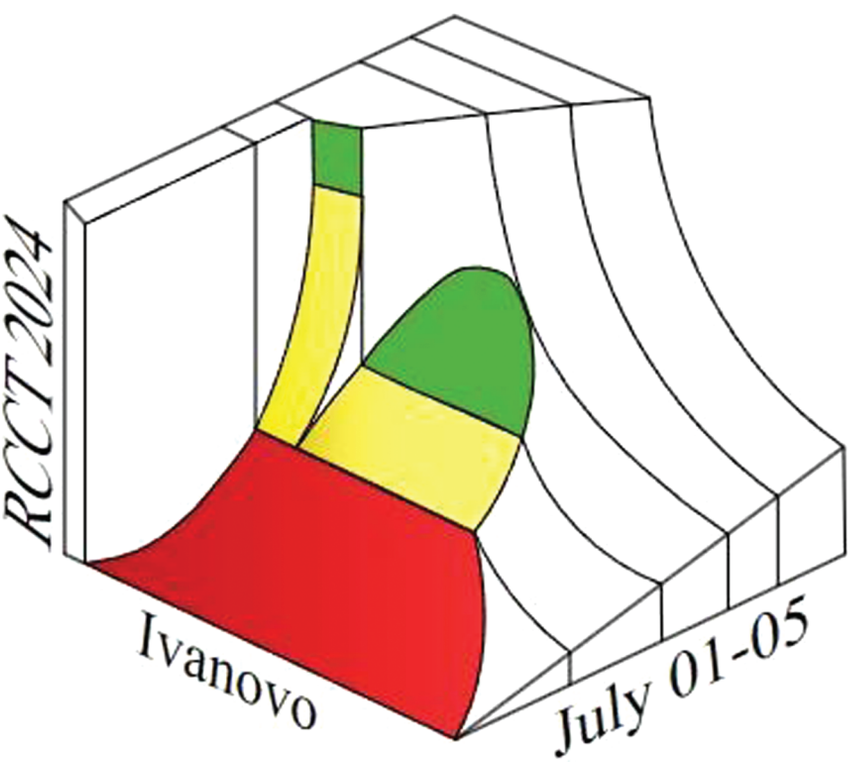
XXIV International Conference on Chemical Thermodynamics in Russia

July 1-5, 2024
Ivanovo, Russia



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BOOK OF ABSTRACTS

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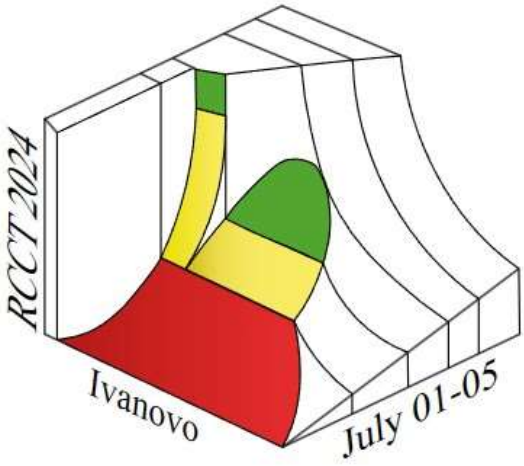

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Russian Academy of Sciences

Mendeleev Russian Chemical Society

Scientific Council for Physical Chemistry of the Russian Academy of Sciences

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THERMODYNAMIC PROPERTIES IN THE Cs_2O-SrO-Al_2O_3-SiO_2 SYSTEM STUDIED BY THE KNUDSEN EFFUSION MASS SPECTROMETRIC METHOD

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Vaporization of cesium and strontium compounds from the core material during severe accidents at nuclear power stations may lead to hazardous radioactive pollution of the environment [1]. For better understanding of high temperature behavior of multicomponent core material, the model Cs_2O-SrO-Al_2O_3-SiO_2 system was considered in the present study since alumino-silicate matrices were mentioned as potential candidates for reliable waste forms for cesium and strontium [2].

In the present study, the vaporization processes and thermodynamic properties of the Cs_2O-SrO-Al_2O_3-SiO_2 system were studied by the Knudsen effusion mass spectrometric (KEMS) method [3]. The samples in the system under consideration were synthesized by the sol-gel method based on Cs_2CO_3, $SrCO_3$, $Al(NO_3)_3 \bullet 9H_2O$, and tetraethoxysilane $(C_2H_5O)_4Si$ as initial components involving nitric acid with the maximum annealing temperature of 1073 K for 622 hours. The samples obtained were characterized by X-ray phase analysis, electron probe microanalysis, differential thermal analysis, and X-ray fluorescence analysis. Identification of the structure as well as phase and chemical compositions of the samples of the Cs_2O-SrO-Al_2O_3-SiO_2 system allowed the KEMS study to be carried out using an MS-1301 mass spectrometer at an ionizing voltage of 30 V from a single tungsten effusion cell.

The vaporization of the samples in the Cs_2O-SrO-Al_2O_3-SiO_2 system was observed at temperatures above 900 K. The main vapor species over the samples were Cs and O_2, leading to selective removal of Cs_2O from the samples. The vaporization of the samples without Cs_2O was found at temperatures above 1800 K, with the main vapor species being Sr, SiO, and O_2. The temperature dependences of the partial pressures of the vapor species over the samples under study were obtained by the KEMS method. These data enabled the partial vaporization enthalpies of Cs_2O as well as mass losses of the samples to be determined. The data obtained were also used to derive the temperature dependences of the component activities and calculate partial molar enthalpies of mixing in the Cs_2O-SrO-Al_2O_3-SiO_2 system. Negative deviations from the ideal behavior were established in the system under study at the temperatures 1000, 1100, and 1200 K. It was shown that introduction of SrO in the Cs_2O-Al_2O_3-SiO_2 system led to decrease in the Cs_2O activity coefficients. As for the influence of Al_2O_3 and SiO_2, the minimum values of the Cs_2O activity coefficients were observed in the samples with the equimolar content of Al_2O_3 and SiO_2, showing the concentration ranges with the optimal thermal stability.

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The financial support of the Ministry of Science and Higher Education of the Russian Federation (project No. 075-15-2021-1383) is gratefully acknowledged. We also express the gratitude to A.V. Fedorova and A.A. Selyutin for the sample synthesis and identification.

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XXIV Международная конференция
по химической термодинамике в России

Тезисы докладов опубликованы в авторской редакции

ISBN 978-5-6051371-2-2

Подписано в печать 05.06.2024.
Формат 60×84 1/8. Бумага офсетная №1, пл. 80 г/кв.м.
Печать плоская. Печ. л. 46,63. Усл. печ. л. 43,36.
Тираж 30 экз. Заказ № 26127.
Отпечатано в АО «Ивановский издательский дом».
153000, г. Иваново, ул. Степанова, 5. Тел. (4932)301411, 303237
e-mail: 301411@rambler.ru

ISBN 978-5-6051371-2-2