

Supplementary Information

The influence of gas-liquid interfacial transport theory on numerical modelling of Plasma Activation of Water

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Table 1: Gaseous and aqueous species included in the one-film and two-film models. The short-lived species only exist in the plasma region. The long-lived species exist in both the plasma and air-gap regions, and are absorbed into the liquid phase. The aqueous species exist in the liquid phase and can be desorbed back into the air gap if a negative concentration gradient occurs across the gas-liquid interface.

Air-plasma species	
Short-lived species	Long-lived species
e^- , $N(^2D)$, $N_2(A^3\Sigma)$, $N_2(B^3\Pi)$, N^+ , N_2^+ , $O(^1D)$, O_2^+ , O^+ , H_2O^+ , H , OH^+ , H^+ , H_2^+ , NO_2^+ , N_3^+ , N_4^+ , O_4^+ , N_2O^+ , NO^+ , H_3^+ , H_3O^+ , O^- , O_2^- , O_3^- , NO^- , NO_2^- , NO_3^- , H^- , OH^- , O_4^- , N_2O^- ,	N_2 , N , O_2 , O , $O_2(a^1\Delta)$, O_3 , H_2O , OH , H_2 , N_2O_5 , NO_3 , NO , N_2O , NO_2 , HNO_3 , HO_2 , N_2O_3 , N_2O_4 , H_2O_2 , HNO , HNO_2 ,
Aqueous species	
O , O_2 , $O_2(a^1\Delta)$, O_3 , OH , $ONOOH$, O_2NOOH , H , H_2 , H_2O_2 , HO_2 , HO_3 , HNO , HNO_2 , HNO_3 , N , N_2 , NO , NO_2 , NO_3 , N_2O , N_2O_3 , N_2O_4 , N_2O_5 , H^+ , O^- , O_2^- , O_3^- , OH^- , $ONOO^-$, O_2NOO^- , HO_2^- , NO_2^- , NO_3^-	

Table 2: Liquid-phase chemical reactions included in both the one-film and two-film models.

Reversible reactions				
No.	Reaction	Forward rate coefficient	Reverse rate coefficient	Source
1	$H_2O \rightleftharpoons H^+ + OH^-$	$1.90 \times 10^5 \times e^{-\frac{6800}{T_{liq}}} s^{-1}$	$1.3 \times 10^{11} M^{-1} s^{-1}$	[1]
2	$H_2O_2 \rightleftharpoons H^+ + HO_2^-$	$5.0 \times 10^{10} \times 2.2 \times 10^{-12} \times e^{-3730 \times (\frac{1}{T_{liq}} - \frac{1}{298})} s^{-1}$	$5.0 \times 10^{10} M^{-1} s^{-1}$	[2] [3]
3	$2HNO_2 \rightleftharpoons NO + NO_2 (+ H_2O)$	$13.4 M^{-1} s^{-1}$	$1.1 \times 10^9 M^{-1} s^{-1}$	[4]
4	$HNO_2 \rightleftharpoons H^+ + NO_2^-$	$9.73 \times 10^9 \times e^{-\frac{1760}{T_{liq}}} s^{-1}$	$5 \times 10^{10} M^{-1} s^{-1}$	[1]
5	$HNO_3 \rightleftharpoons H^+ + NO_3^-$	$2.6 \times 10^9 \times e^{-\frac{1800}{T_{liq}}} s^{-1}$	$5 \times 10^{10} M^{-1} s^{-1}$	[1]
6	$HO_2 + NO_2 \rightleftharpoons O_2NOOH$	$1.8 \times 10^9 M^{-1} s^{-1}$	$5.27 \times 10^{16} \times e^{-\frac{13200}{T_{liq}}} s^{-1}$	[4] [5]
7	$HO_2 \rightleftharpoons H^+ + O_2^-$	$5.0 \times 10^{10} \times 10^{-4.57} s^{-1}$	$5.0 \times 10^{10} M^{-1} s^{-1}$	[3]
8	$HO_3 \rightleftharpoons H^+ + O_3^-$	$1.4 \times 10^5 s^{-1}$	$5 \times 10^{10} M^{-1} s^{-1}$	[4]
9	$NO + NO_2 \rightleftharpoons N_2O_3$	$1.1 \times 10^9 M^{-1} s^{-1}$	$8.4 \times 10^4 s^{-1}$	[4]
10	$2NO_2 \rightleftharpoons N_2O_4$	$4.5 \times 10^8 M^{-1} s^{-1}$	$6.9 \times 10^3 s^{-1}$	[4]

11	$2NO_2 (+H_2O) \rightleftharpoons 2H^+ + NO_2^- + NO_3^-$	$1.5 \times 10^{10} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	$41 M^{-3}s^{-1}$	[2] [4]
12	$NO_2 + O_2^- \rightleftharpoons O_2NOO^-$	$4.5 \times 10^9 M^{-1}s^{-1}$	$1.05 s^{-1}$	[4]
13	$O + O_2 \rightleftharpoons O_3$	$4.0 \times 10^9 M^{-1}s^{-1}$	$3.0 \times 10^{-6} s^{-1}$	[4]
14	$O^- + O_2 \rightleftharpoons O_3^-$	$3.6 \times 10^9 M^{-1}s^{-1}$	$3.3 \times 10^3 s^{-1}$	[4]
15	$O_2NOOH \rightleftharpoons H^+ + O_2NOO^-$	$5 \times 10^5 s^{-1}$	$5 \times 10^5 \times 10^{5.9} M^{-1}s^{-1}$	[1] ¹
16	$OH + OH^- \rightleftharpoons H_2O + O^-$	$1.3 \times 10^{10} M^{-1}s^{-1}$	$1.7 \times 10^6 M^{-1}s^{-1}$	[4]
17	$OH \rightleftharpoons H^+ + O^-$	$1.0 \times 10^{11} \times 10^{-11.9} s^{-1}$	$1.0 \times 10^{11} M^{-1}s^{-1}$	[3]
18	$ONOO^- \rightleftharpoons NO + O_2^-$	$0.02 s^{-1}$	$5 \times 10^9 M^{-1}s^{-1}$	[4]
19	$ONOO^- \rightleftharpoons NO_2 + O^-$	$1 \times 10^{-6} s^{-1}$	$3.5 \times 10^9 M^{-1}s^{-1}$	[4]
20	$ONOOH \rightleftharpoons H^+ + ONOO^-$	$5 \times 10^5 s^{-1}$	$5 \times 10^5 \times 10^{6.6} M^{-1}s^{-1}$	² 1
21	$ONOOH \rightleftharpoons NO_2 + OH$	$0.35 s^{-1}$	$4.5 \times 10^9 M^{-1}s^{-1}$	[4]

¹Reverse rate coefficient calculated from forward reaction using pKa from [4]

²Forward rate coefficient estimated to be the same as that of Reaction 15

Irreversible reactions

No.	Reaction	Rate coefficient	Source
22	$2H \rightarrow H_2$	$7.8 \times 10^9 M^{-1}s^{-1}$	[4]
23	$H + H_2O \rightarrow H_2 + OH$	$11 M^{-1}s^{-1}$	[4]
24	$H + H_2O_2 \rightarrow H_2O + OH$	$9.0 \times 10^7 M^{-1}s^{-1}$	[4]
25	$H + HNO_2 \rightarrow H_2O + NO$	$4.5 \times 10^8 M^{-1}s^{-1}$	[6]
26	$H + HO_2 \rightarrow H_2O_2$	$1.8 \times 10^{10} M^{-1}s^{-1}$	[4]
27	$H + HO_2^- \rightarrow OH + OH^-$	$9.0 \times 10^7 M^{-1}s^{-1}$	[4]
28	$H + N_2O \rightarrow N_2 + OH$	$2.1 \times 10^6 M^{-1}s^{-1}$	[4]
29	$H + NO_2 \rightarrow HNO_2$	$1.0 \times 10^{10} M^{-1}s^{-1}$	[4]
30	$H + NO_2^- \rightarrow NO + OH^-$	$7.1 \times 10^8 M^{-1}s^{-1}$	[4]
31	$H + O^- \rightarrow OH^-$	$1.1 \times 10^{10} M^{-1}s^{-1}$	[4]
32	$H + O_2 \rightarrow HO_2$	$2.1 \times 10^{10} M^{-1}s^{-1}$	[4]
33	$H + O_2^- \rightarrow HO_2^-$	$1.8 \times 10^{10} M^{-1}s^{-1}$	[4]
34	$H + O_3 \rightarrow HO_3$	$3.8 \times 10^{10} M^{-1}s^{-1}$	[4]
35	$H + O_3^- \rightarrow O_2 + OH^-$	$1.0 \times 10^{10} M^{-1}s^{-1}$	[4]
36	$H + OH \rightarrow H_2O$	$7.0 \times 10^9 M^{-1}s^{-1}$	[4]
37	$H^+ + H_2O_2 + NO_2^- \rightarrow H_2O + ONOOH$	$1.1 \times 10^3 M^{-2}s^{-1}$	[7]
38	$H^+ + O_3^- \rightarrow O_2 + OH$	$9.0 \times 10^{10} M^{-1}s^{-1}$	[4]
39	$H^+ + ONOOH \rightarrow 2H^+ + NO_3^-$	$4.3 M^{-1}s^{-1}$	[4]
40	$H_2 + H_2O_2 \rightarrow H + H_2O + OH$	$6 \times 10^6 M^{-1}s^{-1}$	[6]
41	$H_2 + O^- \rightarrow H + OH^-$	$8.0 \times 10^7 M^{-1}s^{-1}$	[4]
42	$H_2 + OH \rightarrow H + H_2O$	$4 \times 10^7 M^{-1}s^{-1}$	[6]
43	$HO_2 + O_2^- (+H_2O) \rightarrow H_2O_2 + O_2 + OH^-$	$1.5 \times 10^{10} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
44	$H_2O + N_2O_3 \rightarrow 2HNO_2$	$1.16 \times 10^4 M^{-1}s^{-1}$	[6]
45	$H_2O + N_2O_4 \rightarrow HNO_2 + HNO_3$	$8.00 \times 10^2 M^{-1}s^{-1}$	[6]
46	$H_2O + N_2O_5 \rightarrow H_2O + NO_2 + NO_3$	$84 M^{-1}s^{-1}$	[6]
47	$H_2O + N_2O_5 \rightarrow 2HNO_3$	$1 M^{-1}s^{-1}$	[6]
48	$H_2O + N_2O_5 \rightarrow 2ONOOH$	$1 M^{-1}s^{-1}$	[6]
49	$H_2O + NO_2^- + O^- \rightarrow NO_2 + 2OH^-$	$1.10 \times 10^{-17} M^{-2}s^{-1}$	[6]
50	$H_2O + NO_3 \rightarrow HNO_3 + OH$	$2.9 \times 10^7 M^{-1}s^{-1}$	[6]

51	$H_2O_2 + HNO_2 \rightarrow H_2O + ONOOH$	$1.4 \times 10^2 \times [H^+] M^{-1}s^{-1}$	[4]
52	$H_2O_2 + HNO_2 \rightarrow 2H^+ + H_2O + NO_3^-$	$3.7 \times 10^{13} \times [H^+] \times e^{-\frac{6700}{T_{liq}}} M^{-1}s^{-1}$	[2]
53	$H_2O_2 + HO_2 \rightarrow H_2O + O_2 + OH$	$0.5 M^{-1}s^{-1}$	[4]
54	$H_2O_2 + NO_3 \rightarrow H^+ + HO_2 + NO_3^-$	$4.0 \times 10^9 \times e^{-\frac{2000}{T_{liq}}} M^{-1}s^{-1}$	[1]
55	$H_2O_2 + O \rightarrow HO_2 + OH$	$1.6 \times 10^9 M^{-1}s^{-1}$	[4]
56	$H_2O_2 + O^- \rightarrow H_2O + O_2^-$	$5.0 \times 10^7 M^{-1}s^{-1}$	[4]
57	$H_2O_2 + O_2^- \rightarrow O_2 + OH + OH^-$	$0.13 M^{-1}s^{-1}$	[4]
58	$H_2O_2 + O_3 \rightarrow HO_2 + O_2 + OH$	$6.5 \times 10^{-3} M^{-1}s^{-1}$	[4]
59	$H_2O_2 + O_3 \rightarrow H_2O + 2O_2$	$7.8 \times 10^{-3} \times [O_3]^{-\frac{1}{2}} M^{-1}s^{-1}$	[2]
60	$H_2O_2 + OH \rightarrow H_2O + HO_2$	$8.4 \times 10^9 \times e^{-\frac{1680}{T_{liq}}} M^{-1}s^{-1}$	[1]
61	$HNO + O_2 \rightarrow HO_2 + NO$	$5.00 M^{-1}s^{-1}$	[6]
62	$HNO + O_3 \rightarrow HNO_2 + O_2$	$5.79 \times 10^6 M^{-1}s^{-1}$	[6]
63	$HNO + OH \rightarrow H_2O + NO$	$4.82 \times 10^{10} M^{-1}s^{-1}$	[6]
64	$HNO_2 + O_2NOOH \rightarrow 2H^+ + 2NO_3^-$	$12 M^{-1}s^{-1}$	[4]
65	$HNO_2 + O_2NOOH \rightarrow 2HNO_3$	$12.0 M^{-1}s^{-1}$	[6]
66	$HNO_2 + OH \rightarrow H_2O + NO_2$	$1.5 \times 10^{11} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
67	$HNO_3 + OH \rightarrow H_2O + NO_3$	$5.3 \times 10^7 M^{-1}s^{-1}$	[4]
68	$HO_2 + HO_3 \rightarrow H_2O_2 + 1.5O_2$	$5 \times 10^9 M^{-1}s^{-1}$	[4]
69	$2HO_2 \rightarrow H_2O_2 + O_2$	$7.6 \times 10^9 \times e^{-\frac{2720}{T_{liq}}} M^{-1}s^{-1}$	[1]
70	$HO_2 + HO_2^- \rightarrow O_2 + OH + OH^-$	$0.5 M^{-1}s^{-1}$	[4]
71	$HO_2 + NO \rightarrow ONOOH$	$3.2 \times 10^9 M^{-1}s^{-1}$	[4]
72	$HO_2 + NO \rightarrow HNO_3$	$3.21 \times 10^9 M^{-1}s^{-1}$	[6]
73	$HO_2 + NO_3 \rightarrow H^+ + NO_3^- + O_2$	$6.9 \times 10^{11} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
74	$HO_2 + O^- \rightarrow O_2 + OH^-$	$6.0 \times 10^9 M^{-1}s^{-1}$	[4]
75	$HO_2 + O_2^- \rightarrow HO_2^- + O_2$	$8.0 \times 10^7 M^{-1}s^{-1}$	[4]
76	$HO_2 + O_3 \rightarrow HO_3 + O_2$	$5.0 \times 10^8 M^{-1}s^{-1}$	[4]
77	$HO_2 + O_3 \rightarrow 2O_2 + OH$	$1.0 \times 10^4 M^{-1}s^{-1}$	[4]
78	$HO_2 + O_3^- \rightarrow 2O_2 + OH^-$	$6.0 \times 10^9 M^{-1}s^{-1}$	[4]
79	$HO_2 + OH \rightarrow H_2O + O_2$	$1.1 \times 10^{12} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
80	$HO_2^- + O \rightarrow O_2^- + OH$	$5.3 \times 10^9 M^{-1}s^{-1}$	[4]
81	$HO_2^- + O^- \rightarrow O_2^- + OH$	$4.0 \times 10^8 M^{-1}s^{-1}$	[4]
82	$HO_2^- + O_2^- \rightarrow O^- + O_2 + OH$	$0.13 M^{-1}s^{-1}$	[4]
83	$HO_2^- + O_3 \rightarrow O_2 + O_2^- + OH$	$5.5 \times 10^6 M^{-1}s^{-1}$	[4]
84	$HO_2^- + OH \rightarrow HO_2 + OH^-$	$7.5 \times 10^9 M^{-1}s^{-1}$	[4]
85	$2HO_3 \rightarrow H_2O_2 + 2O_2$	$5 \times 10^9 M^{-1}s^{-1}$	[4]
86	$HO_3 + O_2^- \rightarrow 2O_2 + OH^-$	$1 \times 10^{10} M^{-1}s^{-1}$	[4]
87	$HO_3 + OH \rightarrow H_2O_2 + O_2$	$5 \times 10^9 M^{-1}s^{-1}$	[4]
88	$HO_3 \rightarrow O_2 + OH$	$1.1 \times 10^5 s^{-1}$	[4]
89	$2N \rightarrow N_2$	$3 \times 10^7 M^{-1}s^{-1}$	[6]
90	$N_2O + NO_2^- \rightarrow N_2 + NO_3^-$	$3 \times 10^8 M^{-1}s^{-1}$	[6]
91	$N_2O_3 + ONOO^- \rightarrow 2NO_2 + NO_2^-$	$3 \times 10^8 M^{-1}s^{-1}$	[4]
92	$N_2O_3 (+ OH^-) \rightarrow H^+ + 2NO_2^-$	$2 \times 10^3 + 1 \times 10^8 \times [OH^-] s^{-1}$	[4]
93	$N_2O_4 (+ H_2O) \rightarrow 2H^+ + NO_2^- + NO_3^-$	$1 \times 10^3 s^{-1}$	[4]
94	$N_2O_5 (+ H_2O) \rightarrow 2H^+ + 2NO_3^-$	$5 \times 10^9 s^{-1}$	[4]
95	$2NO + O_2 \rightarrow 2NO_2$	$2.3 \times 10^6 M^{-2}s^{-1}$	[4]

96	$NO + NO_2 (+ H_2O) \rightarrow 2H^+ + 2NO_2^-$	$3.1 \times 10^{10} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
97	$NO + O_2^- \rightarrow NO_3^-$	$4 \times 10^9 M^{-1}s^{-1}$	[6]
98	$NO + OH \rightarrow H^+ + NO_2^-$	$3.1 \times 10^{12} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
99	$NO + OH \rightarrow HNO_2$	$2.0 \times 10^{10} M^{-1}s^{-1}$	[6]
100	$2NO_2 (+ H_2O) \rightarrow H^+ + HNO_2 + NO_3^-$	$5.0 \times 10^3 \times e^{-\frac{2900}{T_{liq}}} M^{-1}s^{-1}$	[1]
101	$NO_2 + NO_3 \rightarrow N_2O_5$	$1.7 \times 10^9 M^{-1}s^{-1}$	[4]
102	$NO_2 + O_2^- \rightarrow NO_2^- + O_2$	$1 \times 10^9 M^{-1}s^{-1}$	[4]
103	$NO_2 + OH \rightarrow H^+ + NO_3^-$	$2.0 \times 10^{11} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
104	$NO_2 + OH \rightarrow HNO_3$	$1 \times 10^{10} M^{-1}s^{-1}$	[6]
105	$NO_2^- + NO_3 \rightarrow NO_2 + NO_3^-$	$1.8 \times 10^{11} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
106	$NO_2^- + O_3 \rightarrow NO_3^- + O_2$	$8 \times 10^{15} \times e^{-\frac{7000}{T_{liq}}} M^{-1}s^{-1}$	[1]
107	$NO_2^- + OH \rightarrow NO_2 + OH^-$	$1.5 \times 10^{12} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
108	$NO_3 + O_2^- \rightarrow NO_3^- + O_2$	$1.5 \times 10^{11} \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
109	$NO_3 + OH^- \rightarrow NO_3^- + OH$	$8.1 \times 10^{11} \times e^{-\frac{2700}{T_{liq}}} M^{-1}s^{-1}$	[1]
110	$2O \rightarrow O_2$	$2.8 \times 10^{10} M^{-1}s^{-1}$	[4]
111	$O + OH^- \rightarrow HO_2^-$	$4.2 \times 10^8 M^{-1}s^{-1}$	[4]
112	$O (+ H_2O) \rightarrow 2OH$	$50 s^{-1}$	[4]
113	$2O^- (+ H_2O) \rightarrow HO_2^- + OH^-$	$1.0 \times 10^9 M^{-1}s^{-1}$	[4]
114	$O^- + O_2^- (+ H_2O) \rightarrow O_2 + 2OH^-$	$6.0 \times 10^8 M^{-1}s^{-1}$	[4]
115	$O^- + O_3 \rightarrow O_2 + O_2^-$	$5.0 \times 10^9 M^{-1}s^{-1}$	[4]
116	$O^- + O_3^- \rightarrow 2O_2^-$	$7.0 \times 10^8 M^{-1}s^{-1}$	[4]
117	$O^- + OH \rightarrow HO_2^-$	$2.6 \times 10^{10} M^{-1}s^{-1}$	[4]
118	$2O_2^- (+ 2H_2O) \rightarrow H_2O_2 + O_2 + 2OH^-$	$0.3 M^{-1}s^{-1}$	[4]
119	$O_2^- + O_3 \rightarrow O_2 + O_3^-$	$1.6 \times 10^9 M^{-1}s^{-1}$	[4]
120	$O_2^- + O_3 (+ H_2O) \rightarrow 2O_2 + OH + OH^-$	$7.6 \times 10^9 \times e^{-\frac{1500}{T_{liq}}} M^{-1}s^{-1}$	[2]
121	$O_2^- + OH \rightarrow O_2 + OH^-$	$1.4 \times 10^{13} \times e^{-\frac{2120}{T_{liq}}} M^{-1}s^{-1}$	[1]
122	$O_2(\alpha^1\Delta) + OH \rightarrow O_2 + OH$	$2.2 \times 10^3 M^{-1}s^{-1}$	[4]
123	$O_2(\alpha^1\Delta) \rightarrow O_2$	$2.5 \times 10^5 s^{-1}$	[4]
124	$O_2NOO^- \rightarrow NO_2^- + O_2$	$3 \times 10^{18} \times e^{-\frac{12700}{T_{liq}}} s^{-1}$	[5]
125	$O_2NOOH \rightarrow HNO_2 + O_2$	$7 \times 10^{-4} s^{-1}$	[6]
126	$O_3 + OH \rightarrow HO_2 + O_2$	$3 \times 10^9 M^{-1}s^{-1}$	[4]
127	$O_3 + OH^- \rightarrow HO_2^- + O_2$	$40 M^{-1}s^{-1}$	[4]
128	$O_3 + OH^- \rightarrow HO_2 + O_2^-$	$70 M^{-1}s^{-1}$	[4]
129	$O_3^- + OH \rightarrow HO_2 + O_2^-$	$6 \times 10^9 M^{-1}s^{-1}$	[4]
130	$O_3^- + OH \rightarrow O_3 + OH^-$	$2.5 \times 10^9 M^{-1}s^{-1}$	[4]
131	$O_3^- (+ H_2O) \rightarrow O_2 + OH + OH^-$	$25 s^{-1}$	[4]
132	$OH + N_2O \rightarrow HNO + NO$	$2.3 \times 10^4 M^{-1}s^{-1}$	[6]
133	$2OH \rightarrow H_2O_2$	$5.0 \times 10^9 M^{-1}s^{-1}$	[4]
134	$OH + ONOO^- \rightarrow H^+ + NO_2^- + O_2^-$	$4 \times 10^9 M^{-1}s^{-1}$	[4]
135	$OH + ONOO^- \rightarrow NO + O_2 + O_2^-$	$4.8 \times 10^9 M^{-1}s^{-1}$	[4]
136	$ONOO^- + ONOOH \rightarrow NO_2^- + O_2NOOH$	$1.3 \times 10^3 M^{-1}s^{-1}$	[4]
137	$ONOO^- \rightarrow NO_3^-$	$8 \times 10^{-6} s^{-1}$	[4]

138	$ONOOH \rightarrow H^+ + NO_3^-$	$7.3 \times 10^{15} \times e^{-\frac{10800}{T_{liq}}} s^{-1}$	[5]
139	$ONOOH \rightarrow HNO_3$	$8.5 \times 10^9 \times e^{-\frac{7500}{T_{liq}}} s^{-1}$	[8]

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