Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information

Two reaction regimes in the oxidation of larger cationic tantalum clusters (Ta_n^+, n = 13 - 40) under multi-collision conditions

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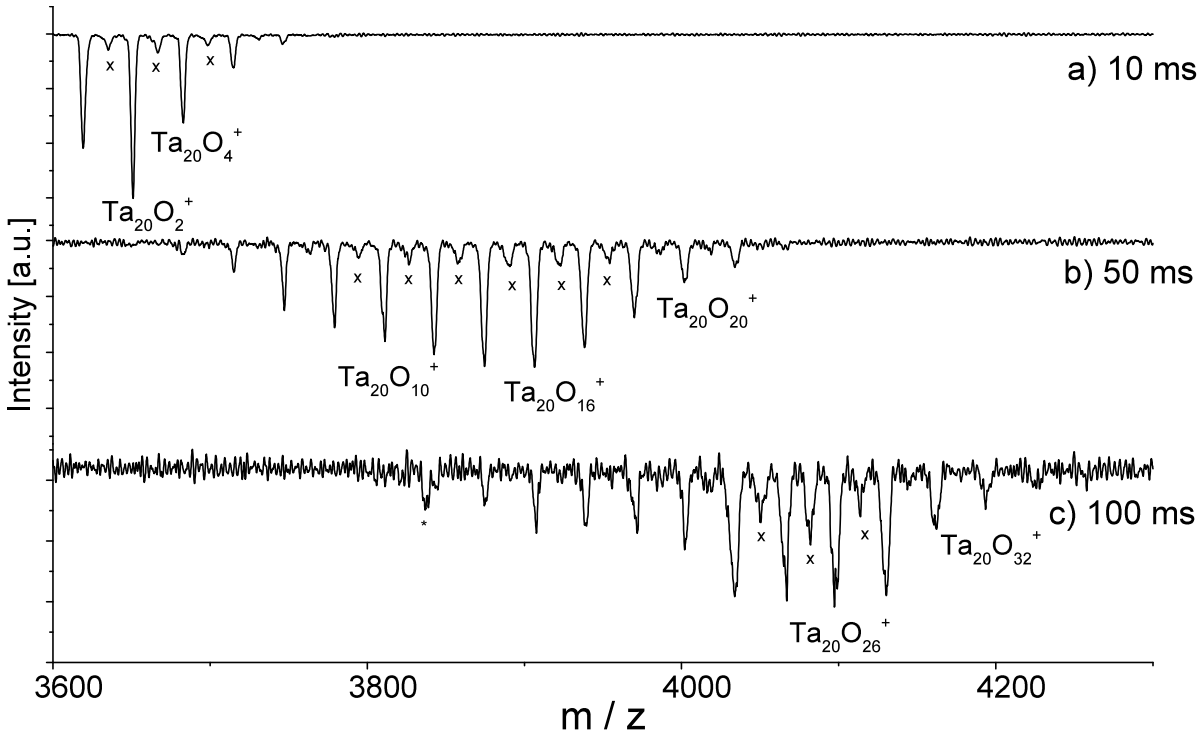

Fig. 1 Mass spectra of Ta_{20}^+ clusters exposed to 100 ppm O_2 at 50 K for reaction times of a) 10 ms, b) 50 ms, c) 100 ms. The reaction proceeds via subsequent attachment of O_2 units. In contrast to the reaction at 300 K, no final reaction product is observed. Instead, additional oxygen molecules are loosely bound to the cluster. Peaks marked with x are due to a side reaction with water background, peaks denoted by an * are due fragmentation of the cluster.

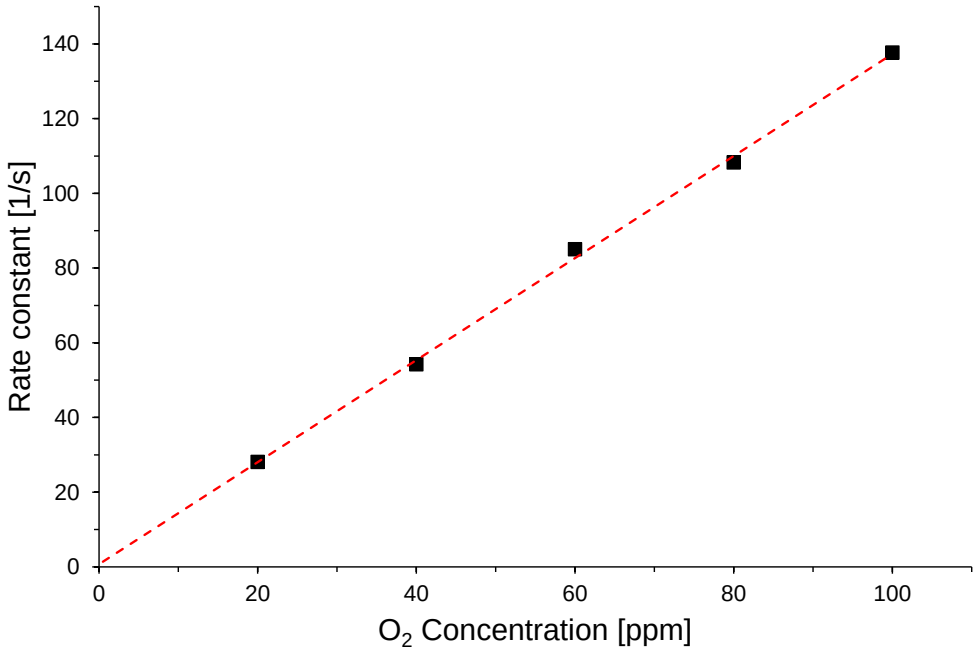

Fig. 2 Rate constant k_1 for the first reaction step as a function of the oxygen concentration within the helium buffer gas. The dashed red line represents a linear regression.

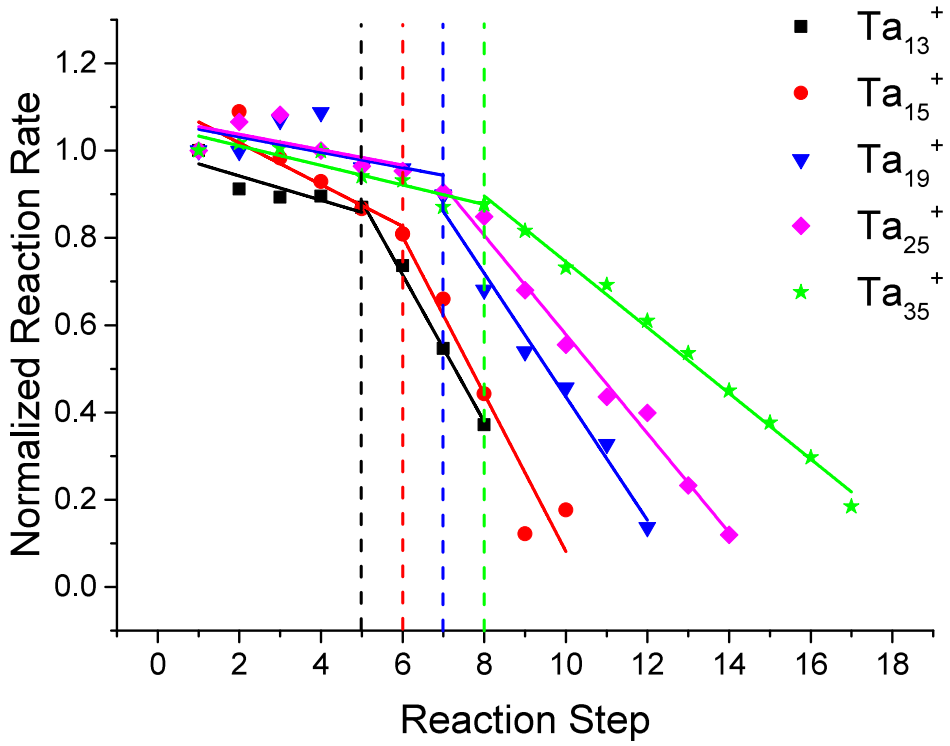

Fig. 3 Rate constants for the oxidation of Ta_{13}^+, Ta_{15}^+, Ta_{25}^+ and Ta_{35}^+. All rate constants are normalized to the first reaction step. For all cluster sizes two reaction regimes are observed. The transition reaction step in general increases with cluster size. Solid lines represent a linear regression of the rate constants for both reaction regimes.

Table 1 Rate constants for the various reaction steps in the consecutive oxidation reaction of Ta^+_{13-40} at 300 K and with 100 ppm oxygen.

	Ta^+_{13}	Ta^+_{14}	Ta^+_{15}	Ta^+_{16}	Ta^+_{17}	Ta^+_{18}	Ta^+_{19}	Ta^+_{20}	Ta^+_{21}	Ta^+_{25}	Ta^+_{30}	Ta^+_{35}	Ta^+_{40}
k_1 [$10^{-10}cm^3/s$]	19	21	13	24	20	23	14	18	15	15	18	21	22
k_2 [$10^{-10}cm^3/s$]	17	19	14	20	18	21	15	16	16	17	19	22	22
k_3 [$10^{-10}cm^3/s$]	17	18	13	20	20	20	16	16	14	17	20	21	21
k_4 [$10^{-10}cm^3/s$]	17	18	13	20	20	19	16	16	13	15	19	21	21
k_5 [$10^{-10}cm^3/s$]	17	17	12	20	16	18	13	14	13	15	18	20	21
k_6 [$10^{-10}cm^3/s$]	14	17	11	17	14	15	5	14	13	15	17	20	20
k_7 [$10^{-10}cm^3/s$]	10	12	9	12	14	14	13	15	11	14	16	19	21
k_8 [$10^{-10}cm^3/s$]	7	9	6	10	15	12	10	12	9	13	16	19	19
k_9 [$10^{-10}cm^3/s$]			2	6	9	13	7	10	8	11	14	17	18
k_{10}[$10^{-10}cm^3/s$]			2	2	6	12	7	8	7	9	13	16	17
k_{11} [$10^{-10}cm^3/s$]					1	3	5	6	5	7	11	15	17
k_{12} [$10^{-10}cm^3/s$]							2	3	2	6	9	13	15
k_{13}[$10^{-10}cm^3/s$]								2	1	3	7	11	14
k_{14} [$10^{-10}cm^3/s$]										2	6	10	13
k_{15} [$10^{-10}cm^3/s$]											3	8	11
k_{16} [$10^{-10}cm^3/s$]												6	9
k_{17} [$10^{-10}cm^3/s$]												4	8
k_{18} [$10^{-10}cm^3/s$]												2	5
k_{19} [$10^{-10}cm^3/s$]													4
k_{20} [$10^{-10}cm^3/s$]													3
k_{21} [$10^{-10}cm^3/s$]													2