

Steps Towards Understanding Water-Responsiveness

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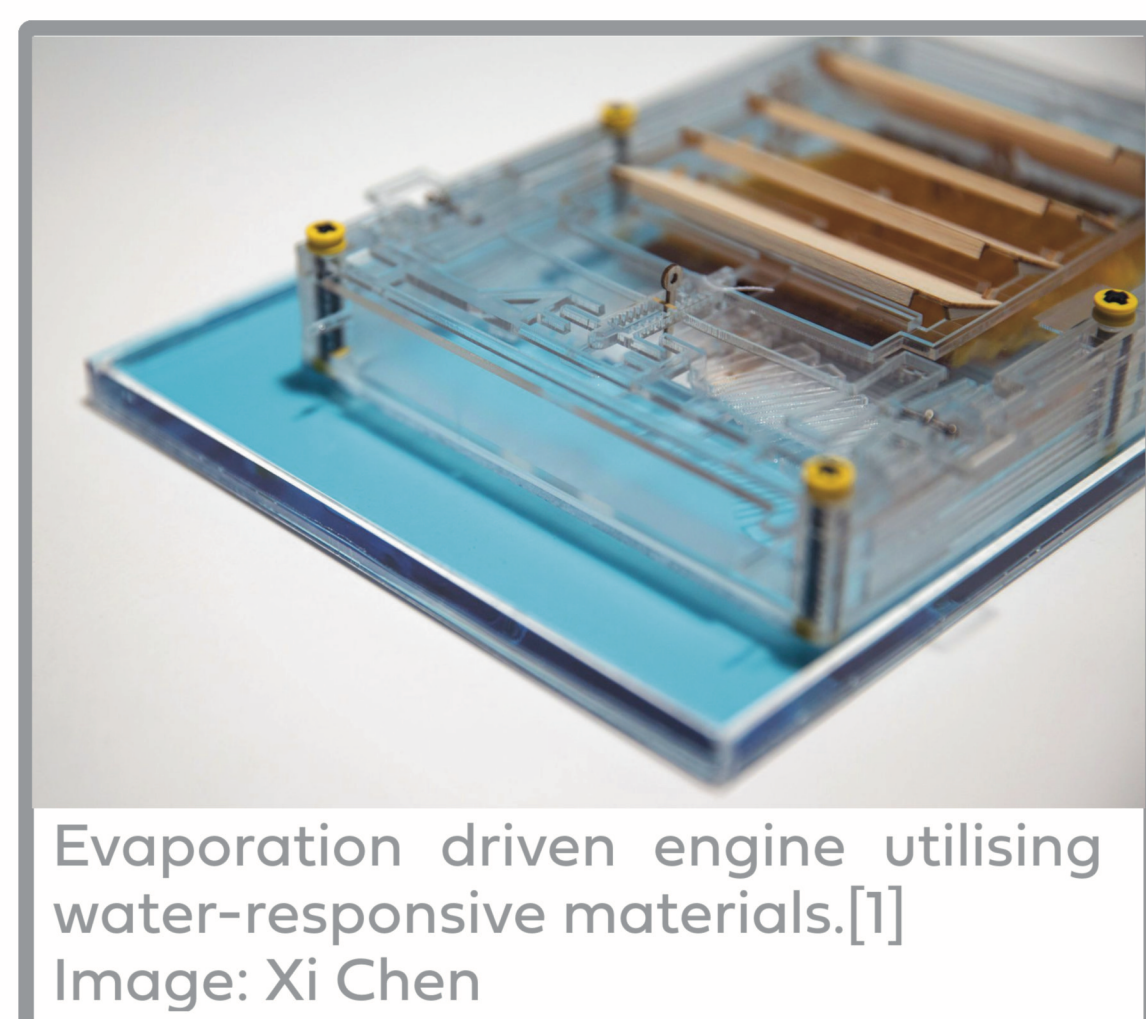


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Water-Responsiveness

Water responsive materials dramatically alter their size or shape in response to changes in the humidity of their environment, with promising applications in energy storage, soft robotics and wearable technology.[1]

Nature provides many examples of water responsive processes, including the opening of pine cones. Evaporation powered engines have been demonstrated, which run at an air-water interface, and are able to generate enough power to illuminate an LED.[1]



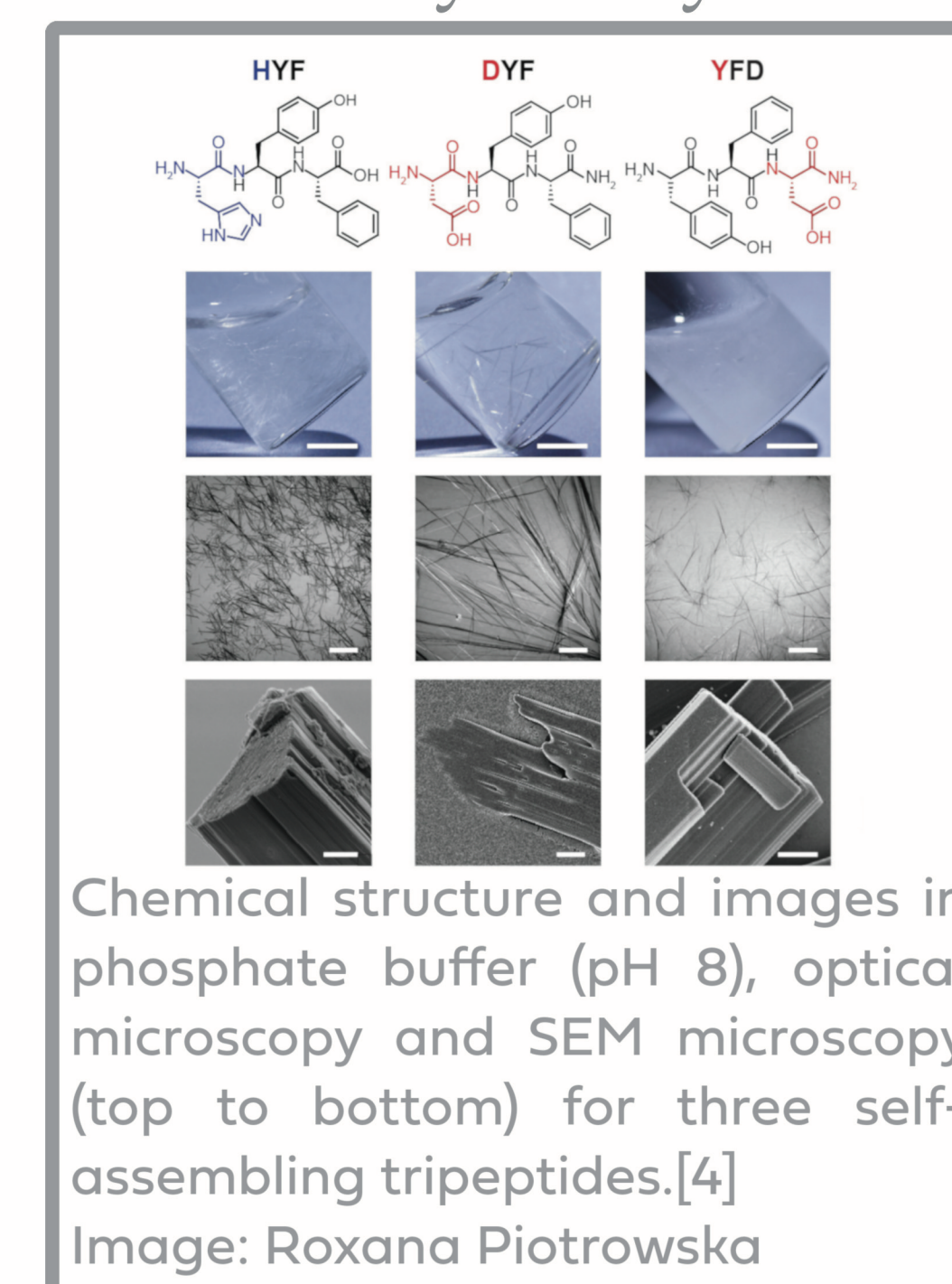
Despite the promising applications of these materials, the mechanisms underpinning water-responsiveness are poorly understood.

Self-Assembled Tripeptides

Self-assembly is the spontaneous formation of ordered patterns or structures from bulk solute molecules. The process is ubiquitous in nature and has been used widely in synthetic supramolecular chemistry.[2]

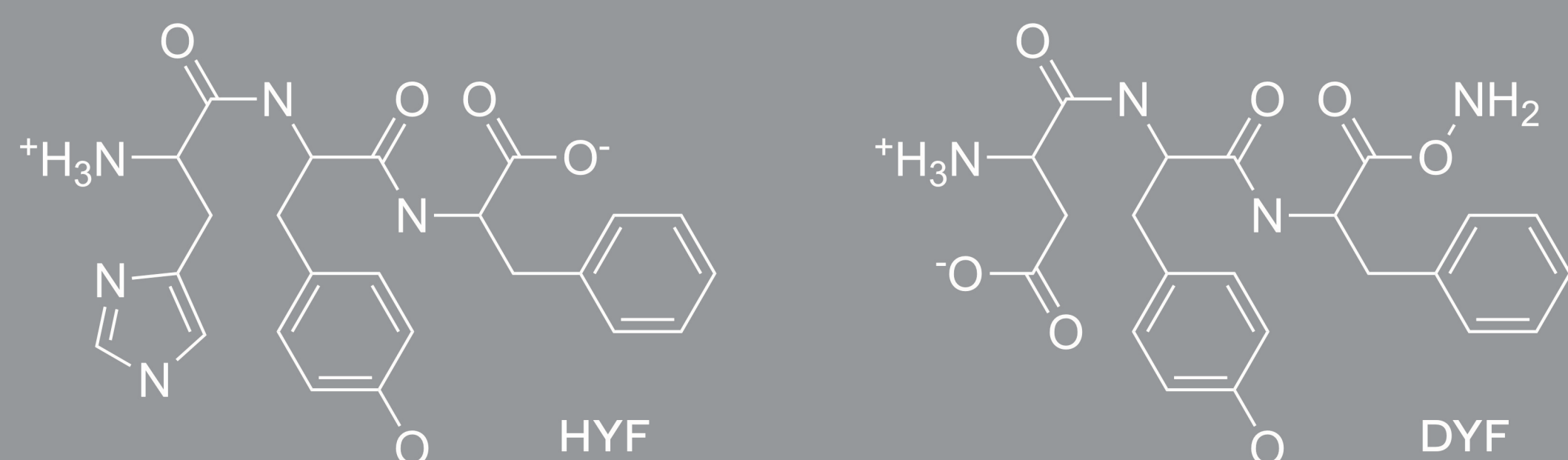
Self-assembly is an error correcting process and enables simple molecules, such as small amino acid sequences (peptides), to form larger, more complicated structures.[2,3]

As tripeptides are small, tunable, and highly sequence dependent, these are useful test systems for probing assembly mechanisms using molecular dynamics.[3]



Aims

Simulate the water responsive tripeptides *HYF* and *DYF* at various hydration levels, observing structural changes as water content is altered.

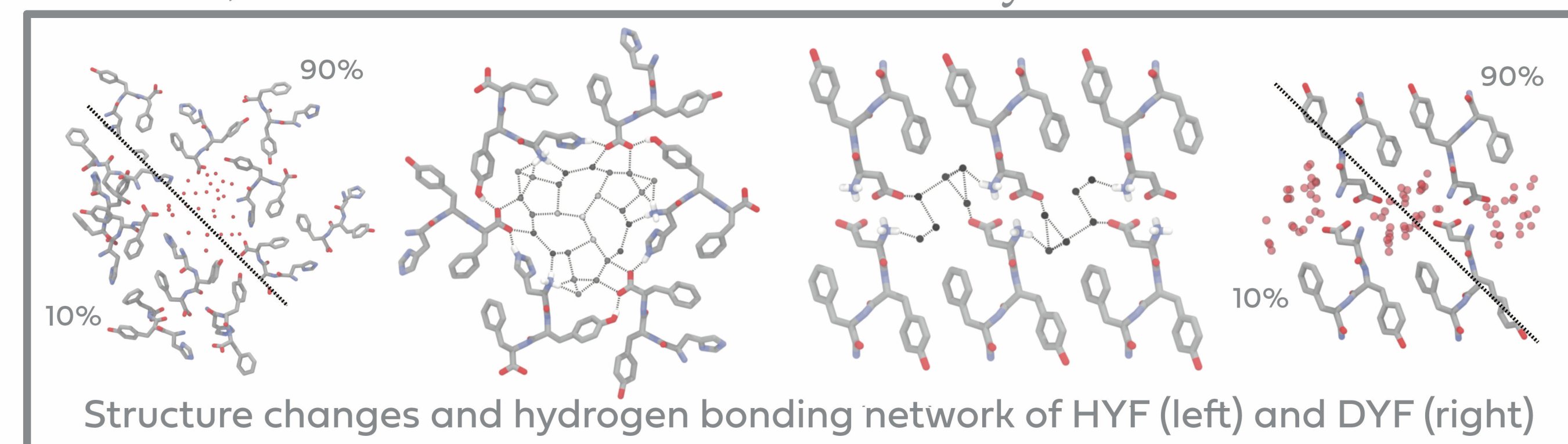


Rationalise the mechanism of water-responsiveness in *HYF* in terms of molecular interactions. Compare this with *DYF*, which is water responsive to a lesser degree.

If possible, determine a general mechanism for water-responsiveness.

Conclusion

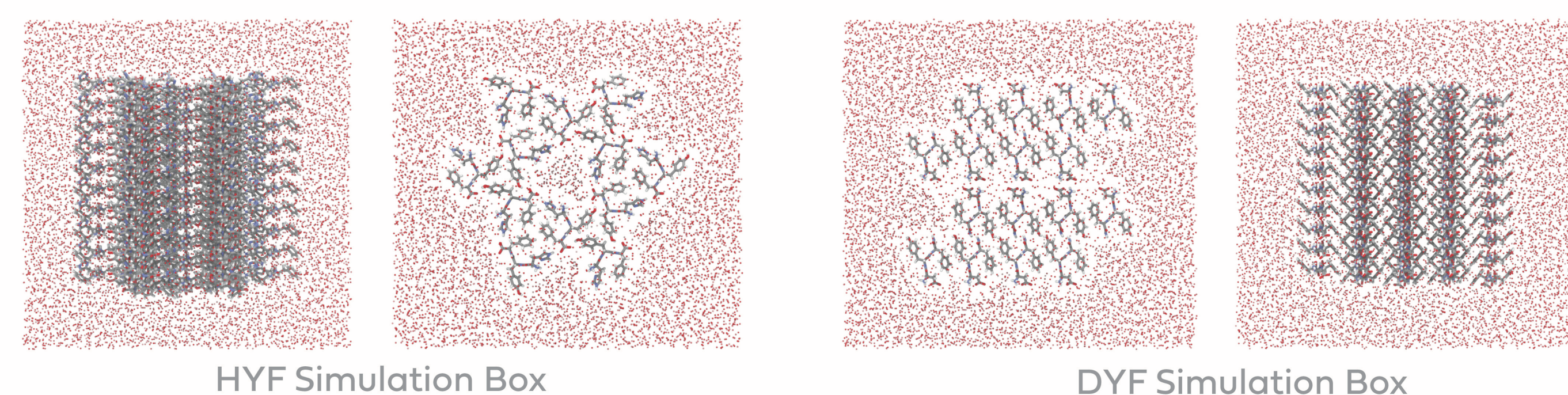
In *HYF*, stronger water binding at lower relative humidity levels leads to a difference in chemical potential between bound and unbound water. This difference is converted to mechanical energy through a strong H-bonding network, causing contraction of the pore. *DYF*, with its smaller pores and weaker network, is less able to translate humidity to mechanical work.



We believe this observation could be of significance in other natural and synthetic water responsive materials, and could be used to improve their design.[4]

Methods

Simulations of the crystal structures (10 repeating units high) for both *HYF* and *DYF* were run in triplicate in the NVT ensemble using the CHARMM36 forcefield (NAMD 2.12). Simulations were run for 20 ns at 298 K. The peptides were first solvated at standard density.



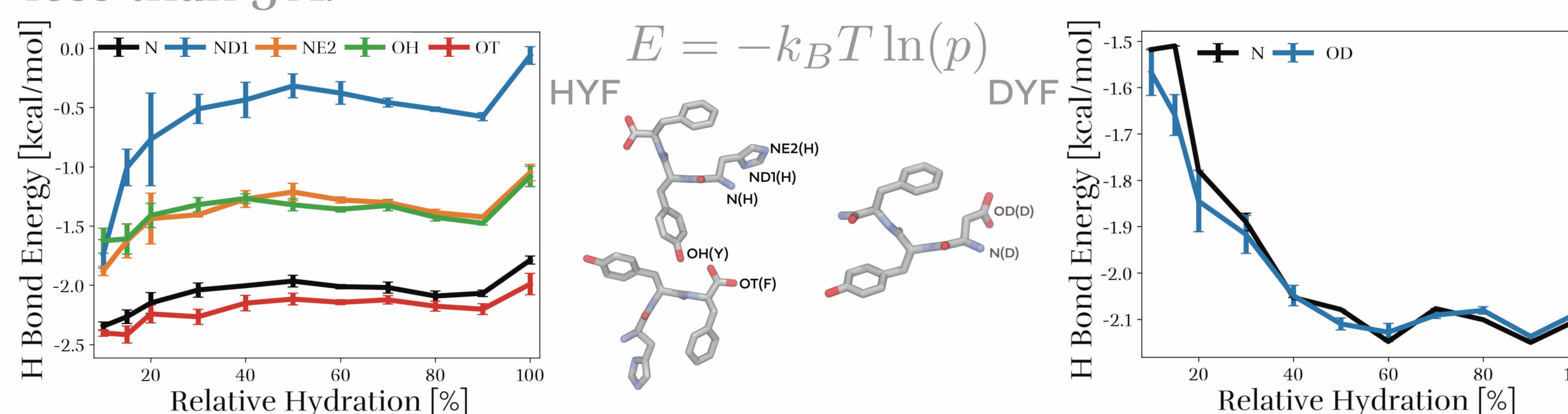
In order to simulate different humidity levels, water was removed such that only n% of the water molecules remained, where n was the intended humidity level. Distances and H-bonding were probed using the MDAnalysis package for Python 3.

Results

Pore width for both tripeptides was examined - trends were consistent with experimental results.[4]

Hydrogen bond analysis showed that while energies increase as *HYF* is dehydrated, they decrease for *DYF*.

H-bonds were defined as D-H-A angle > 120°, and H-A distance less than 3 Å.



References

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