SCIENCES & BIOENGINEERING SCIENCES

The Research Group

General Chemistry

has the honor to invite you to the public defense of the PhD thesis of

Tom Bettens

to obtain the degree of Doctor of Sciences

Title of the PhD thesis:

Quantum Chemical Modeling of Mechanochemical Reactivity: Insights from Conceptual DFT and Modified Potential Energy Surfaces

Promotors:
Prof. dr. Frank De Proft
Prof. dr. Mercedes Alonso
Co-promotor:
Em. Prof. Dr. Paul Geerlings

The defense will take place on
Monday, December 13, 2021 at 17h30

The defense can be followed through a live stream. Contact tom.bettens@vub.be for more information.

Members of the jury

Prof. dr. Ulrich Hennecke (VUB, chair)
Prof. Dr. Ir. Freija De Vleeschouwer (VUB, secretary)
Prof. Dr. Ir. Wim Versées (VUB)
Prof. Dr. Andreas Dreuw (Ruprecht-Karls-Universität)
Prof. Dr. Célia Fonseca Guerra (Vrije Universiteit Amsterdam)
Prof. Dr. Christophe Morell (Université de Lyon)

Curriculum vitae

Tom Bettens obtained the degree of Master of Science in Chemistry in 2017 before joining the ALGC group pursuing a PhD in computational chemistry.

His PhD was devoted to the understanding of the mechanical activation of molecular system to design new mechanochemical experiments. This research and several side activities resulted in a total of seven peer-reviewed scientific articles and a book chapter.

Abstract of the PhD research

In chemistry, mechanical force is typically applied to a chemical system through ball-milling or sonication experiments. Such experiments activate the bulk system and the true activation of a chemical process owing to the direct absorption of mechanical energy by the reagents in a chemical reaction is overshadowed by the complex interplay of simultaneous processes, surface effects and local heating among others. Recent pioneering force microscopy experiments have proven the possibility to trigger pure mechanical deformations of individual molecules and study unique mechanochemical reactions. Reliable approaches for calculating force-modified molecular geometries are wonderful tools to gain mechanistic insight into force-induced reactions. But little attention has been devoted to a true understanding of mechanical activation and the associated change in the chemistry and electronic properties of molecules. Prior to this thesis, very few tools were available for the identification of these chemical reactions that can be triggered by mechanical activation; faithful to tradition, chemical intuition and trial and error are often trusted instruments.

In this thesis, two approaches were followed to understand the mechanochemical reactivity of individual molecules. In a first approach, the reactivity of chemical bonds and angles was studied by several reactivity indices upon the introduction of bond length alternations and angular distortions, respectively. In a second approach, the mechanical alterations of molecular potential energy surfaces were investigated. We demonstrated that the directional character of mechanical force can be used to trigger different reaction pathways. In other words, a molecule responds according to the structural distortion owing to the direction of the mechanical force. Via a systematic approach, structural distortions that trigger a variety of chemical reactions were anticipated and validated through quantum chemical calculations: ring openings, topological interconversions, and bimolecular reactions. As the range of applications of molecular mechanochemistry continues to be explored, two new tools based on quantum chemical foundations are now available for identifying new mechanochemical reactions.