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Aluminum-silicon nanoalloys: search for the most stable structures up to 13 atoms.

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Abstract: Clusters are aggregates of a countable number of particles, where a hetero-atomic cluster is constituted of two or more different species. Metal clusters may be constituted of a single metallic element or of more than one, giving rise to nanoalloys [1]. The study of hetero-atomic clusters opens a great channel to design new mixed materials, whose physicochemical properties can differ from those of their pure elemental counterparts [2]. Silicon based clusters possess great interest due to their potential applications in semiconductor and optoelectronic industries and aluminum-silicon compounds possess considerable significance in nanomaterials fields [2]. However, nowadays, there is a lack of studies of these nanoalloys [2]. The properties of alloy clusters may be "tunable" by changing the composition of a cluster of a given size, and to find the most stable structures there is a need to study all possible isomers that appear from atom permutations (homotops) [3,4]. This is a very difficult task, given that the number of local minima increases very fast with cluster size. The goal of this work is to find the most stable structure and composition of aluminum and silicon nanoalloys, using computational calculations. For this purpose, we consider nanoalloys that possess between three and thirteen atoms, and all possible composition of aluminum and silicon. In each case, all possible isomers are considered, permuting the position of all the atoms that constitute each cluster. There are a lot of possible permutations, and these lead to thousands of structures to be analyzed. The initial coordinates are taken from the singly aluminum doped silicon neutral clusters obtained by Tam *et al* [2], and from these coordinates the structures are optimized using the Møller-Plesset (MP) Perturbation Theory methodology. The MP2 calculations consume a considerable amount of time and in this work, the basis set chosen is the LANL2DZ with an Effective Core Potential (ECP) [5], which makes the calculations of all homotops faster. In order to generate all homotops, we developed an algorithm that reads the skeleton of the molecule from Cartesian coordinates inserted in a separated file and select the desired spin multiplicity. After performing these simple steps, the program generates a GAMESS input for all possible homotops to be locally optimized. Vibrational frequencies are also calculated for the optimized structures. At the same time, when executed, the program presents the nuclear repulsion energy of each structure and search for similar values. This is useful to compare the generated inputs and verify if they represent the same structure, avoiding a repeated calculation. If the energies differ only in the last decimal places, the program accuses a possible equivalence of the structures. After performing all calculations, the optimized energies are analyzed and the lowest energy minimum is selected, therefore, the most stable structure with that specific composition is determined. Average binding

energies and excess energies of each composition and cluster size are calculated. After performing these calculations, for selected compositions, the homotops will be re-optimized using a more sophisticated method and basis set. This will enable a comparison between the methods, and verify if the most stable structure obtained with the MP2/ECP method is the most stable structure obtained by a more refined method. All electronic structure calculations are carried out using the GAMESS 2016 program package.

Key-words: Nanoclusters; electronic structure; optimization.

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References:

[1] Roy L. Johnston, "Atomic and Molecular Clusters" (2002), Taylor & Francis, London and New York.
[2] Several authors, quoted by Nguyen Minh Tam, Truong Ba Tai, Vu Thi Ngan, and Minh Tho Nguyen, " Structure, Thermochemical Properties, and Growth Sequence of Aluminum-Doped Silicon Clusters Si_nAl_m (n = 1 – 11, m = 1-2) and their anions" (2013), The Journal of Physical Chemistry, ACS Publications.
[3] Mateus A. M. Paiva, Barbara M. T. C. Peluzo, Jadson C. Belchior and Breno R. L. Galvão, "Structure and Stability of neutral Al-Mg nanoclusters up to 55 atoms" (2016), Phys. Chem. Chem. Phys, Royal Society of Chemistry.
[4] J. Jellinek and E. B. Krissinel, Chem. Phys. Lett., 258,283 (1996).
[5] LANL2DZ ECP Basis set, available in: <https://bse.pnl.gov/bse/portal/user/anon/panel/Main/template/courier_content/js_peid/11535052407933> Accessed on 21/07/2017.