



Contribution ID: 204

Type: Verbal

The Pourbaix diagram of astatine in aqueous medium

Astatine (At), element 85, is below iodine in the periodic table of elements. One of its isotopes, ^{211}At , is a promising candidate as a therapeutic agent in nuclear medicine [1]. Although it is clear that much of the chemistry described for halogens is applicable to astatine, a more metallic character is expected as compared to its nearest halogen neighbor, iodine. However, At chemistry in aqueous solution remains poorly understood. There are no stable isotope of astatine, its longest-lived isotope having a half-life of 8.3 h. All investigations are thus derived from radiochemical studies at ultra-trace concentration, typically between 10⁻¹² and 10⁻¹⁵ mol/L and no spectroscopic tools can be used to identify unambiguously the formed species. The chemical forms of astatine are usually deduced from its behavior in given conditions with respect to the behavior of expected model compounds.

In this work, a combined experimental and theoretical approach is used to define the potential/pH diagram of astatine (Pourbaix diagram) in non complexing medium with the aim of answering the two main questions raised in the literature: does At(0) exist in aqueous solution and what is the chemical form of At(III), if it exists? The experimental methodology considers that a given species is characterized by its distribution coefficient (D) experimentally determined in a biphasic system. The change in speciation arising from a change in experimental conditions is observed by a change in D value [2]. Unlike most of previous studies, we present a quantitative analysis of the experimental data based on equilibrium reactions, to identify the species formed and derive the thermodynamic parameters. The theoretical methodology is based on quasi-relativistic quantum chemistry computations and solvation free energy calculations using polarizable continuum models. The results show that At at the oxidation state 0 cannot exist in aqueous solution. The three oxidation states present in the range of water stability are At(-I), At(I) and At(III) and exist as At⁻, At⁺ and AtO⁺, respectively, in the 1 to 2 pH range [3]. When the pH increases, AtO⁺ reacts with water to form two hydrolysis species, AtO(OH) and AtO(OH)₂⁻.

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[2] J. Champion, C. Alliot, S. Huclier, D. Deniaud, Z. Asfari and G. Montavon, *Inorg. Chim. Acta*, 362 issue 8, 2654 (2009)

[3] J. Champion, C. Alliot, E. Renault, B.M. Mokili, M. Chérel, N. Galland and G. Montavon, submitted to *J. Chem. Phys. A*.

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Track Classification: Separation Methods, Speciation