Vitrimeric and self-healing electrolytes

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Abstract:

Generating novel electrolyte-systems is of urgent need, for batteries as well as for supercapacitors acting as short-time power-storage devices. While solid electrolytes offer promising alternatives to the conventional liquid electrolytes, they undergo irreversible failures during cycling. Intrinsic self-healing materials [1] bearing dynamic bonding are the proper candidates as electrolytes [2], since reversible bonding can compensate failure of the materials consequently extending the lifespan of a battery.

We here report on the use of dynamic covalent bonding systems to embed self-healing [3] and vitrimeric properties [4] into electrolyte-systems, requiring fundamental knowledge about bonding strength and dynamics in highly polar and even ionic-liquid environment. We have focused on two different bonding systems, one related to hydrogen bonding; the other to covalent dynamic bonds, enabling vitrimeric exchange during charge-transport through the electrolyte-medium. Critical is to maintain the dynamic feature in the temperature window of interest, still keeping the required ion-conductivities intact. The materials we are reporting herewith not only suffice these requirements, but also allow to embed self-healing properties into the final (solid) material. Using specifically designed polymers, we also report on changes in thermodynamic properties exerted by the surrounding medium, required for the electrolyte to maintain its function. Furthermore, we demonstrate the 3D-printability of these electrolyte-composites [5].

[1] a*Polymers* **2021**, *13*; b *Materials Today: Proceedings* **2021**, *34*, 243-249; c *ACS Applied Polymer Materials* **2020**, *2*, 4127-4139; d *Composites Part B: Engineering* **2019**, *157*, 1-13; e*Macromolecular Rapid Communications* **2018**, 1700739-n/a.

[2] a *Advanced Functional Materials* **2020**, *30*, 1909912; b*Advanced Energy Materials* **2020**, *10*, 2002815.

[3] *Polymers* **2022**, *14*, 4090.

[4] a *Polymers* **2022**, *14*, 2456; b *Chemical Engineering Journal* **2022**, *433*, 133261; c *Composites Part B: Engineering* **2020**, *184*, 107647.

[5] a*Advanced Materials Technology* **2022**, ASAP, admt.202200088R202200081; b *Frontiers in Chemistry* **2021**, *9*; c *Nanomaterials* **2022**, *12*, 1859.