Firstly, what led you to investigate the thermodynamic properties of the periodic table?

Thermodynamic properties serve as a critical gauge for the performance of any computational chemistry methodology. They are less forgiving than many other properties, as simple error cancellation is less likely to occur in the calculation of properties such as enthalpies of formation. Knowing that a methodology can properly predict enthalpies (a measure of the total energy of a thermodynamic system) is a good indicator that the methodology is more likely to be effective in determining other energetic properties. Thermochemical properties are also of great importance in understanding reactivity.

What are the main aims and objectives of your research?

Some of the main aims and objectives of my research are to extend the quantitative predictive powers of computational chemistry to molecules of increasing size, and to do so with similar levels of accuracy that are possible for smaller molecules. To accomplish this, one challenge is to address the computational cost (the amount of time, memory, and disk space) required in quantum mechanical ab initio calculations. Whilst this is a challenge facing all computational scientists, the improvement of computer technology does help this to some extent. Of course, the production of large amounts of additional data by ever more powerful systems is a problem in and of itself.

Can you explain the correlation consistent Composite Approach (ccCA)? How was it developed?

There are many ways to try to reduce the computational cost of ab initio methods. Among the most popular approaches are composite. These attempt to reproduce the accuracy of energetics obtained with a sophisticated, albeit costly, computational approach, using a series of less accurate, and less computationally costly calculations. The methodology was developed by considering a number of factors including efficiency, accuracy, and applicability. We wanted a scheme that could be extended to molecules of increasing size, that would aim for the same level of accuracy (‘chemical accuracy’) achieved, on average, in gas-phase experiment-based studies for a diverse set of molecules. This amounts to 1.0 kcal/mol for main group species and 3.0 kcal/mol for transition metal species. We did not want a method that relied upon parameterisation based upon experimental data, as we were concerned that this may not effectively capture much of the unique chemistry that can occur for the transition metals and heavier elements.

Why are you also investigating the scission of lignin linkage models via a transition metal catalyst?

Chemical problems drive our methodology development. Presently, we are considering many problems of importance to green chemistry, from studies of lignin to our extensive work on CO₂ reduction, utilisation, and sequestration. Lignin is the second most common organic matter on Earth, surpassed only by cellulose, and is often a highly under-utilised component of plant matter, since it is usually burned to heat the reactors. However, lignin can be used to derive several useful, value-added products such as methanol, organic acids, benzene and vanillin. While there are various processes for producing useful chemicals on the industrial scale from cellulose, economically viable means of using lignin as a starting material for useful chemicals has lagged. Also, the selectivity for a specific product has generally been very limited. Our studies are providing molecular level insight about potentially useful catalytic options. Beyond this, our initial studies have provided theoretical insight about the utility of our computational models, and have illustrated the potential utility of our models and methods for more complex catalysts and lignin models.

What value has your team and students brought to this research?

I certainly want to acknowledge my research team, my postdoctoral fellows, graduate students, undergraduate students, and high school students. They have all been an absolutely critical component of my research programme. Their efforts, creativity, drive, and teamwork have enabled us to pursue a wide range of interesting research from the development of computational methodologies, to studies in green chemistry. One of these investigations was for CO₂ ‘formatics’, which has been a particularly fruitful line of investigation. We have also been involved in areas such as highly energetic materials, computer-based drug design, mechanical properties of materials, and many other areas.

Dr Angela Wilson explains her work on the periodic table, in which she is trying to produce a basis for ever more accurate computational chemistry by utilising thermodynamics, thereby building complete solutions from the basic laws of science.
Researchers at the University of North Texas is using computational models to push forward our understanding of chemistry, but this science is producing its own challenges, requiring the team’s innovation to overcome them.

**NEW INSIGHTS**

The group has used these new computational methods to help with research in various areas, such as to identify novel highly energetic nitrogen-containing compounds that should be considered in next generation explosives, to suggest new design for carbon capture and carbon reduction techniques, to understand bond activation in biomass species, and to propose where new experiments may be needed.

This work is extremely important, demonstrating that the team is not only using their work as a tool for discovery, but also uncovering elements of the computer science behind their computations. Furthermore, their modelling is facilitating discoveries in chemistry which could not have been produced without the powerful approach that has been developed. Dr Angela Wilson is leading the group and is pleased with these results, as she explains: “Development and discovery that aids not only our group, but also other researchers around the world has been the most rewarding element of our work”. Their research has been translated into global innovations, and this has in turn led to scientific discoveries. Being part of these ongoing developments has proved exciting, and their continued research developing computational software is set to keep pushing forward innovative work, providing the computational gains needed for further experimental breakthroughs.

**OPPORTUNITIES AHEAD**

These successes have been achieved despite significant challenges facing both the team in Texas, as well as computational scientists.
This study has provided a rich, integrated research and education environment, and has aided in drawing very strong doctoral, undergraduate, and high school students, who have been critical assets to research efforts around the world. Such issues, which unify computational groups of researchers, also comprise a number of opportunities within the discipline. The first of these is practical and concerns hardware. Although the need for increasingly powerful computation may seem to be the major problem, there is actually a number of issues beyond this. Increasingly, high complexity parallel computing, including memory and processing divisions, means that computational programming could have a more difficult task of utilising the new hardware that becomes available. Furthermore, when the hardware is used to its potential, the amount of data produced requires new techniques in data farming, as well as ways of forging paths through intelligent searches or other techniques. Although there are challenges, there are many opportunities ahead, particularly as researchers learn of the potential of computing resources and demand for different models increases.


**TECHNOLOGY**

**OBJECTIVES**

To develop methods for the calculation of thermochemical and bonding properties (e.g. enthalpies of formation and reaction, ionisation potentials, electron affinities, proton affinities, bond dissociation energies, reaction barriers, excitation energies) of main group and transition metal species to near chemical accuracy, i.e. to within one kcal/mol or less for main group species, within three kcal/mol or less for enthalpies of transition metal species. Correct predictions of these properties is core to the understanding of chemical reactivity, from the design of new catalysts, molecules and materials.

**KEY COLLABORATORS**

Dr Wibe de Jong, Pacific Northwest National Laboratory

Dr Thomas R Cundari, University of North Texas

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**CONTACT**

Dr Angela K Wilson
Principal Investigator
University of North Texas
Department of Chemistry
1155 Union Circle, #305070
Denton
Texas 76203-5017
USA

T +1 940 565 4296
E akwilson@unt.edu

**ANGELA WILSON** earned her PhD in Chemical Physics from the University of Minnesota and was a postdoctoral fellow at Pacific Northwest National Laboratory. She is now Regents Professor of Chemistry and Director for the Center for Advanced Scientific Computing and Modelling at the University of North Texas. She is a Fellow of the American Chemical Society and National Associate of the US National Academies.

**TRANSITIONAL RESEARCH**

This research moved beyond the traditional role of ab initio composite approaches, which merely predict the ground state energies of main group species, to a whole new approach. Wilson realises that there are a variety of possibilities opened up by their work: “Our composite strategies can be used not only for ground states, but also in addressing transition and excited states, transition metals, species that require multi-reference treatment and entire potential energy surfaces”. By possessing the capability to deal with multiple close-lying electronic configurations, as well as large numbers of other chemical problems, the net effect is one of increased applicability. Consequently, the team is in the position to provide prediction of not only qualitative trends, but also providing quantitative energetic data with high degrees of accuracy. This means that the scope of chemical problems that are being addressed is far wider than has ever previously been possible with composite schemes, making their work ever important in the ongoing development and application of computational chemistry.

**IMPORTANT PLACEMENT**

These advances have been possible thanks to the dedication and support of the University of North Texas, who over the past decade have built one of the largest computational chemistry programmes in the US. By investing in high performance computing, the university has been able to facilitate an extremely strong programme of investigation. The research has also been furthered by two Chemistry Research Instrumentation Facilities grants, which have enabled the creation of a computational chemistry high performance computing facility. The group has also benefited from NSF-supported computing facilities, which are needed for very large calculations. Wilson is pleased with what the group has been able to achieve as a result: “This study has provided a rich, integrated research and education environment, and has aided in drawing very strong doctoral, undergraduate, and high school students, who have been critical assets to research efforts”. The researchers are excited about the work that lies ahead and the hope of producing pioneering computational results that will have important application for their field and research in general.