2.37514200	1.87256700
O	0.51543200	0.65888700	0.41907100
O	-0.67437700	2.51594000	-0.57506900
C	2.29126000	2.79019200	-0.60671400
O	1.52064500	2.89950300	0.49399800
H	3.16765500	3.43567800	-0.49670300
O	2.06737500	2.09492700	-1.54901700
H	-0.80364500	-0.30930700	-1.72586000
S	-3.15708200	-1.35264700	-0.52722200
O	-3.67483200	-2.14280200	-1.59065400
O	-3.57688500	-1.66484300	0.82116000
O	-1.72160600	-1.05739200	-0.59577100
C	-3.67905300	1.17348500	-0.02743400
O	-3.85405900	0.14547100	-0.86392000
H	-4.21642800	2.04886700	-0.40686200
O	-3.04040600	1.16497000	0.98177900
H	-2.31472000	-0.95063500	1.98048600
N	-1.36613600	-0.58667900	2.17802500
H	-0.85635900	-0.64272800	1.28578500
H	-0.87118200	-1.13454600	2.87367000
H	-1.41779800	0.40307900	2.43147200
S	3.20510500	-1.11588600	-0.33809600
O	4.57086000	-1.07250000	-0.69183800
O	2.87098500	-0.02555500	0.67088900
O	2.18193400	-1.22709200	-1.33181300
C	2.00153200	-2.76533200	1.30340800
O	3.10048700	-2.48276700	0.57126600
H	2.13258000	-3.74911100	1.76309100
O	1.05323000	-2.06021800	1.44114400
H	1.85807900	0.24373200	0.62113600

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254

(SA)₃·(A)₃

Atom	X	Y	Z
S	2.61550300	-1.21713100	0.07787100
O	1.50419700	-2.11956500	0.33153900
O	3.94552200	-2.09812300	0.14786300

O	2.66374100	-0.67863000	-1.27165400
H	3.94723000	-2.59642300	0.97400000
S	-2.80519300	-0.98920500	-0.06753700
O	-2.88751900	0.34179100	0.50484700
O	-4.14297800	-1.67881400	0.45755400
O	-2.85144000	-1.05270500	-1.52017700
H	-4.26188300	-2.51704700	-0.00442200
S	-0.33046500	2.24661500	0.03312700
O	0.21614600	0.92532100	-0.34071300
O	-0.53745000	2.32266200	1.47103800
O	-1.71152100	2.40034500	-0.67450000
O	0.49429700	3.31883300	-0.51434200
H	1.96174700	2.66009600	-0.62515700
O	2.74348300	-0.20042500	1.12633500
N	-0.09442900	-0.29203800	2.17408000
N	2.82992600	2.04774500	-0.59450900
N	-0.09600400	-1.28903300	-1.85188200
H	3.67861200	2.57105100	-0.77685000
H	2.87821600	1.57373000	0.31751000
H	2.72943200	1.26102800	-1.24862100
H	-2.31296100	1.69948700	-0.30930200
H	0.20493900	-1.93442300	-1.11044700
H	0.50390300	-1.39420000	-2.66253600
H	-1.09902400	-1.41255800	-2.04950500
H	0.02767100	-0.33907600	-1.43743600
H	0.89281000	-0.35062400	1.89832100
H	-0.65647300	-0.89341900	1.53205000
H	-0.39668000	0.69889100	2.05200600
H	-0.21685000	-0.59772400	3.13279700
O	-1.69080900	-1.77318400	0.47325800

255

256 (SA)₂·(A)₃·FSA

Atom	X	Y	Z
S	2.54617000	-1.61306500	0.17337400
O	2.53073400	-1.36697700	-1.25655200
O	3.45690900	-2.91890000	0.29359500
O	3.20340600	-0.57689400	0.96273600
O	1.24418100	-1.98353300	0.72922600
H	-0.06330300	-1.89373500	-0.74184800
H	3.48671000	-3.19854000	1.21636200
S	0.53066300	2.33941500	-0.17921600
O	0.17450500	2.49218600	1.22770400
O	0.77296300	0.92495900	-0.50459000

O	-0.68640100	2.81865800	-1.03655700
O	1.63632400	3.18459700	-0.60929500
H	0.15632000	1.05760800	1.94961500
H	3.03934200	0.60258500	-1.61078700
N	0.13954700	0.09425900	2.39442900
H	0.71510400	0.08969000	3.22966200
H	-0.82844200	-0.14317700	2.61268200
H	0.49563400	-0.60542600	1.71987000
N	-0.34546600	-1.35648200	-1.57066200
H	-1.35760200	-1.45842700	-1.71549200
H	-0.11532200	-0.37569100	-1.32520200
H	0.21126100	-1.64797400	-2.36726500
S	-2.61335700	-0.05628800	0.52624900
O	-2.95163800	0.67828500	1.70612200
O	-2.31535000	0.72497100	-0.66244500
O	-1.71447800	-1.17225400	0.73835200
C	-4.13486100	-1.58315700	-0.94138500
O	-4.07585500	-0.78824800	0.12364400
H	-5.15461800	-1.96391800	-1.05876800
O	-3.23553400	-1.86575600	-1.68171200
H	-1.43543700	2.19580100	-0.88160100
N	3.50117100	1.31384300	-1.03524100
H	2.86161200	2.14900800	-0.93597000
H	4.40902100	1.55980500	-1.41211400
H	3.58837200	0.84024400	-0.11885000

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258 SA·(A₃)₂(FSA)₂

Atom	X	Y	Z
N	0.83387200	-0.43401100	2.18783400
H	1.83389100	-0.65793000	2.09210800
H	0.55630300	-0.36783400	3.16077200
H	0.60144600	0.45302000	1.68709900
S	2.70283700	-0.48528200	-0.63007100
O	2.88110700	-0.34723300	-2.03825700
O	2.72100500	0.72551700	0.16993400
O	1.68179000	-1.43461900	-0.21621700
C	4.33574600	-1.55761700	1.10917500
O	4.13897600	-1.24754600	-0.16631800
H	5.31915900	-2.02416100	1.22792800
O	3.58011800	-1.37482900	2.02297600
H	0.27895200	-1.14925000	1.70325800
S	-2.44346000	-0.75224800	0.58884300
O	-2.78436500	-0.16036100	1.85451000

O	-2.67741800	0.09593000	-0.56675100
O	-1.21277000	-1.50876600	0.54864300
C	-3.63487700	-2.70660000	-0.65158100
O	-3.62775700	-1.93907500	0.44108300
H	-4.45210500	-3.43205200	-0.58213000
O	-2.88250100	-2.63845300	-1.57915900
H	0.25793400	-1.47566400	-1.39132900
N	-0.38344300	-1.06744000	-2.08835900
H	-0.41701600	-0.04875900	-1.86347800
H	-1.31050300	-1.49358200	-1.99162100
H	0.00156300	-1.19188200	-3.01919000
S	0.00000200	2.38018500	-0.34011300
O	-0.91643900	3.50541100	-0.51249300
O	0.12099600	1.52027000	-1.50393100
O	-0.32908200	1.63241100	0.88522400
O	1.39814700	3.05241500	-0.12406800
H	-2.61835800	2.10423700	1.57133600
H	2.06716500	2.33176000	-0.11600600
N	-3.00983500	2.53990000	0.73431700
H	-2.23497100	3.10334500	0.26088200
H	-3.83550100	3.08519800	0.95075400
H	-3.22184200	1.74865900	0.10755200

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260

(A)₃·(FSA)₃

Atom	X	Y	Z
N	0.43250300	0.17461100	-1.90860200
H	0.10701400	1.14755100	-1.73877900
H	-0.31737800	-0.45690900	-1.59577500
H	0.61293800	0.03033900	-2.89673700
S	-2.25698800	-1.81816200	0.04075300
O	-1.44494000	-3.00259800	-0.00029100
O	-2.33250900	-1.17406200	1.33768600
O	-2.07128100	-0.89108400	-1.06173500
C	-4.85272000	-1.67047200	-0.27173300
O	-3.77813400	-2.46023900	-0.21420500
H	-5.74320400	-2.28242200	-0.44533700
O	-4.87464800	-0.48030200	-0.15658100
H	1.30592500	0.00696000	-1.37120700
S	3.70101600	-0.65750200	0.17026800
O	5.07379000	-0.31215000	0.26556000
O	3.06031900	-1.10038400	1.40161500
O	2.85804500	0.21667100	-0.62798700
C	2.65607400	-2.69384300	-1.10528300

O	3.77598400	-2.08121800	-0.75588400
H	2.88569800	-3.59050600	-1.69151400
O	1.53798100	-2.34265500	-0.83717600
H	1.53197900	-1.72176300	1.60556600
N	0.51467200	-1.88462600	1.79271000
H	0.04921600	-0.97118800	1.69558700
H	0.35394200	-2.25282500	2.72446000
H	0.10351400	-2.51521000	1.09235900
S	-0.28232600	3.53522200	-0.18513700
O	0.32545100	4.72113900	-0.67222700
O	-1.23090200	3.63661400	0.89519200
O	-0.70156900	2.56942900	-1.21050900
C	0.96933200	1.56572600	1.06846000
O	1.05910300	2.76585000	0.54378400
H	1.89157400	1.30031300	1.59196500
O	0.01410300	0.82215900	0.99078000
H	-2.19413900	1.68039600	-0.42187000
N	-2.81105700	1.45973500	0.37213500
H	-2.96880000	2.33503000	0.86744900
H	-2.29677800	0.80508800	0.97083100
H	-3.66401200	0.97836400	0.07232500

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262