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SUPPORTING INFORMATION

COMPUTATIONAL METHODS

The exploration of the potential energy surface (PES) was performed using the AUTOMATON program, which applies the cellular automaton criterion to generate the initial population followed by genetic algorithm operations over several iterations until the lowest energy structure (global minimum) is found. Specific technical details can be found in the original publication.

Geometrical optimizations and search procedures were initially carried out at the M06-2X/def2SVP level and subsequently refined at the M06-2X, MP2, and CCSD(T) levels for OHe_4^{2+} and at the M06-2X, and MP2 levels for ONe_4^{2+} using the aug-cc-pVTZ basis set. Due to the high computational cost, ONe_4^{2+} could not be optimized at the CCSD(T) level.

In all cases, vibrational analyses were carried out to verify that the obtained structures correspond to true minima on their respective potential energy surfaces. Default options SCF=tight (Accuracy of 10^{-12}), integral=ultrafine (99590 grid points) were employed.

Wavefunction analyses were performed at the CCSD level, since Gaussian16 does not provide wavefunctions at the CCSD(T) level. Accordingly, single-point CCSD calculations were conducted using the CCSD(T) geometry for OHe_4^{2+} and the MP2 geometry for ONe_4^{2+} using the density=current option in Gaussian16. Wiberg bond indices and natural population analyses (NPA) were obtained with NBO 7.0, while QTAIM analyses were performed with Multiwfn. Full reference details are provided in the original manuscript.

Table S1. ZPE-corrected total electronic energies (in a.u.) for ONg_4^{2+} singlet D_{4h} and triplet C_{2v} at different levels using the aug-cc-pVTZ basis set.

ONg_4^{2+}	CCSD(T)		MP2		M06-2X	
	Singlet	Triplet	Singlet	Triplet	Singlet	Triplet
OHe_4^{2+}	-84.994363	-85.013824	-84.927452	-84.952538	-85.115409	-85.139084
ONe_4^{2+}	---	---	-588.670641	-588.688541	-589.284605	-589.32118

Table S2. Cartesian Coordinates (in Å) at the MP2/aug-cc-pVTZ level for ONg_4^{2+} at the triplet state

OHe_4^{2+}				**ONe_4^{2+}**			
M06-2X							
8	0.000000000	0.000000000	0.309805000	8	0.000000000	0.000000000	0.532059000
2	0.000000000	1.430955000	0.321916000	10	0.000000000	1.713946000	0.629860000
2	1.278630000	0.000000000	-0.941526000	10	1.449768000	0.000000000	-0.842683000
2	-1.278630000	0.000000000	-0.941526000	10	-1.449768000	0.000000000	-0.842683000
2	0.000000000	-1.430955000	0.321916000	10	0.000000000	-1.713946000	0.629860000
MP2							
8	0.000000000	0.000000000	0.330249000	8	0.000000000	0.000000000	0.536593000
2	0.000000000	1.413655000	0.316122000	10	0.000000000	1.625310000	0.623981000
2	1.311609000	0.000000000	-0.976620000	10	1.481371000	0.000000000	-0.838618000
2	-1.311609000	0.000000000	-0.976620000	10	-1.481371000	0.000000000	-0.838618000
2	0.000000000	-1.413655000	0.316122000	10	0.000000000	-1.625310000	0.623981000
CCSD(T)							
8	0.000000000	0.000000000	-0.324918754				
2	-1.433164053	0.000000000	-0.322675929				
2	1.433164053	0.000000000	-0.322675929				
2	0.000000000	-1.314772238	0.971884722				
2	0.000000000	1.314772238	0.971884722				

Table S3. Gibbs free energies of dissociation (ΔG_{diss}) at 298K for the possible dissociation pathways of ONg_4^{2+} (Ng = He, Ne) computed at the MP2/aug-cc-pVTZ level.

Dissociation pathway		**ΔG_{diss} (kcal·mol^{-1})**	
		OHe_4^{2+}	**ONe_4^{2+}**
1	$ONg^{2+}_4 \rightarrow ONg^{2+}_3 + Ng$	−0.6	0.9
2	$ONg^{2+}_4 \rightarrow ONg^{2+}_2 + 2Ng$	9.8	7.6
3	$ONg^{2+}_4 \rightarrow ONg^{+}_2 + Ng^{+}_2$	−105.7	−97.8
4	$ONg^{2+}_4 \rightarrow Ng^{+}_2 + O^{+} + 2Ng$	−72.4	−58.3
5	$ONg^{2+}_4 \rightarrow ONg^{2+} + 3Ng$	12.3	−123.7
6	$ONg^{2+}_4 \rightarrow O^{2+} + 4Ng$	101.7	157.2