Lay summary

Developing new methods in computational chemistry is an important area of research in STEAM fields. Theoretical models are implemented into computer codes and then executed on supercomputers. By doing so, scientists are able to solve many problems in (general) chemistry, even modeling chemical compounds that are difficult to handle experimentally due to their instability, toxicity or radioactivity. The main objective quantum chemistry is to properly describe the correlated motion of electrons. Specifically, a crucial point is to propose a theoretical approach that will provide a reliable description of the investigated problem with reasonable computational cost. Most methods are computationally very expensive and their computational cost scales exponentially with the size of the system, a fact that is known as the curse of dimension. Hence, their applicability is strongly limited. In the following project, the applicant proposes to develop new methods that will be a hybrid of existing geminal-based approaches with Configuration-Interaction theory. Geminal methods are based on two-electron functions, which are the fundamental building blocks of the (approximated) electronic wavefunction. Numerical results show that these approaches can accurately model the strong part of the correlated motion of the electrons, encountered, for instance, in molecules with stretched bonds as well as complexes or clusters containing actinide atoms. One of these methods is the Antisymmetric Product of 1-reference orbital Geminal (AP1roG). To make AP1roG applicable to challenging problems in chemistry, we have to extend it in a suitable way. The applicant proposes to develop AP1roG-CI-type methods that should accurately capture the strong and the missing weak correlations effects. These effects are due to the repulsive Coulomb interactions between the electrons. The devised hybrid methods will be both robust and inexpensive. Furthermore, in the proposed project, the applicant will benchmark the new methods against small-sized molecules (with a focus on main group chemistry). If successful, the applicant will apply the new methodology to heavy-element chemistry, in particular to actinide-containing compounds. All methods developed in this project will be included in the Piernik software packages, an open-source code free of charge for the general public.