Supporting Information

Guanosine Dianions Hydrated by One to Four Water Molecules

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1. Methodology

1.1 Experimental methods

The NIPE spectra were recorded using a home-made cryogenic magnetic-bottle time-of-flight (TOF) photoelectron spectrometer, coupled with an electrospray ionization (ESI) source and a temperature-variable cryogenic ion trap. The [dGMP-2H]$_2^-$ dianion and its solvated clusters were produced by spraying 0.1 mM solutions of the corresponding sodium salt in H$_2$O/CH$_3$CN (1:3) under a humidity-controlled environment. The clusters were guided by an RF quadrupole ion guide and detected by a quadrupole mass spectrometer to optimize ESI conditions to ensure stable and intense solvated cluster beams. Then, the anions were directed into the cryogenic 3D ion trap, where they were accumulated and cooled by collisions with a cold buffer gas (20% H$_2$ balanced in helium). In the present work, the temperature inside the ion trap was set at 20 K. The cryogenic anions were then pulsed out into the extraction zone of a TOF mass spectrometer at a 10 Hz repetition rate. Each of the desired clusters was mass-selected and maximally decelerated before being photodetached in the interaction zone with 157 nm (7.867 eV) light from an F$_2$ excimer laser. The laser was operated at a 20 Hz repetition rate with the ion beam off at alternating laser shots for shot-by-shot background subtraction. The distribution of the detached electrons was analyzed using a 5.2 m-long magnetic-bottle electron flight tube, and then converted into kinetic energy spectrum after calibration with the known data of I$^-$ and OsCl$_6^{2-}$. The electron binding energy (EBE) spectra were obtained by subtracting the kinetic energy spectra from the photon energy used. The electron energy resolution ($\Delta E/E$) was about 2%, i.e., $\sim$20 meV for 1 eV kinetic energy electrons.

1.2 Computational methods

To interpret the obtained PES spectra of the [dGMP-2H]$_2^-$•nH$_2$O anions, quantum chemistry calculations preceded by molecular dynamics based conformational search were conducted for all solvated systems. The initial [dGMP-2H]$_2^-$ geometry was taken from our previous study. In order to identify the most important structures for the geometries generated in the MD simulation, a clustering against each of four microsolvated groups was used. Then the contributions of particular structures were computed based on the differences in their Gibbs free energies calculated quantum-chemically. Finally, the VDE and ADE values for the most represented anionic geometries were calculated and compared to the experimental data.

1.2.1 Molecular dynamics conformational search

The geometries of complexes for QM calculations were obtained from clustering the MD simulations performed with Gromacs18. The nucleoside dianion starting structure was taken from our previous study and parametrized with parmbsc1 forcefield (DG3) and phosaa10
modifications (for the additional hydrogen atom) with charges calculated explicitly with Gaussian16, B3LYP/6-31++G(d,p). One to four waters were placed around the nucleoside dianion randomly and the complex put into a geometric center of cubic box with side length 40 Å. Three replicas with different starting points (water orientations) were prepared. After a short steepest descent minimization (up to 5000 steps) 1 microsecond NVT simulations were performed with a timestep of 2 fs for each replica in 200 and 300 K resulting in 6 simulations for every n=1 to 4 waters of the [dGMP-2H]2−•nH2O systems. Note that the targeted hydrated clusters were generated from ESI solutions at ambient conditions and the rapid solvent evaporation and cryogenic cooling afford to freeze and retain conformational spaces accessible at near room temperatures. A Verlet cut-off and a twin range cut-off of 18 Å was used along with PBC conditions. LINear Constraint Solver (LINCS) was used to constraint hydrogen bonds and Berendsen thermostat. The center of mass translation and rotation around the center of mass were removed. Next, we clustered the last 900 ns of each of the generated MD trajectories using the gromos algorithm implemented in Gromacs. For that root mean square deviation (RMSD) was computed based on all atoms and the cut-offs of 0.1, 0.15, 0.2 and 0.25 Å were applied for 1, 2, 3 and 4 waters, respectively. The representative structures of each cluster were further transferred to QM calculations.

1.2.2 Quantum chemistry methodology adjustment

In order to choose an appropriate DFT level, four XC functionals were tested on the set of ten manually prepared singly solvated [dGMP-2H]2−•H2O dianions (manual-1 to manual-10). We compared the experimental PES spectrum with the VDE and ADE values calculated for those ten systems using the following four methods: i) B3LYP and ii) CAM-B3LYP both combined with the 6-31++G(d,p) basis set. These two methods were used in our previous work on non-solvated [dGMP-2H]2−. Additionally iii) the ωB97XD functional combined with the aug-cc-pvdz basis set, suggested in Wang et al. benchmark studies on multiply charged anions as performing similarly well as the CAM-B3LYP functional, was tested. Finally, we also considered iv) the HSE06 functional combined with the aug-cc-pvdz basis set, which was found to be the best method for reproducing the experimental values of VDE in anionic silicon clusters. The best agreement between experimental and calculated VDE values for the [dGMP-2H]2−•H2O dianions was observed for the CAM-B3LYP/6-31++G(d,p) method, which only slightly underestimates the experimental VDE (1.63 eV vs. 1.8 eV found experimentally). VDEs obtained for the remaining functional were calculated at 1.52, 1.49 and 1.42 eV for B3LYP, ωB97XD and HSE06, respectively. Thus, CAM-B3LYP/6-31++G(d,p) (gas phase) level of theory was chosen as the most reliable one for the current study.
1.2.3 Contribution of particular geometries to the equilibrated mixture of conformers

Molecular dynamics based conformational search led to quite large number of initial geometries for each microsolvated system: 35 geometries (plus set of 10 geometries prepared manually during methodology adjustment part) for [dGMP-2H]^{2−}•H$_2$O, 36 geometries for [dGMP-2H]^{2−}•2H$_2$O, 31 geometries for [dGMP-2H]^{2−}•3H$_2$O and 43 geometries for the [dGMP-2H]^{2−}•4H$_2$O anion. VDEs calculated for the singly hydrated [dGMP-2H]^{2−}•H$_2$O system span a wide range from 1.23 to 1.74 eV. Since PES spectrum usually represents the VDE energies of the most stable anionic conformers, we decided to calculate the measured characteristics for only those geometries that are the most stable thermodynamically. In a thermodynamically equilibrated system one can define the equilibrium constant $K_M$ as:

$$K_M = \frac{[M]}{[\text{Ref}]} = e^{-\Delta G/RT}$$

where $\Delta G$ is the difference in Gibbs free energy between the geometry of M (e.g. any conformation of dianionic [dGMP-2H]^{2−}•nH$_2$O) and the reference geometry Ref (the most stable geometry of dianionic [dGMP-2H]^{2−}•nH$_2$O). Obviously, $K_{\text{Ref}}$ is then equal to 1. The equilibrium fraction, $x_M$, of conformation M in the system containing n conformations can be then estimated as:

$$x_M = \frac{K_M}{(K_{\text{Ref}} + K_1 + K_2 + ... + K_n)}$$

Considering only the structures of $x_M \geq 0.01$ for further discussion the number of conformations is reduced to 13, 7, 9 and 11 for the system solvated with 1, 2, 3 and 4 water molecules, respectively. Those the most represented conformations were discussed in the main discussion, while for full data see Table S1-S4.

1.2.4 VDE and ADE definitions

Vertical detachment energy (VDE) of [dGMP-2H]^{2−}•nH$_2$O is defined as the difference in electronic energy of dianionic system and its corresponding anion radical, both in the optimized dianion geometry, while adiabatic detachment energy (ADE) is the difference in Gibbs free energy between dianionic and anion radical systems, both in their optimal geometries. To reduce the set of VDE/ADE values to a single number, we also calculated a weighted average VDE ($VDE_{avg}$) and ADE ($ADE_{avg}$), taking into account the contribution of each conformer to the equilibrated mixture of conformers (for the complete rather than reduced data sets):

$$VDE_{avg} = \sum_{M} VDE_M \cdot x_M$$
1.2.5 Hydration energies

For particular hydrates, hydration energies were calculated as the difference between the sum of the free energies ($\Delta G_{\text{hyd}}$) of non-interacting monomers and that of the cluster, all calculated at the optimal geometries:

$$\Delta G_{\text{hyd}} = G([\text{dGMP-2H}]^{2-} \cdot n\text{H}_2\text{O}) - n \cdot G(\text{H}_2\text{O}) - G([\text{dGMP-2H}]^{2-})$$

where $G(X)$ denote the free energy of species $X$ obtained by correcting its electronic energy for zero-point vibration, thermal contributions to energy from vibrations, rotations, and translations, entropy and the $pV$ term. These terms were determined in the harmonic oscillator-rigid rotor approximation for $T = 298$ K and $p = 1$ atm. The values of $\Delta G_{\text{hyd}}$ corrected for BSSE energy using the counterpoise procedure of Boys and Bernardi\textsuperscript{27} are denoted $\Delta G_{\text{hyd-BSSE}}$:

$$\Delta G_{\text{hyd-BSSE}} = \Delta G_{\text{hyd}} + \text{BSSE energy}$$
2. Relative stability, VDE, ADE and water binding energy for all the obtained [dGMP-2H]²⁻•nH₂O systems

Table S1. Relative Gibbs free energy values (ΔG, in kcal/mol), vertical and adiabatic detachment energy (VDE and ADE, in eV) and equilibrium fraction values (xₘ) calculated for the [dGMP-2H]²⁻•nH₂O systems. Data for the structures of xₘ≥0.01 as well as weighted average VDE and ADE bolded.

| Name              | ΔG  | VDE | ADE | xₘ      |
|-------------------|-----|-----|-----|---------|
| manually prepared structures |     |     |     |         |
| manual-1          | 7.2 | 1.62| 1.19| 3.48E-06|
| manual-2          | 12.3| 1.47| 1.02| 7.35E-10|
| manual-3          | 12.4| 1.40| 0.97| 5.98E-10|
| manual-4          |     |     |     |         |
| manual-5          | 11.2| 1.27| 0.91| 4.17E-09|
| manual-6          | 14.5| 1.28| 0.86| 1.60E-11|
| manual-7          | 12.4| 1.40| 0.97| 5.85E-10|
| manual-8          | 10.3| 1.23| 0.87| 1.91E-08|
| manual-9          | 2.5 | 1.63| 1.20| 1.08E-02|
| manual-10         | 5.1 | 1.62| 1.31| 1.26E-04|
| based on the 200K MD clustering |     |     |     |         |
| one-200-m1        | 7.7 | 1.70| 1.38| 1.68E-06|
| one-200-m2        | 9.4 | 1.64| 1.34| 8.74E-08|
| one-200-m3        | 10.2| 1.35| 1.10| 2.24E-08|
| one-200-m4        | 11.4| 1.63| 1.33| 3.23E-09|
| one-200-m5        | 12.6| 1.57| 1.30| 3.95E-10|
| one-200-m6        | 13.8| 1.33| 1.10| 5.39E-11|
| based on the 300K MD clustering |     |     |     |         |
| one-m1            | 2.2 | 1.59| 1.31| 1.69E-02|
| one-m2            | 1.6 | 1.64| 1.33| 4.87E-02|
| one-m3            | 4.0 | 1.27| 1.05| 7.71E-04|
| one-m4            | 3.5 | 1.36| 1.11| 1.82E-03|
| one-m5            | 2.6 | 1.65| 1.36| 8.53E-03|
| one-m6            | 5.2 | 1.58| 1.30| 1.19E-04|
| one-m7            | 2.0 | 1.65| 1.33| 2.45E-02|
| one-m8            | 4.8 | 1.50| 1.22| 2.08E-04|
| one-m9            | 2.0 | 1.36| 1.18| 2.55E-02|
| one-m10           | 4.2 | 1.29| 1.05| 5.66E-04|
| one-m11           | 3.7 | 1.26| 1.03| 1.30E-03|
| one-m12           | 2.6 | 1.59| 1.31| 8.77E-03|
| one-m13           | 4.6 | 1.63| 1.32| 2.85E-04|
| one-m14           | 1.9 | 1.36| 0.97| 3.11E-02|
| one-m15           | 3.2 | 1.61| 1.24| 3.23E-03|
| one-m16           | 3.8 | 1.30| 1.07| 1.12E-03|
| one-m17           | 1.6 | 1.67| 1.35| 4.93E-02|
| one-m18           | 2.6 | 1.65| 1.36| 8.55E-03|
| one-m19           | 4.8 | 1.34| 1.10| 2.16E-04|
|     |     |     |     |     |
|-----|-----|-----|-----|-----|
| one-m20 | 2.0 | 1.70 | 1.38 | 2.60E-02 |
| one-m21 | 3.8 | 1.29 | 1.06 | 1.13E-03 |
| one-m22 | 6.9 | 1.28 | 1.03 | 6.24E-06 |
| one-m23 | 0.0 | 1.70 | 1.37 | 7.14E-01 |
| one-m24 | 4.8 | 1.50 | 1.20 | 2.25E-04 |
| one-m25 | 5.5 | 1.59 | 1.29 | 6.94E-05 |
| one-m26 | 8.1 | 1.37 | 1.13 | 8.76E-07 |
| one-m27 | 6.4 | 1.30 | 1.07 | 1.36E-05 |
| one-m28 | 5.5 | 1.67 | 1.35 | 7.02E-05 |
| one-m29 | 2.3 | 1.33 | 0.97 | 1.58E-02 |

- weighted average: 1.66, 1.34
- experimental: 1.80, 1.55
**Table S2.** Relative Gibbs free energy values ($\Delta G$, in kcal/mol), vertical and adiabatic detachment energy (VDE and ADE, in eV) and equilibrium fraction values ($x_M$) calculated for the [dGMP-2H]$^2$•2H$_2$O systems. Data for the structures of $x_M \geq 0.01$ as well as weighted average VDE and ADE bolded.

| Name               | $\Delta G$ | VDE  | ADE  | $x_M$     |
|--------------------|------------|------|------|-----------|
| **based on the 200K MD clustering** |            |      |      |           |
| two-200-m1         | 7.4        | 2.09 | 1.70 | 1.71E-06  |
| two-200-m2         | 7.6        | 1.81 | 1.47 | 1.32E-06  |
| two-200-m3         |            |      |      |           |
| two-200-m4         | 9.9        | 2.09 | 1.58 | 2.54E-08  |
| two-200-m5         | 9.3        | 1.69 | 1.40 | 6.63E-08  |
| two-200-m6         | 7.3        | 2.03 | 1.65 | 1.92E-06  |
| two-200-m7         | 11.3       | 1.79 | 1.46 | 2.60E-09  |
| **based on the 300K MD clustering** |            |      |      |           |
| two-m1             | 2.7        | 1.69 | 1.40 | 5.18E-03  |
| two-m2             | 2.6        | 1.68 | 1.40 | 5.82E-03  |
| **two-m3**         | 1.4        | 2.00 | 1.62 | 4.37E-02  |
| two-m4             |            |      |      |           |
| two-m5             | 1.9        | 1.81 | 1.46 | 1.95E-02  |
| two-m6             | 2.0        | 1.74 | 1.42 | 1.48E-02  |
| two-m7             | 5.9        | 1.37 | 1.15 | 2.16E-05  |
| two-m8             | 5.4        | 1.68 | 1.40 | 5.49E-05  |
| two-m9             | 2.8        | 1.75 | 1.45 | 4.36E-03  |
| two-m10            |            |      |      |           |
| two-m11            | 6.0        | 1.80 | 1.50 | 1.72E-05  |
| two-m12            | 5.4        | 1.66 | 1.39 | 5.20E-05  |
| two-m13            | 4.6        | 1.36 | 1.13 | 1.90E-04  |
| two-m14            | 3.1        | 1.73 | 1.46 | 2.39E-03  |
| two-m15            | 4.3        | 2.08 | 1.63 | 3.22E-04  |
| two-m16            | 4.6        | 1.98 | 1.60 | 1.93E-04  |
| two-m17            | 2.5        | 1.65 | 1.37 | 6.26E-03  |
| two-m18            | 4.3        | 1.99 | 1.61 | 3.30E-04  |
| two-m19            | 0.0        | 1.81 | 1.32 | 4.62E-01  |
| two-m20            | 5.6        | 1.38 | 1.15 | 3.75E-05  |
| two-m21            | 5.0        | 1.78 | 1.44 | 9.39E-05  |
| **two-m22**        | 2.1        | 1.75 | 1.42 | 1.25E-02  |
| two-m23            | 0.1        | 1.81 | 1.53 | 3.93E-01  |
| two-m24            | 1.7        | 2.07 | 1.68 | 2.77E-02  |
| two-m25            | 5.5        | 1.73 | 1.43 | 4.65E-05  |
| two-m26            | 7.4        | 1.38 | 1.12 | 1.78E-06  |
| two-m27            | 3.8        | 1.70 | 1.29 | 7.13E-04  |
| two-m28            | 8.1        | 1.85 | 1.50 | 5.16E-07  |
| two-m29            | 7.5        | 2.02 | 1.43 | 1.52E-06  |
| **weighted average** | 1.82      | 1.43 |      |           |
| experimental       | 2.05       | 1.80 |      |           |
**Table S3.** Relative Gibbs free energy values ($\Delta G$, in kcal/mol), vertical and adiabatic detachment energy (VDE and ADE, in eV) and equilibrium fraction values ($x_M$) calculated for the $[\text{dGMP}-2\text{H}]^2\cdot3\text{H}_2\text{O}$ systems. Data for the structures of $x_M \geq 0.01$ as well as weighted average VDE and ADE bolded.

| Name                   | $\Delta G$ | VDE  | ADE  | $x_M$      |
|------------------------|------------|------|------|------------|
| **based on the 200K MD clustering** |            |      |      |            |
| three-200-m1           | 5.8        | 2.14 | 1.75 | 9.74E-06   |
| three-200-m2           | 5.8        | 2.19 | 1.79 | 1.04E-05   |
| three-200-m3           |            |      |      |            |
| three-200-m4           |            |      |      |            |
| three-200-m5           | 9.2        | 2.29 | 1.85 | 2.99E-08   |
| three-200-m6           | 9.4        | 2.17 | 1.77 | 2.32E-08   |
| **based on the 300K MD clustering** |            |      |      |            |
| three-m1               | 0.2        | 2.17 | 1.76 | 1.33E-01   |
| three-m2               | 0.2        | 2.17 | 1.76 | 1.35E-01   |
| three-m3               | 0.5        | 2.17 | 1.77 | 7.86E-02   |
| three-m4               | 0.2        | 2.09 | 1.72 | 1.18E-01   |
| three-m5               | 0.2        | 2.11 | 1.72 | 1.21E-01   |
| three-m6               | 0.4        | 2.08 | 1.71 | 8.89E-02   |
| three-m7               | 3.3        | 1.73 | 1.44 | 6.56E-04   |
| three-m8               | 3.1        | 1.83 | 1.45 | 9.26E-04   |
| three-m9               | 2.3        | 1.79 | 1.48 | 3.53E-03   |
| three-m10              | 3.8        | 2.24 | 1.80 | 3.12E-04   |
| three-m11              |            |      |      |            |
| three-m12              | 3.9        | 2.26 | 1.82 | 2.68E-04   |
| **three-m13**          | 0.3        | 2.08 | 1.71 | 1.09E-01   |
| three-m14              | 4.1        | 1.81 | 1.45 | 1.72E-04   |
| **three-m15**          | 0.0        | 2.07 | 1.69 | 1.80E-01   |
| three-m16              | 2.4        | 1.82 | 1.50 | 3.06E-03   |
| three-m17              | 3.1        | 2.09 | 1.70 | 9.57E-04   |
| three-m18              | 1.8        | 1.90 | 1.55 | 8.50E-03   |
| three-m19              | 3.2        | 2.10 | 1.71 | 8.64E-04   |
| three-m20              | 6.5        | 1.96 | 1.60 | 2.97E-06   |
| **three-m21**          | 1.4        | 2.37 | 1.93 | 1.73E-02   |
| three-m22              | 7.0        | 1.76 | 1.46 | 1.30E-06   |
| three-m23              | 4.0        | 2.30 | 1.85 | 2.20E-04   |
| three-m24              | 5.0        | 1.83 | 1.48 | 3.71E-05   |
| three-m25              | 5.4        | 1.77 | 1.48 | 2.03E-05   |
| **weighted average**   | 2.11       | 1.73 |      |            |
| **experimental**       | 2.20       | 1.95 |      |            |
Table S4. Relative Gibbs free energy values ($\Delta G$, in kcal/mol), vertical and adiabatic detachment energy (VDE and ADE, in eV) and equilibrium fraction values ($x_M$) calculated for the $[\text{dGMP-2H}]^2 \cdot 3\text{H}_2\text{O}$ systems. Data for the structures of $x_M \geq 0.01$ as well as weighted average VDE and ADE bolded.

| Name based on the 200K MD clustering | $\Delta G$ | VDE  | ADE  | $x_M$     |
|-------------------------------------|-----------|------|------|-----------|
| four-200-m1                         | 10.5      | 2.30 | 1.88 | 7.49E-09  |
| four-200-m2                         | the same as four-200-m1, excluded |
| four-200-m3                         | 10.4      | 2.32 | 1.88 | 8.74E-09  |
| four-200-m4                         | 10.2      | 2.24 | 1.83 | 1.22E-08  |
| four-200-m5                         | the same as four-200-m1, excluded |
| four-200-m6                         | 7.8       | 2.56 | 1.95 | 6.69E-07  |
| four-200-m7                         | 7.6       | 2.27 | 1.86 | 1.04E-06  |
| four-200-m8                         | 6.8       | 2.29 | 1.87 | 3.67E-06  |
| four-200-m9                         | 7.9       | 2.50 | 1.91 | 6.42E-07  |
| four-200-m10                        | 10.5      | 2.16 | 1.77 | 7.43E-09  |

| Name based on the 300K MD clustering | $\Delta G$ | VDE  | ADE  | $x_M$     |
|-------------------------------------|-----------|------|------|-----------|
| four-m1                             | 1.6       | 2.23 | 1.76 | 2.52E-02  |
| four-m2                             | 5.5       | 2.10 | 1.66 | 3.59E-05  |
| four-m3                             | 2.0       | 2.13 | 1.75 | 1.25E-02  |
| four-m4                             | 1.1       | 2.14 | 1.76 | 6.29E-02  |
| four-m5                             | 1.7       | 2.14 | 1.76 | 2.03E-02  |
| four-m6                             | 5.0       | 2.18 | 1.80 | 8.45E-05  |
| four-m7                             | 3.3       | 2.46 | 1.78 | 1.53E-03  |
| four-m8                             | 1.6       | 2.18 | 1.79 | 2.58E-02  |
| four-m9                             | 5.0       | 2.34 | 1.90 | 8.82E-05  |
| four-m10                            | 3.4       | 2.34 | 1.80 | 1.20E-03  |
| four-m11                            | 3.4       | 2.32 | 1.79 | 1.29E-03  |
| four-m12                            | 5.0       | 2.41 | 1.93 | 7.85E-05  |
| four-m13                            | 0.7       | 2.39 | 1.97 | 1.20E-01  |
| four-m14                            | 1.1       | 2.17 | 1.79 | 5.81E-02  |
| four-m15                            | 1.1       | 2.25 | 1.84 | 6.43E-02  |
| four-m16                            | 0.8       | 2.12 | 1.75 | 9.98E-02  |
| four-m17                            | the same as four-m15, excluded |
| four-m18                            | 2.9       | 1.86 | 1.56 | 2.77E-03  |
| four-m19                            | 5.0       | 1.88 | 1.57 | 8.67E-05  |
| four-m20                            | 5.4       | 1.91 | 1.49 | 3.97E-05  |
| four-m21                            | 6.3       | 2.35 | 1.88 | 8.55E-06  |
| four-m22                            | 2.8       | 2.14 | 1.76 | 3.59E-03  |
| four-m23                            | 0.0       | 2.34 | 1.90 | 3.80E-01  |
| four-m24                            | 4.8       | 2.21 | 1.80 | 1.22E-04  |
| four-m25                            | 6.1       | 2.38 | 1.91 | 1.18E-05  |
| four-m26                            | 5.7       | 1.83 | 1.48 | 2.34E-05  |
| four-m27                            | 1.7       | 2.24 | 1.82 | 2.22E-02  |
| four-m28                            | 3.3       | 2.12 | 1.75 | 1.36E-03  |
| four-m29                            | 6.6       | 2.40 | 1.80 | 5.52E-06  |
Table S5. Water binding energies (WBE) – free energy of cluster formation ($\Delta G_{\text{hyd}}$, in kcal/mol), BSSE corrected hydration energies ($\Delta G_{\text{hyd-BSSE}}$, in kcal/mol), WBE per water molecule ($\Delta G_{\text{hyd-BSSE}}/n$, in kcal/mol) and experimental VDE increment ($\Delta VDE_{\text{exp}} = nVDE_{\text{exp}} - (n-1)VDE_{\text{exp}}$, in eV), calculated for the most stable \([\text{dGMP-2H}]^{2-}n\text{H}_2\text{O}\) systems.

| $N \text{H}_2\text{O}$ | Name       | $\Delta G_{\text{hyd}}$ | $\Delta G_{\text{hyd-BSSE}}$ | $\Delta G_{\text{hyd-BSSE}}/n$ | $\Delta VDE_{\text{exp}}$ |
|-----------------------|------------|--------------------------|-------------------------------|--------------------------------|---------------------------|
| 1                     | one-m23    | -18.74                   | -18.01                        | -18.01                         | 0.50                      |
| 2                     | two-m19    | -24.74                   | -22.93                        | -11.46                         | 0.25                      |
| 3                     | three-m15  | -29.14                   | -26.68                        | -8.89                          | 0.15                      |
| 4                     | four-m23   | -34.27                   | -30.48                        | -7.62                          | 0.10                      |

Figure S1. Water binding energy per water molecule ($\Delta G_{\text{hyd-BSSE}}/n$) dependence on experimental VDE increment ($\Delta VDE_{\text{exp}}$).

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