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Evaluation of Adsorption Enthalpies in Isothermal Chromatography Experiments under non-optimal Conditions - the case of Tl and TlOH on fused silica

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The determination of the enthalpy of adsorption of an analyte on a defined stationary surface is one of the most important methods for the determination of chemical properties of superheavy elements (SHEs). Isothermal gas-adsorption chromatography setups have been used in various experiments to chemically study compounds of rutherfordium (Rf, Z=104), dubnium (Db, Z=105), seaborgium (Sg, Z=106), and bohrium (Bh, Z=107). In all these experiments, Monte Carlo simulations were used to derive the adsorption enthalpy for the investigated chemical system from the experimental data. This approach allows for predicting the breakthrough yield of a chemical species on a specific stationary phase as a function of the applied fixed experimental parameters e.g. gas flow and temperature profile. Including the decay (half-life) of the used radioisotope, leaves the enthalpy of adsorption as the only free parameter in the model. If the measurements can be performed over a sufficiently wide temperature range, saturation of the breakthrough yields at about 100% can be achieved. Therefore, data evaluation based on relative chemical yields of the external chromatograms was introduced in the past. These relative chemical yields were calculated with respect to a reference measurement at sufficiently high temperature, assumed to represent the 100% breakthrough yield. The adsorption enthalpy was then deduced by determining the Monte Carlo simulation that reproduced the experimentally observed external chromatogram best.
Recently, in preparation for experiments with nihonium (Nh, Z=113), model studies were performed with its lighter homolog thallium, using both isothermal vacuum as well as isothermal gas-adsorption chromatography. It became evident that it was difficult to reliably identify the saturation level of the external chromatogram for the species studied due to experimental limitations regarding the maximum temperatures, isothermal temperature profile stability as well as the statistics of the detected events. All of these limitations are clearly due to non-optimal experimental conditions and make the use of established data analysis questionable. This has motivated the development of an algorithm to predict the breakthrough yield independently of a reference measurement, which can be used in connection with Monte-Carlo simulation to estimate the enthalpy of adsorption.
Here, we will present this evaluation algorithm and its application to isothermal chromatography experiments using the example of past experiments performed at the JAEA tandem accelerator in Tokai-mura (Japan) and at the JINR U-400 Cyclotron in Dubna (Russia).

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