

Understanding noble gas reactivity at high pressure

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Keywords: noble gas chemistry, van der Waals compounds, first principles calculations

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The chemical reactivity of the noble gas elements has been an active area of research for decades. At ambient conditions, it is known that the outer electronic shells of the heaviest noble gases can be broken up to form gas phase or solid compounds [1,2]. External pressure generally increases the reactivity of all noble gas elements, leading to formation of numerous compounds that are dominated by van der Waals, ionic, or covalent interactions [3-5]. This has potentially far-reaching consequences, for instance to understand the low abundance of xenon in the atmosphere, to estimate the helium storage capacity in Earth's crust and upper mantle, or to model the interiors of giant gas planets.

Electronic structure calculations can give detailed insights into what stabilizes such compounds. Here, we discuss a variety of studies by us and others into the chemistry (or lack thereof) behind the formation of compressed noble gas compounds [6-8].

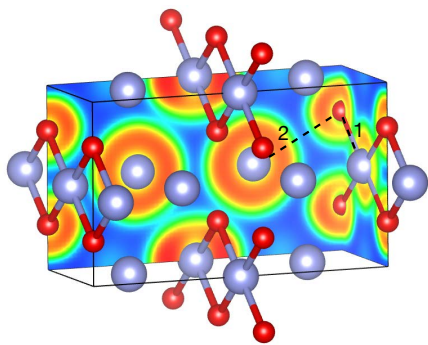


Figure 1. Electronic localization function (ELF) in Xe_3O_2 at 100GPa [6].

We will discuss molecular van der Waals compounds formed by He or Ne with small polar and non-polar molecules in the 1-10GPa range. Wave-function based approaches can fully capture dispersion interactions and allow us to assess the ability of standard density functional theory to describe the weak hydrophobic-hydration interactions in these systems as function of pressure.

We will also discuss the propensity of noble gases to form covalently bound solids in the 10-100GPa range, a

research area where crystal structure searching has true predictive power.

Lastly, we will show how the absence of chemical bonds, as seen in the Na_2He compound [9], does not preclude formation of noble gas compounds at even higher pressures, a concept that lends itself to interesting predictions to find noble gases in places they were not hitherto considered.

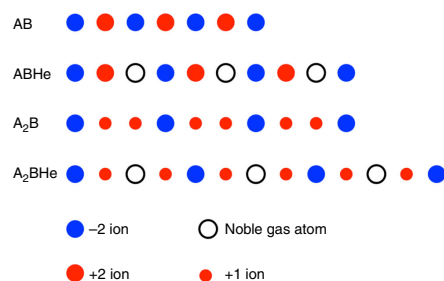


Figure 2. Schematic 1D illustration of noble gas inclusion in ionic compounds [7].

Acknowledgments: Computing resources provided by the UK national high performance computing service, ARCHER, and the UK Materials and Molecular Modelling Hub, which is partially funded by EPSRC (EP/P020194), and for which access was obtained via the UKCP consortium funded by EPSRC grant no. EP/P022561/1, are gratefully acknowledged.

- [1] N. Bartlett, *Proc. Chem. Soc.* 1962, **1962**, 218.
- [2] D.S. Brock, G. Schrobilgen, *J. Am. Chem. Soc.* 2011, **133**, 6265.
- [3] D. Londono *et al.*, *Nature* 1988, **332**, 141.
- [4] C. Sanloup *et al.*, *Science* 2005, **310**, 1174.
- [5] A. Dewaele *et al.*, *Nat. Chem.* 2016, **8**, 784.
- [6] A. Hermann, P. Schwerdtfeger, *J. Phys. Chem. Lett.* 2014, **5**, 4336.
- [7] P. Teeratchanan, A. Hermann. *J. Chem. Phys.* 2015, **143**, 154507.
- [8] Z. Liu *et al.*, *Nat. Commun.* 2018, **9**, 951.
- [9] X. Dong *et al.*, *Nat. Chem.* 2017, **9**, 440.