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## **Introducing modifiers in hybrid glasses**

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Hybrid glasses, including glassy metal-organic frameworks and coordination polymers, consist of metal ion nodes connected by organic linkers through coordination bonds, often resembling the network structure of, e.g., silicate glasses. However, the latter may be modified by adding so-called “fluxes” or “modifiers” to tune their structure and properties. Such approach for structure-property tuning does not yet exist for hybrid glasses, which is a major limitation for enabling their applications. In this work, we present methods for continuous composition and thus structure modification in hybrid glasses, mimicking the concept of modifiers in oxide glasses. Specifically, we show that water (and other electron donating compounds) addition to both hybrid cobalt bis-acetamide and zeolitic imidazolate framework (ZIF) crystals significantly modifies their melting and glass transition temperatures as well as mechanical properties. Based on a combination of *ab initio* simulations and spectroscopic analyses, we show how the modifiers depolymerize the continuous network, ultimately affecting the macroscopic thermal and mechanical properties of the hybrid glasses. As such, our results may pave the way for melt-quenching previously un-meltable crystals as well as continuous tuning of glass forming propensity and material properties.