Supporting Information

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High-Voltage Aqueous Mg-Ion Batteries Enabled by Solvation Structure Reorganization

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Figure S1. Ionic conductivity (κ) and viscosity (η) of a series of designed aqueous Mg-ion electrolytes (AMEs). Ionic conductivity (a), viscosity (b), and log (η⁻¹) vs log(κ) plot (c) of 0.8 m Mg(TFSI)₂-xPEG-(1-x)H₂O, where all measurements were done at room temperature and 25°C, respectively, x=0, 75%, 80%, 85%, 90%, and 95%. When x=0, it has a high ionic conductivity of 37.75 mS cm⁻¹ and the lowest viscosity of 2 × 10⁻³ Pa s. Pure PEG solvent has viscosity of ~0.110 Pa s. Note it took around 2 days to completely dissolve Mg(TFSI)₂ into the 95%PEG solvent due to the relatively low solubility of Mg(TFSI)₂ and the strong viscosity of the electrolyte.
Figure S2. Flammability evaluation. (a) A burning cotton swab was soaked in the commercial LP30 (1 M LiPF$_6$ in ethylene carbonate:dimethyl carbonate in a ratio of 1:1, w/w) (snapshot of Video S1) and 0.8 m Mg(TFSI)$_2$-85%PEG-15%H$_2$O (snapshot of Video S2), where LP30 was fired immediately, while the fire of cotton swab was extinguished in 0.8 m Mg(TFSI)$_2$-85%PEG-15%H$_2$O. (b) Flammability testing of glass fiber immersed in LP30 (snapshot of Video S3) and 0.8 m Mg(TFSI)$_2$-85%PEG-15%H$_2$O (snapshot of Video S4).

Figure S3. Electrochemical stability window of AMEs 0.8 m Mg(TFSI)$_2$-85%PEG-15%H$_2$O on GC and SS between −2.5 V and 2.0 V vs AgCl/Ag at 0.5 mV s$^{-1}$. These potential limits correspond to 0.08 V and 3.78 V vs Mg$^{2+}$/Mg shifted by 2.58 V against the values for the used AgCl/Ag reference.
Figure S4. Illustration of the evolution of the total energy and root-mean-square deviation (RMSD) of Mg in the 0.8 m Mg(TFSI)$_2$-H$_2$O (a, b) and 0.8 m Mg(TFSI)$_2$-85%PEG-15%H$_2$O (c, d) system during MD simulation at 300 K. The radial distribution function (RDF) g(r) (solid lines) and corresponding integrated coordination numbers (ICN) n(r) (dashed lines) (e) of Mg$^{2+}$-TFSI' in both 0.8 m Mg(TFSI)$_2$-H$_2$O and 0.8 m Mg(TFSI)$_2$-85%PEG-15%H$_2$O.

The g(r) profile of Mg$^{2+}$-TFSI' in 0.8 m Mg(TFSI)$_2$-H$_2$O electrolyte shows a first peak at around 0.43 nm, whereas it displays one first peak at around 0.21 nm with the ICN of 0.73 in 0.8 m Mg(TFSI)$_2$-85%PEG-15%H$_2$O (Figure S4e), where TFSI' is much closer to Mg$^{2+}$ than that in 0.8 m Mg(TFSI)$_2$-H$_2$O electrolyte and belongs to the first solvation shell of Mg$^{2+}$, demonstrating the “quasi-water-in-salt” property of 0.8 m Mg(TFSI)$_2$-85%PEG-15%H$_2$O electrolyte.
Figure S5. Structure and morphology study of V$_2$O$_5$ nanowires (Mo K$_\alpha_1$ radiation, $\lambda=0.70932$ Å). (a) Rietveld refinement based on X-ray diffraction data and (b) SEM image of pristine V$_2$O$_5$ nanowires.

Figure S6. Cyclic voltammetry (CV) in 0.8 m Mg(TFSI)$_2$-100%H$_2$O at a scan rate of 0.05 mV s$^{-1}$ in the potential range of 1.88-3.33 V vs Mg$^{2+}$/Mg (-0.75, 0.75 V vs AgCl/Ag).

Figure S7. Charge–discharge curves (a) and cycling performance (b) of V$_2$O$_5$ in 0.8 m Mg(TFSI)$_2$-100%H$_2$O at 50 mA g$^{-1}$ in 0.8 m Mg(TFSI)$_2$-100%H$_2$O in the potential range of 1.88-3.33 V vs Mg$^{2+}$/Mg.
Figure S8. Rietveld refinement based on X-ray diffraction data of V$_2$O$_5$ nanowires at 2.15 V, about 50% of depth of discharge (a), where an additional small reflection at 16.7° might be related to MgO and the other one at 21.06° cannot be indexed on any known phase. Le Bail fitting based on synchrotron diffraction data of V$_2$O$_5$ at 1.58 V, 100% of depth of discharge (b), and at 2.76 V, about 50% of depth of charge (c).
| Electrolyte                   | Ionic conductivity (mS cm\(^{-1}\)) | Viscosity (Pa s) |
|------------------------------|--------------------------------------|-----------------|
| 1 0.8 m Mg(TFSI)\(_2\)-100%H\(_2\)O | 37.75                                | 2 × 10\(^{-3}\) |
| 2 0.8 m Mg(TFSI)\(_2\)-75%PEG-25%H\(_2\)O | 0.634                                | 0.289(17)       |
| 3 0.8 m Mg(TFSI)\(_2\)-80%PEG-20%H\(_2\)O | 0.346                                | 0.516(20)       |
| 4 0.8 m Mg(TFSI)\(_2\)-85%PEG-15%H\(_2\)O | 0.183                                | 0.951(30)       |
| 5 0.8 m Mg(TFSI)\(_2\)-90%PEG-10%H\(_2\)O | 0.093                                | 1.995(67)       |
| 6 0.8 m Mg(TFSI)\(_2\)-95%PEG-5%H\(_2\)O | 0.043                                | 4.752(38)       |
| Electrolyte                                                                 | Battery-type | Ionic conductivity (mS cm\(^{-1}\)) | Year | Reference |
|---------------------------------------------------------------------------|--------------|--------------------------------------|------|-----------|
| “Water-in-Salt” 21 m LiTFSI                                              | LIBs         | 8.21 (25 °C)                         | 2015 | [1]       |
| “Water-in-Bisalt” 21 m LiTFSI-7 m LiOTf                                   | LIBs         | 6.5 (25 °C)                          | 2016 | [2]       |
| “Hydrate Melt” Li(TFSI)\(_{0.7}\)(BETI)\(_{0.3}\)2H\(_2\)O                   | LIBs         | 3.0 (30 °C)                          | 2016 | [3]       |
| (19.4 m LiTFSI-8.3 m LiBETI)                                              | LIBs         | 5.0 (30 °C)                          | 2018 | [4]       |
| “Hybrid Aqueous/Nonaqueous” 21 m LiTFSI in H\(_2\)O-9.25 m LiTFSI in DMC\(\text{wt\%}=1:1\) | LIBs         | 0.1 (30 °C)                          | 2019 | [5]       |
| “Monohydrate Melt” Li(PTFSI)\(_{0.6}\)(TFSI)\(_{0.4}\)1H\(_2\)O              | MIBs         | several                              | 2000 | [8]       |
| (22.2 m LiTFSI-33.3 m LiPTFSI) Li(TFSI)\(_{0.3}\)(MM3411)\(_{0.2}\)-1.4H\(_2\)O | MIBs         | 0.33 (25 °C)                         | 2019 | [6]       |
| 31.4 m LiTFSI-7.9 m LiMM3411                                              | MIBs         | 0.5 (25 °C)                          | 2014 | [7]       |
| 0.5 M Mg[TFSI]\(_2\) in a glyme/diglyme mixture                           | MIBs         | 5.2 (25 °C)                          | 2017 | [9]       |
| Mg(AlCl\(_2\)BuEt)\(_2\)/Tetrahydrofuran                               | MIBs         | 0.07 (600 °C)                        | 2000 | [10]      |
| (PhMgCl)\(_2\)-AlCl\(_3\)/Tetrahydrofuran                              | MIBs         | 10 \(^{-3}\) (150 °C)               | 2014 | [12]      |
| 0.6 M MgBOR(hfip)/DME                                                    | MIBs         | 7.23 (725 °C)                        | 2016 | [14]      |
| MgCl\(_2\)-Mg(TFSI)\(_2\)/1,2-dimethoxyethane (DME)                     | MIBs         | 6.8 (25 °C)                          | 2017 | [10]      |
| Mg(BH\(_4\))(NH\(_2\))                                                | MIBs         | 6 \times 10 \(^{-3}\) (30 °C)       | 2017 | [15]      |
| Mg\(_{1-2x}\)(Zr\(_1-x\)Nb\(_x\))P\(_2\)O\(_{24}\) x = 0.15 in MgZr\(_4\)P\(_6\)O\(_{24}\) | MIBs         | 2.9-5.8 (25 °C)                     | 2013 | [11]      |
| Magnesium Ethylenediamine Borohydride, Mg(NH\(_2\)CH\(_2\)CH\(_2\)NH\(_2\))\(_2\) | MIBs         | 6 \times 10 \(^{-2}\) (70 °C)       | 2017 | [15]      |
| MgSc\(_2\)Se\(_4\)                                                    | MIBs         | 0.01 (25 °C)                         | 2017 | [16]      |
| poly(ethylene glycol) dimethacrylate (PEGDMA)-Mg(TFSI)\(_2\) gel electrolyte | MIBs         | 0.1 (25 °C)                          | 2019 | [17]      |
| poly(methylmethacrylate) (PMMA)-Mg(CF\(_3\)SO\(_3\))\(_2\) gel electrolyte | MIBs         | 0.4 (20 °C)                          | 2002 | [18]      |
| Mg(AlCl\(_2\)EtBu)\(_2\)/tetruglyme/PVdF polynomial (ethylene oxide) (PEO) | MIBs         | 3.7 (25 °C)                         | 2003 | [19]      |
| poly(vinylidene fluoride) (PVDF)-Mg(TFSI)\(_2\)                         | MIBs         | 1.2 \times 10 \(^{-2}\) (70 °C)     | 2016 | [20]      |
| LiTFSI-Mg(TFSI)\(_2\)-acetamide with molar ratio of 10:1:40             | LMIBs        | 85 (25 °C)                           | 2020 | [21]      |
Table S3. The electrochemical stability window (ESW) of several Mg-ion organic electrolytes

| Electrolyte                                                                 | ESW  | Reference |
|-----------------------------------------------------------------------------|------|-----------|
| Mg Organoborate Moieties (Mg(BBu₃Ph₂)₂ in THF)                               | ~1.9 V | [22]      |
| Magnesium haloalky aluminate complex                                         | ~2.2 V | [8]       |
| (Mg(AlCl₃BuEt)₂)                                                           |      |           |
| Mg(TFSI)₂-MgCl₂ in DME                                                      | ~3.0 V | [23]      |
| Magnesium hexamethyldisilazide Mg(HMDS)₂–4MgCl₂ in THF                      | ~2.8 V | [24]      |
| Mg(BH₄)₂ in THF                                                             | ~2.3 V | [25]      |
| (PhMgCl)₂–AlCl₃ in THF (APC)                                                | ~3.3 V | [9]       |
| Magnesium aluminum                                                          | ~3.5 V | [26]      |
| hexafluoroisopropoxideMg[Al(HFIP)₄]₂                                        |      |           |
| [Mg(THF)₆][AlCl₄]₂ in PYR14TFSI/THF                                        | ~2.5 V | [27]      |
| Magnesocene/THF                                                             | ~1.8 V | [28]      |
| Magnesium tetrakis(hexafluoroisopropyloxy) borate                           | ~4.5 V | [29]      |
| Mg-[B(hfip)₄]₂ (hfip = OC(H)(CF₃)₂)                                         | ~3.5 V | [10]      |
| Boron-centered anion-based magnesium electrolyte (BCM electrolyte)          |      |           |
| Hybrid AlCl₃/MgCl₂/Mg(TFSI)₂-THF                                            | ~2.6 V | [30]      |
| [Mg(G4)][TFSA]₂/[PYR13][TFSA], tetraglyme(G4)                               | ~4.1 V | [31]      |
| Perfluorinated pinacolatoborate, Mg[B(O₂C₂(CF₃)₄]₂                            | ~4.0 V | [32]      |
| (Mg-FPB)                                                                   |      |           |
| LiTFSI-Mg(TFSI)₂-acetamide with molar ratio of 10:1:40                      | ~3.0 V | [21]      |

Table S4. Hydrogen-bonds (HBs) contribution of both electrolytes and lifetime of HBs from molecular dynamics (MD) calculations

| Electrolyte | Origin of HBs contribution | lifetime of HBs |
|-------------|-----------------------------|-----------------|
|             | TFSI-H₂O                    | H₂O-H₂O         | PEG-H₂O         |
| 0.8 m Mg(TFSI)₂-100%H₂O | 5.4/TFSI                  | 1.5/H₂O       | --              | 50.86 ps |
| 0.8 m Mg(TFSI)₂-85%PEG-15%H₂O | 1.6/TFSI                | 1/H₂O         | 1.9/PEG        | 79.33 ps |

Table S5. The first peak position and corresponding integrated coordination numbers (ICN) of solvation structure in both electrolytes from MD calculations

| Electrolyte | First peak position (nm)/corresponding ICN |
|-------------|---------------------------------------------|
|             | Mg²⁺-H₂O               | Mg²⁺-TFSI   | Mg²⁺-PEG |
| 0.8 m Mg(TFSI)₂-100%H₂O | 0.20/5.47               | 0.43/--     |         |
| 0.8 m Mg(TFSI)₂-85%PEG-15%H₂O | 0.20/3.5                | 0.21/0.73  | 0.19/2.0 |
### Table S6. Electrochemical performance comparison of V2O5 in different electrolytes

| Electrolyte | Working voltage (V vs Mg2+/Mg)/ Temperature | Initial (max) discharge capacity (mAh g⁻¹)/ current density (mA g⁻¹) | Capacity retention (cycles) | Ref |
|-------------|--------------------------------------------|---------------------------------------------------------------------|-----------------------------|-----|
| V2O5        | 0.8 m Mg(TFSI)₂−85%PEG−15%H2O              | 1.58-3.68                                                           | 359/50                      | 80% (100) This work |
| metastable ζ- V2O5 | 0.2 M Mg(TFSI)₂ in PC | 0.2-3.4/50°C                                                      | 140/6                       | 64 (100) [33] |
| V2O5 nanoclusters /carbon | 0.2M [Mg₂(μ-Cl)₂(DME)₄][AlCl₄]₂ in DME | 0.5-2.8                                                           | 340 /40                     | 26% (40) [34] |
| V2O5-PEO nanocomposites | 0.5 M Mg(ClO₄)₂ in acetonitrile | 1.0-3.0                                                           | 125/10                      | 77%(35) [35] |
| V2O5        | 1.0 M Mg(TFSI)₂ in diglyme with 2600 ppm water | 0.5-4.2                                                           | 60(260)/(20μA cm⁻²)         | [36] |
| GO/V2O5     | 0.25 M Mg(AlCl₂EtBu)₂ in THF               | 1.0-2.8                                                            | 178/0.2C                   | 79%(20) [37] |
| V2O5 film   | 0.1 M Mg(TFSI)₂ in acetonitrile            | 2.2-3.0                                                            | 150/ (0.5μA cm⁻²)          | [38] |
| ζ-V2O5      | 0.5 M MgTFSI₂−PY₁₄TFSI in 3.6/120°C       | 0.7-1.1                                                             | 130/15                     | [39] |
| Amorphous V2O5 | 0.3 M Mg(TFSI)₂ in glyme: diglyme | 1.1-3.4                                                           | 30(180)/30                 | 71%(40) [40] |
| V2O5        | 0.5 M MgTFSI₂−PY₁₄TFSI in 3.5/110°C        | 1.3-3.5                                                             | 295/(C/20, C/5)            | 76%(50) [41] |

**Video S1:** Flammability testing of commercial LP30 in EC/DMC electrolyte. The commercial LP30 in the EC/DMC electrolyte started to burn after an ignited cotton swab was immersed in the electrolyte, identifying that the electrolyte is flammable.

**Video S2:** Flammability testing of the 0.8 m Mg(TFSI)₂−85%PEG−15%H₂O electrolyte. The fire of an ignited cotton swab was extinguished after being immersed in the 0.8 m Mg(TFSI)₂−85%PEG−15%H₂O electrolyte, demonstrating that the electrolyte is non-flammable and safe.

**Video S3:** Flammability of LP30. LP30 electrolyte soaked glass fiber was ignited, indicating LP30 electrolyte is flammable.

**Video S4:** Flammability of 0.8 m Mg(TFSI)₂−85%PEG−15%H₂O. 0.8 m Mg(TFSI)₂−85%PEG−15%H₂O electrolyte soaked glass fiber was not ignited, while LP30 electrolyte soaked one was ignited, indicating that 0.8 m Mg(TFSI)₂−85%PEG−15%H₂O electrolyte displays superior safety compared to LP30 electrolyte.
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