Formation and growth of carbonaceous particles in a flame environment

Kimberly Bowal, Jacob Martin, Laura Pascazio, Markus Kraft
Computational Modelling Group
Department of Chemical Engineering and Biotechnology, University of Cambridge

CoMo GROUP

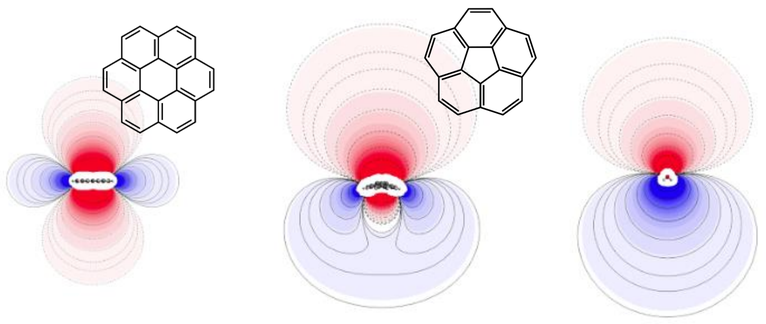
Current descriptions of soot particles are unable to explain their formation at high temperatures [1]

Recent work suggests interactions between polar curved aromatics and ions may play an important role [2] but this has not been investigated in dynamic systems

An intermolecular potential is developed for curved aromatic molecules and used within molecular dynamics to investigate the clustering dynamics and structure of soot particles

1. Intermolecular potential for curved aromatics

Curvature in aromatics produces a flexoelectric dipole moment which is not captured in existing intermolecular potentials [3]

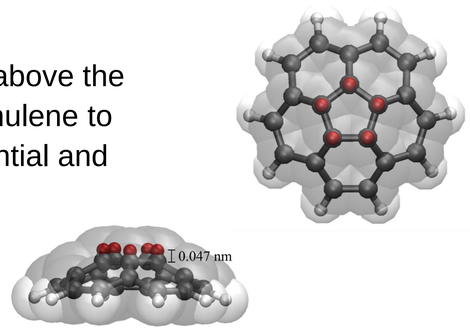


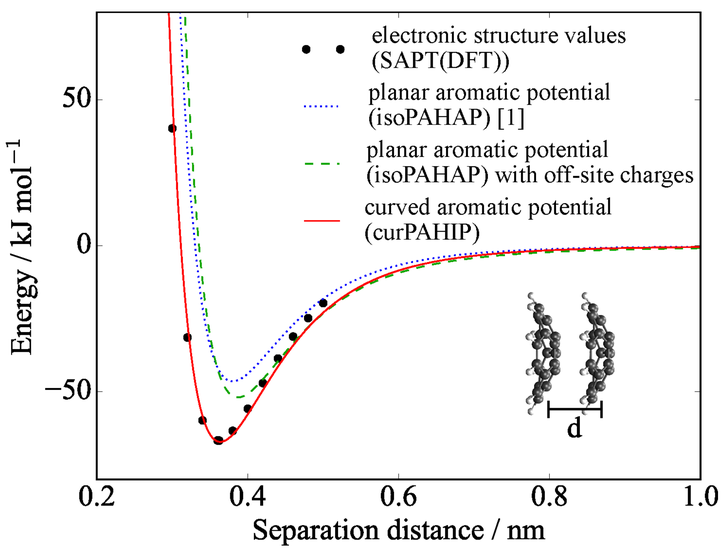
planar aromatic coronene $C_{24}H_{12}$ **0 Debye**

curved aromatic corannulene $C_{24}H_{12}$ **2.07 Debye**

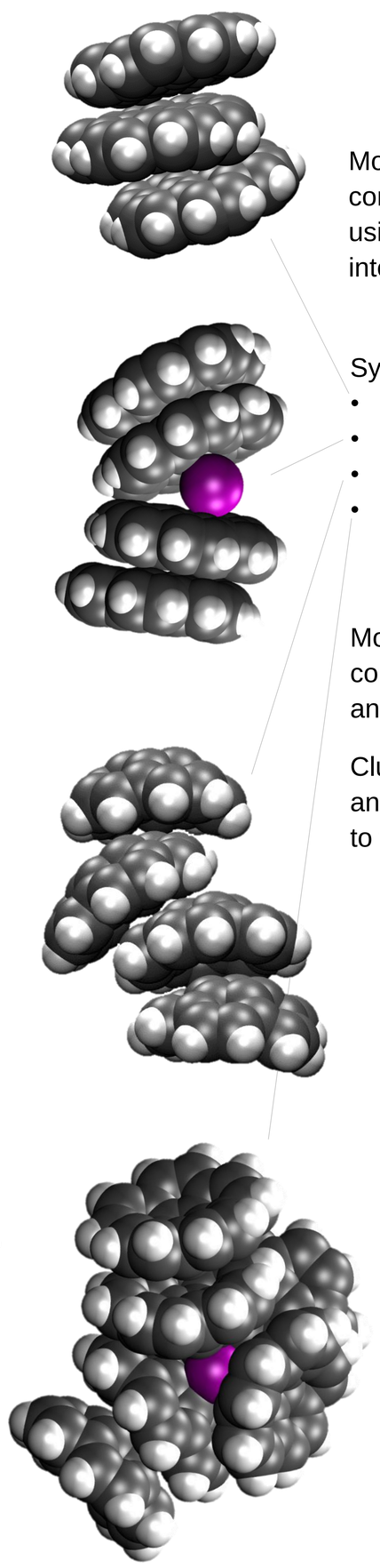
water (reference) H_2O **1.85 Debye**

Off-site charges were added above the pentagonal carbons of corannulene to describe its electrostatic potential and dipole moment



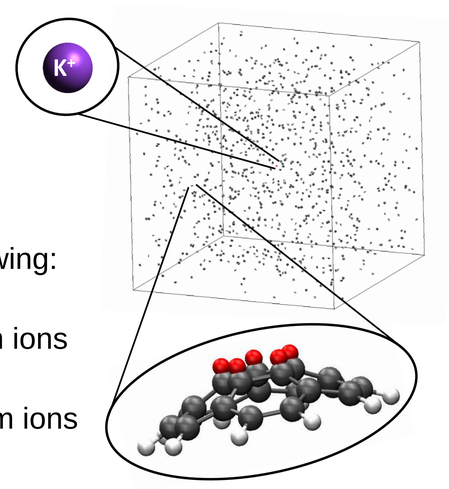


New interaction parameters were determined to produce curPAHIP, a potential that is able to describe the enhanced interactions of corannulene



2. Dynamic particle formation

Molecular dynamics simulations conducted for 1 ns at 500 – 1500 K using the newly developed curPAHIP intermolecular potential

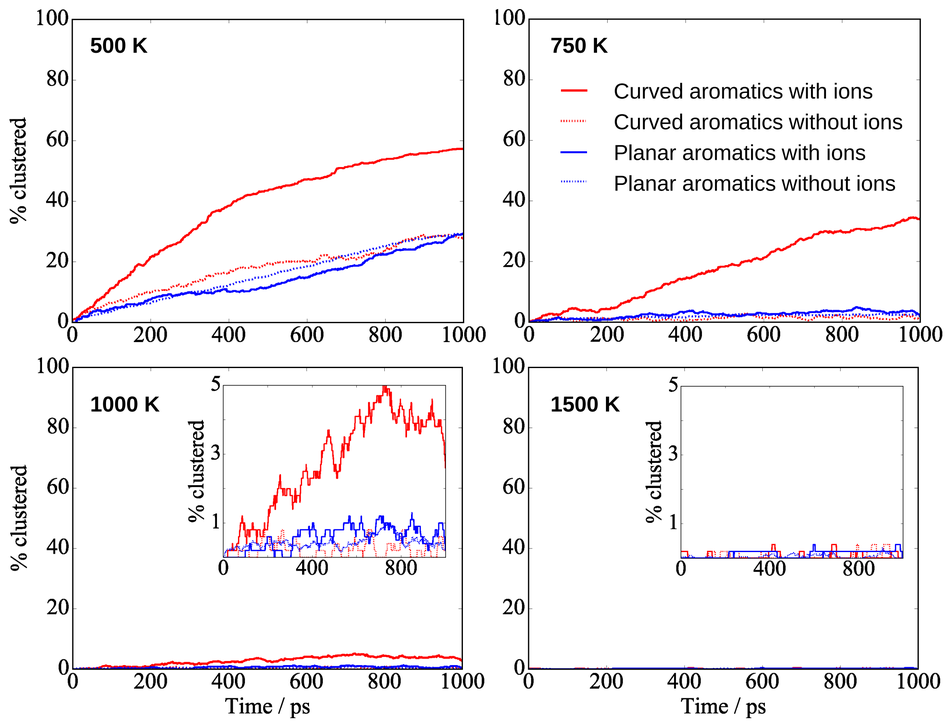


Systems contain 1000 of each of the following:

- Planar aromatic molecules
- Planar aromatic molecules & potassium ions
- Curved aromatic molecules
- Curved aromatic molecules & potassium ions

More rapid and abundant clustering of the curved aromatic corannulene with potassium ions in comparison to non-ionic systems and those containing the planar aromatic coronene

Clusters containing curved aromatics are stabilised by electrostatic and dispersive interactions and present cluster morphologies similar to those seen in experimental images of soot



3. Conclusions

- A new intermolecular potential was developed that is able to describe the polarity and enhanced interactions of curved aromatic molecules
- Molecular dynamics simulations show significant clustering of the curved aromatic corannulene with potassium ions, forming particles in which molecules are arranged compactly around the ions
- These results highlight the enhanced clustering and concentric arrangements of curved aromatics and their role in soot formation and growth
- This work has been recently published [4]

4. Future work

- Metadynamics simulations to provide quantitative nucleation properties for larger curved aromatics and chemi-ions
- Extend to other systems containing curved carbons such as batteries

References

1. Totton *et al.* (2010). *J. Chem. Theory Comput. 6*, 683-695
2. Martin *et al.* (2019). *Proceedings of the Combustion Institute*, 37, 1117-1123
3. Martin *et al.* (2017) J. Phys. Chem. C 121, 27154-27163
4. Bowal *et al* (2019). *Combustion Science and Technology*

Contact author at klb83@cam.ac.uk