Distribution ratio prediction of group 4 elements using machine learning, toward chemical study of element 104, rutherfordium

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Superheavy elements (SHEs) with $Z > 100$ are synthesized via heavy-ion-induced nuclear reactions. SHEs are expected to exhibit chemical properties that differ from the lighter homologous elements in the periodic table due to the influence of relativistic effects. It is important to understand chemical properties of SHEs. However, the methods and reagents that were used in the studies are limited due to the complexity of the chemical experiment of SHEs so chemical studies have been scarce.[1] Considering complex formation of SHEs, ligands have conventionally been chosen based on their bonding and electronic properties. This approach is empirical and considers only a few parameters, resulting in a limited selection of ligands, such as inorganic and organic ligands which have simple structures. In this work, we focused on machine learning, which nonempirically considers many factors. It is expected that effective ligands that have been overlooked might be found by using machine learning. This report presents the predictions of the distribution ratio (D) in a solvent extraction produced by the developed machine learning model. These predictions were compared with the distribution coefficient (K_d) obtained from actual solid-liquid extraction experiments using Zr and Hf to evaluate their validity.

A model was constructed to design the appropriate ligand for the solvent extraction of rutherfordium (Rf), $Z = 104$. It predicted the $\log D$ values of its homologous elements, Ti, Zr, and Hf, and the elements that accept tetravalent cations in aqueous solutions, Pb, Th, and Pu. The experimental data used for training were obtained from SEDATA,[2] a database specializing in solvent extraction. These experimental data were converted into input data for machine learning following the methods in a prior study,[3] in which the $\log D$ of lanthanides in solvent extraction is predicted. Then, a fully connected neural network (FCNN) model was trained using these data. The $\log D$ predicted using the validation data was similar to the experimental values with an R^2 and RMSE of 0.59 and 0.77.

We conducted solid-liquid extraction using ^{88}Zr and ^{175}Hf tracers which were generated using the AVF ring cyclotron at the RIKEN Nishina center and we compared the obtained K_d values and model-predicted $\log D$ value. In this experiment, the tracer solutions with nitric acid 0.13, 0.22, and 0.53 mol L^{-1} were used. Resin and tracers were mixed and shaken for 60 min using a vortex shaker in the extraction with DGA resin, which is impregnated with N, N, N', N'-tetraoctyldigrycolamide (TODGA). The K_d values were calculated as

$$K_d = (A_{STD} - A_s)V/(A_s w_r)$$

where A_{STD} and A_s are radio-activities of ^{88}Zr and ^{175}Hf in the control solution and aqueous phase, respectively; V is the volume of the aqueous phase (mL); and w_r is the mass of the dry resin (g).

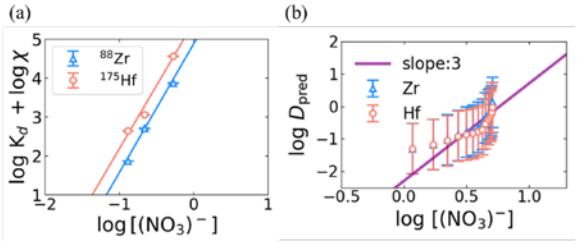

Fig. 1. (a) Experimentally scaled K_d values of ^{88}Zr and ^{175}Hf. (b) D values of Zr and Hf predicted using developed machine learning model.

Figure 1(a) shows the K_d values scaled by χ:

$$\chi = 1 + \sum_{i=1}^{4} \beta_i [(NO_3)^-]^i$$

where β_i is the overall formation constant, which was calculated as $\beta_i = \beta_i = [ML_i]/([M]\,[L]^i)$. The slopes of the line fit 3.31 ± 0.18 for Zr and 3.25 ± 0.21 for Hf. The $\log D$ values of Zr and Hf in the TODGA extraction at different nitric acid concentrations were predicted, and the results are shown in Fig. 1(b). The tendency of the $\log D$ values for $[HNO_3]$, basically consistent with the experimental K_d values, indicated that this model can predict the dependence of acid concentration of the D values during solvent extraction.

In this study, we developed a machine learning model that predicts $\log D$ values of M^{4+} ions when influenced by an acid. We are planning to refine the machine learning model to increase predictive accuracy, search for ligands with different in the D values among Zr^{4+}, Hf^{4+} and Th^{4+} for an experiment of Rf. We also aimed to determine the factors that cause the difference, by the analysis of our machine learning model.

References

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