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Caustic-Side Solvent Extraction Chemical and Physical Properties: Equilibrium Modeling of Distribution Behavior

December 2001

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**CAUSTIC-SIDE SOLVENT EXTRACTION
CHEMICAL AND PHYSICAL PROPERTIES:
EQUILIBRIUM MODELING OF DISTRIBUTION BEHAVIOR**

Lætitia H. Delmau, Tamara J. Haverlock, Tatiana G. Levitskaia,
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ABSTRACT

A multivariate mathematical model describing the extraction of cesium from different mixtures of sodium hydroxide, sodium nitrate, sodium chloride, and sodium nitrite containing potassium at variable concentrations has been established. It was determined based on the cesium, potassium, and sodium distribution ratios obtained with simple systems containing single salts. These experimental data were modeled to obtain the formation constants of complexes formed in the organic phase based on specified concentrations of components in both organic and aqueous phases. The model was applied to five different SRS waste simulants, and the corresponding cesium extraction results were predicted satisfactorily, thus validating the model.

1. INTRODUCTION

The solvent extraction process proposed and considered for cesium removal from the waste present at the Savannah River Site (SRS) is being investigated with respect to the behavior of system components under different conditions. A thorough understanding of the process is in part demonstrable by establishing a model that predicts the extraction of cesium based on the major components of the waste (or simulant). The ability to predict distribution behavior facilitates appropriate flowsheet design to accommodate changing feed composition and temperature. It also provides greater confidence in the robustness of the process overall. Finally, given the knowledge of the composition of any particular feed, a reliable model yields an immediate estimate of expected flowsheet performance for comparison with process data. The scope of this modeling study was directed toward predicting the cesium distribution ratios obtained with five different SRS simulants corresponding to five real-waste tanks. Chemical analyses of the tanks provided the concentrations of sodium, potassium, cesium, nitrate, and free hydroxide.¹ When preparing the simulants, the total concentration of cations could be as high as 5.6 M. The nitrate and hydroxide concentrations measured in the tanks could not balance the cation concentration. The quantity of anion still not accounted for by these analyses was filled either with chloride or with nitrite anions. Based on the total composition of the SRS waste, these four anions and three cations were determined to be the main components. The model will include species of these ions, and corresponding formation constants will be determined by the sequential modeling of simple systems containing first one cation and one anion at the same time, then systematically increasing the number of components. A model representing the extraction of cesium from different media will then be established and cesium extraction behavior could be predicted by a simple input of the concentrations in the aqueous phase before extraction.

2. EXPERIMENTAL PROGRAM

2.1 MATERIALS

Stock solutions of HNO_3 , NaNO_3 , NaNO_2 , NaCl , and NaOH were prepared and all other concentrations prepared as a dilution of the stock. Sodium hydroxide was diluted from 50% wt/wt received from J. T. Baker, Lot J17045. All salts were dried at $110\text{ }^\circ\text{C}$ for >18 hours and stored in a desiccator prior to solution preparation. Sodium chloride was received from EM Scientific, Lot 33131325; NaNO_2 was received as 99.5% from Aldrich, Lot 07012MS; NaNO_3 was received from J. T. Baker as reagent grade crystal, Lot M14156. Cesium nitrate was received from Alpha Aesar, 99.9% and dried prior to use. Potassium nitrate was received from EM Science. Sodium concentrations prepared were 5.6, 4.5, 2.25, 1.00, 0.50, 0.10, and 0.01 M. CsNO_3 and KNO_3 were added at 0.5 mM and 60 mM, respectively, directly to the sodium salts, effecting a slight dilution of the initial sodium in solution. Binary salt solutions at anion ratios of 0, 0.25, 0.50, 0.75, 0.90, and 1.0 and total sodium concentration of 4.5 or 5.6 M were also prepared with CsNO_3 added at 0.5 mM as well as with and without KNO_3 at 60 mM. Potassium extractions from KNO_3 solutions at concentrations of 1.0, 0.30, 0.10, 0.01 M were also performed. Measurements of cesium extraction from nitric acid involved pristine solvent that had not been preequilibrated with the corresponding solution of nitric acid without cesium. The organic phase consisted of washed solvent Cs-7SB/ Isopar® L, ORNL Lot# PVB-B000718-156W (7-28-2000). The radiotracers ^{22}Na and ^{137}Cs were obtained from Isotope Products, Burbank, CA.

2.2 GENERAL CONTACTING AND COUNTING PROCEDURE

The capped vials were mounted by clips on a disk that was rotated in a constant-temperature air box at $25.0 \pm 0.5\text{ }^\circ\text{C}$ for 90 minutes. After the contacting period, the vials were centrifuged for 3 minutes at 3600 RPM and $25\text{ }^\circ\text{C}$ in a Sanyo MSE Mistral 2000R temperature-controlled centrifuge. A 300 μL aliquot of each phase was subsampled and counted using a Packard Cobra II Auto-Gamma counter. All samples were counted for a period of 10 minutes using a window of 580-750 keV.

2.3 VARIABLE TEMPERATURE EXPERIMENT

A series of experiments to determine cesium distribution using ^{137}Cs tracer techniques was completed. The distribution of cesium in response to increasing concentrations of NaOH and NaNO_3 at two temperatures, $20\text{ }^\circ\text{C}$ and $35\text{ }^\circ\text{C}$, was examined. Contacting experiments were carried out using an

O/A of unity. All contacts were performed in duplicate. The aqueous phase consisted of 0.5 mM CsNO₃ and varied concentrations of either NaOH or NaNO₃ at 0.01, 0.1, 0.5, 1.0, 2.25, 4.5, and 5.6 M. ¹³⁷Cs tracer was introduced at 0.1 μCi/mL aqueous phase. The series of 0.5 mM CsNO₃ in NaOH or NaNO₃ solutions were prepared by aqueous dilutions. The contacts were carried out for a period of 90 minutes in 5.0 mL VWR Cat. No. 66008-400 clear polypropylene vials.

The capped vials were mounted by clips on a disk which was rotated in a constant-temperature air box at 20.2 ± 0.5 °C for 90 min or on a similar wheel located in a LabLine Imperial III Model 306M Incubator at 35.8 °C for the same period of time. After the 90 minute contacting period, the vials were centrifuged for 3 minutes at 3600 RPM and 22 °C in a Sanyo MSE Mistral 2000R temperature-controlled centrifuge. A 300 μL aliquot of each phase was subsampled and counted.

2.4 EXPERIMENTS WITH CALIXARENE-FREE SOLVENT

In this experiment cesium extraction as a function of the solvent/modifier/trioctylamine (TOA) system was investigated. A contacting experiment was carried out, using an O/A volume ratio of unity, in which 1 mL of washed calix-free CSSX solvent was contacted at 25 °C with 1 mL of an aqueous phase consisting of 0.5 mM CsNO₃ and variable concentrations of NaOH (0.01, 0.1, 0.5, 1.0, 2.25, 4.5, and 5.6 M).

The calix-free organic phase was prepared by adding Cs-7SB modifier (Lot no. B000718-24DM) at 0.5 M, and trioctylamine (Lot no. B000718-105L) at 0.001M to Isopar® L (Lot no. 0306-10967). This solvent was then washed in Teflon® FEP labware using an O/A volume ratio of unity, twice with 0.1 M NaOH and 50 mM HNO₃, and three times with DDI water. The aqueous phases were made by appropriately diluting a 5.6 M working stock of NaOH and a 50 mM solution of CsNO₃. ¹³⁷Cs tracer was introduced at 0.1 μCi/mL aqueous phase. The contacts were carried out for a period of 90 minutes in 5.0 mL VWR Cat. No. 66008-400 clear polypropylene vials.

2.5 ION-CHROMATOGRAPHY EXPERIMENTS

The solvent (Lot B000718-156W) was contacted with an equal volume of the appropriate salt solution in 2 mL polypropylene vials for 1 hour by rotation in a thermostated air box set at 25 ± 0.1 °C. All samples were centrifuged for 3 minutes at 3500 rpm to confirm complete phase disengagement. The organic phase was then contacted with a five to ten-fold volume of dilute HNO₃ (1 mM) to strip the metal ions into the aqueous phase. Results were based on the first strip since the metal recovery was equal or greater than 98%. The strip solutions were analyzed with a Dionex Model DX500 equipped with a GP40

pump and a CD20 conductivity detector. The cations sodium, potassium and cesium were separated and analyzed using a CS12A analytical column coupled with a CG12A guard column. The analysis used 20 mM H₂SO₄ eluent at 1 mL/min in an isocratic run of 20 minutes. Background conductivity was 0.2 μS using CSRS-Ultra suppressor in auto-regeneration mode set at 300 mA. A five-level external standardization for each metal, Na, K, and Cs, was used. Duplicates were run for each sample and were analyzed with ± 2 % error.

3. PROGRAM SXFIT

3.1. DESCRIPTION OF THE PROGRAM

The program SXFIT is a program that can model thermodynamics data based on the constituents of the systems and the species that are being formed. Although the program's ability to model different kinds of systems is almost limitless, we will describe its capability to handle distribution ratios of ions, since our interest here is to be able to predict cesium extraction behavior. Like the preceding codes SXLSQ², SXLSQA³, and SXLSQI⁴, SXFIT⁵ is a program written in FORTRAN that refines a series of given inputs based on the least-squares minimization of the difference between the observed and the calculated quantities. The main improvement of SXFIT over predecessor codes is the fact that an unlimited number of constituents can be input. The program then calls for the parameters that are used to calculate the activity effects occurring in the aqueous phase (Masson and Pitzer coefficients) and the organic phase (Solubility parameters). In addition, molecular weights and non-aqueous molar volumes of the different constituents need to be provided along with the dielectric constant and the solubility parameter of the diluent in the organic phase. All initial concentrations of constituents are entered in a data file. Finally, based on the knowledge of the extraction reactions that occur during the process, a few reasonable species (products of the extraction system) may also be supplied with their formation constants. The program then calculates all concentrations of all constituents at equilibrium and the distribution coefficients of the ion of interest. Depending on the differences between the observed and calculated values, the program will then adjust the formation constants of the input species until the best fit is obtained. Of course, this could easily become a simple curve-fitting exercise in which a large number of parameters are used to fit a smaller number of data points. However, the user must ensure that all the species and their relative formation constants are chemically reasonable. Usually, the preference will be given to a model that contains the lowest number of species for a given goodness of fit, represented in the program by the agreement factor. A perfect fit with an accurate experimental error on all the data points yields an agreement factor of 1. A value greater than 1 indicates a poorer fit or an underestimate of the experimental error, while a value between 0 and 1 indicates an overestimate of the experimental error.

3.2 ASSUMPTIONS

The solvent used in this system comprises 0.01 M Calix[4]arene-bis(tert-octylbenzo crown-6) (BOBCalixC6), 0.5 M 1-(2,2,3,3-tetrafluoropropoxy)-3-(4-*sec*-butylphenoxy)-2-propanol (Cs7-SB modifier), and 0.001 M trioctylamine (TOA) in Isopar® L. The concentrations of the modifier and of TOA are held constant. The concentration of the modifier is large enough to neglect the amount that is being complexed during the extraction of the cations. Therefore, it will not appear in any species of the model. Regarding TOA, we chose not to include it in the model. Its only influence occurs when the aqueous phase contains enough acid to convert TOA into its acidic form, which in turn increases the amount of nitrate in the organic phase. In this work, only two sets of data involved nitric acid, and it was found that TOA did not have any influence on the cesium or potassium distribution ratios. In future modeling, this restriction will need to be lifted to properly account for acid balance in scrubbing and stripping. Likewise, an accurate accounting of volume and concentration changes would benefit from knowledge of water transferred to and from the solvent; this was omitted from the present treatment.

3.3 PARAMETERS USED

The program requires a series of input parameters, most of which are available in published handbooks and literature. Those parameters involved in the activity coefficients in the organic and aqueous phases can be refined by the program (Pitzer parameters, solubility parameters). However, for present purposes, the parameters were either calculated prior to any modeling or obtained from referenced sources and kept constant. The only parameters refined during the modeling process itself were the formation constants of the species in the organic phase.

Table 1. Molecular weights and non aqueous molar volumes of the constituents

Constituent	Molecular Weight (g/mol)	Non aqueous molar volume (cm ³ /mol)*
Na ⁺	22.990	10
K ⁺	39.098	9
Cs ⁺	132.91	21.5
H ⁺	1.008	0
NO ₃ ⁻	62.005	29
Cl ⁻	35.450	18
NO ₂ ⁻	46.006	26
OH ⁻	17.008	18
BOBCalixC6	1149.53	500
Diluent (Isopar® L)	170	227
Water		18

*The values for the ions are based on their aqueous molar volumes V_0 presented in Table 2. The value for sodium is a personal communication from Charles F. Baes, Jr.

The values for the constituents presented in Table 1 are those called by the program and changeable by the users. The molecular weight of water is 18.015 g/mol. This value is a constant and non-changeable.

Table 2. Masson coefficients⁶ of ions present in the system.

Constituent	V_0	S_v
Na ⁺	-1.5	1.89
K ⁺	8.73	1.10
Cs ⁺	21.40	1.29
H ⁺	0	0
NO ₃ ⁻	29.33	0.543
Cl ⁻	18.12	0.83
NO ₂ ⁻	26.5	2.00
OH ⁻	-1.04	2.32

Table 3. Pitzer parameters⁷ for the interactions between ions present in the system.

Interaction	β_0	β_1	C_ϕ
H ⁺ -NO ₃ ⁻	0.11190	0.36860	0.002470
Na ⁺ -NO ₃ ⁻	0.06800	0.01783	-0.000720
Na ⁺ -Cl ⁻	0.07650	0.26640	0.001270
Na ⁺ -NO ₂ ⁻	0.06410	0.10150	-0.004900
Na ⁺ -OH ⁻	0.08640	0.25300	0.004000
K ⁺ -NO ₃ ⁻	-0.08160	0.04940	0.006600
K ⁺ -Cl ⁻	0.04835	0.21220	-0.000840
K ⁺ -NO ₂ ⁻	0.01510	0.01500	0.000700
K ⁺ -OH ⁻	0.12980	0.32000	0.000410
Cs ⁺ -NO ₃ ⁻	-0.07580	-0.06690	0.000000
Cs ⁺ -Cl ⁻	0.03478	0.03974	-0.000496
Cs ⁺ -NO ₂ ⁻	0.04270	0.06000	-0.005100
Cs ⁺ -OH ⁻	0.15000	0.30000	0.000000

All β_2 values are set to 0. Parameter $\alpha_1 = 2$, and $\alpha_2 = 0$, since all the interactions are between two monocharged ions.

Regarding the activity coefficients in the organic phase, all the product species were assigned a similar value. The solubility parameter of the diluent (Isopar® L) and the extractant BOBCalixC6 were determined by group contribution calculations⁸. A best estimate of the values of the product species was made. They were kept constant as no reliable source for better values is available. In addition, the solubility parameters usually do not have a major impact on the determination of the product species formation constants. They avoid assuming ideality in the organic phase, but do not have a crucial effect on the final results, as the mole fraction of extracted species in the solvent is very small. The dielectric constant of the diluent equals 2.014; its solubility parameter is set to 18.40 J^{1/2}cm^{-3/2}. The solubility parameter of water is set to 51.13 J^{1/2}cm^{-3/2}.

The calixarene solubility parameter was estimated with the group contributions and determined to be 21 J^{1/2}cm^{-3/2}. All organic species formed in the organic phase were assigned a solubility parameter of 19.8 J^{1/2}cm^{-3/2}, which is also the solubility parameter of the modifier. Previous studies showed that at least one molecule of modifier was included in the complexes, and the solubility parameter is close enough to the value for the calixarene to avoid any major activity effect.

4. RESULTS AND DISCUSSION

Tables with all experimental results used in this modeling can be found in the appendix. By way of brief explanation, we include here a description of the approach chosen to find the best model. The method used to model the first set of data is discussed in detail. Fewer details will be given for subsequent groups, as the modeling technique and approach remain the same.

4.1. EXTRACTION MODELING FROM NITRATE MEDIA

The first step was to model data that involved only one anion. Indeed, all the other sodium salts were spiked with potassium nitrate and/or cesium nitrate, and the corresponding amount of anion, however small, was taken into account.

Data on cesium extraction from cesium nitrate and nitric acid yielded the formation constants of $\text{CsNO}_3\text{Calix(o)}$ and $(\text{CsNO}_3)_2\text{Calix(o)}$; the notation (o) refers to the organic phase. Addition of data on potassium extraction from potassium nitrate alone or mixed with cesium nitrate yielded the formation constant of $\text{KNO}_3\text{Calix(o)}$. Finally, addition of data on potassium and cesium extraction from sodium nitrate allowed us to calculate the formation constant of $\text{NaNO}_3\text{Calix(o)}$.

Table 4. Species and formation constants for the model derived for nitrate data

Species	Formation constant
$\text{CsNO}_3\text{Calix(o)}$	$\text{Log}_{10} K = 3.615$
$(\text{CsNO}_3)_2\text{Calix(o)}$	$\text{Log}_{10} K = 4.317$
$\text{KNO}_3\text{Calix(o)}$	$\text{Log}_{10} K = 1.387$
$\text{NaNO}_3\text{Calix(o)}$	$\text{Log}_{10} K = -0.943$

The fit of all the data is presented in Figures 1 and 2. The overall agreement factor is 2.7, with an assumption of a uniform 5% error on all the data points.

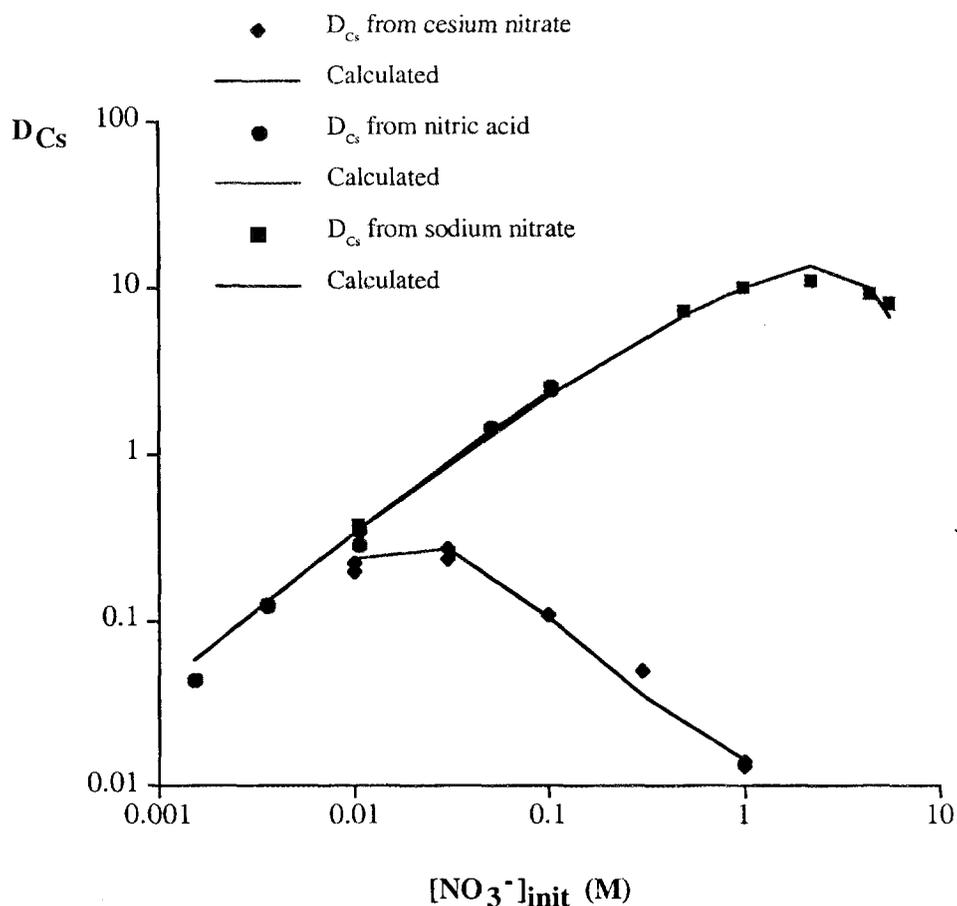
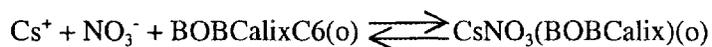


Fig. 1. Fit of cesium distribution ratios for nitrate media

A few comments need to be made regarding the dependence of cesium extraction with the nitrate concentration. A slope of 1 is expected when a complex involving an ion-paired cesium nitrate species is the major product formed in the organic phase.



This is well-demonstrated with the nitric acid experiment. The other two depart from the previous statement as the calixarene is loaded when cesium is extracted from increasing concentrations of cesium nitrate. A similar phenomenon is observed when reaching high concentrations of sodium nitrate where loading and activity effects give a trend that shows the cesium distribution ratios reach a maximum and then decrease.

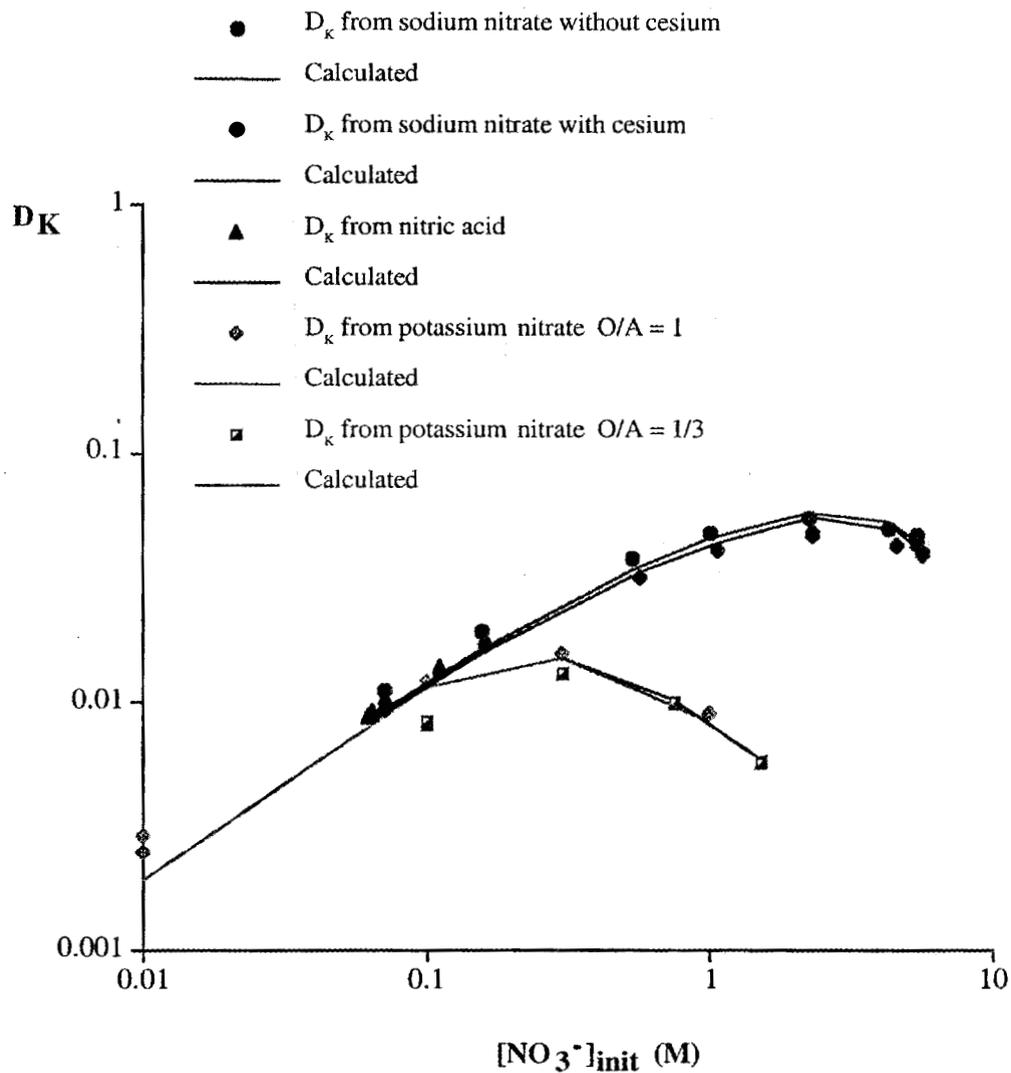


Fig. 2 . Fit of potassium distribution ratios for nitrate media

Similar conclusions can be drawn from the potassium extraction experiment. Potassium is extracted much less than cesium, but its initial concentration in sodium nitrate or nitric acid is about 100 times greater than cesium under similar conditions. The same trend of loading effects appears here, too.

4.2. EXTRACTION MODELING FROM HYDROXIDE MEDIA

The second set of data involves the fitting of cation extraction from sodium hydroxide. The results found previously for the nitrate system are required since all the potassium and the cesium were added as

spikes of nitrate solutions. Preliminary results on sodium extraction showed that the presence of calixarene was not required to extract this cation, as the amount of sodium present in the organic phase was the same whether or not the calixarene was present in the solvent.

The species listed Table 5 were added to the model to achieve a fit of data obtained in hydroxide systems. Figure 3 summarizes the fit as a function of hydroxide concentration.

Table 5. Species and formation constants for the model derived for hydroxide data

Species	Formation constant
CsOH(o)	$\text{Log}_{10} K = -2.264$
CsOHCalix(o)	$\text{Log}_{10} K = 3.332$
KOHCalix(o)	$\text{Log}_{10} K = 1.549$
NaOH(o)	$\text{Log}_{10} K = -0.565$

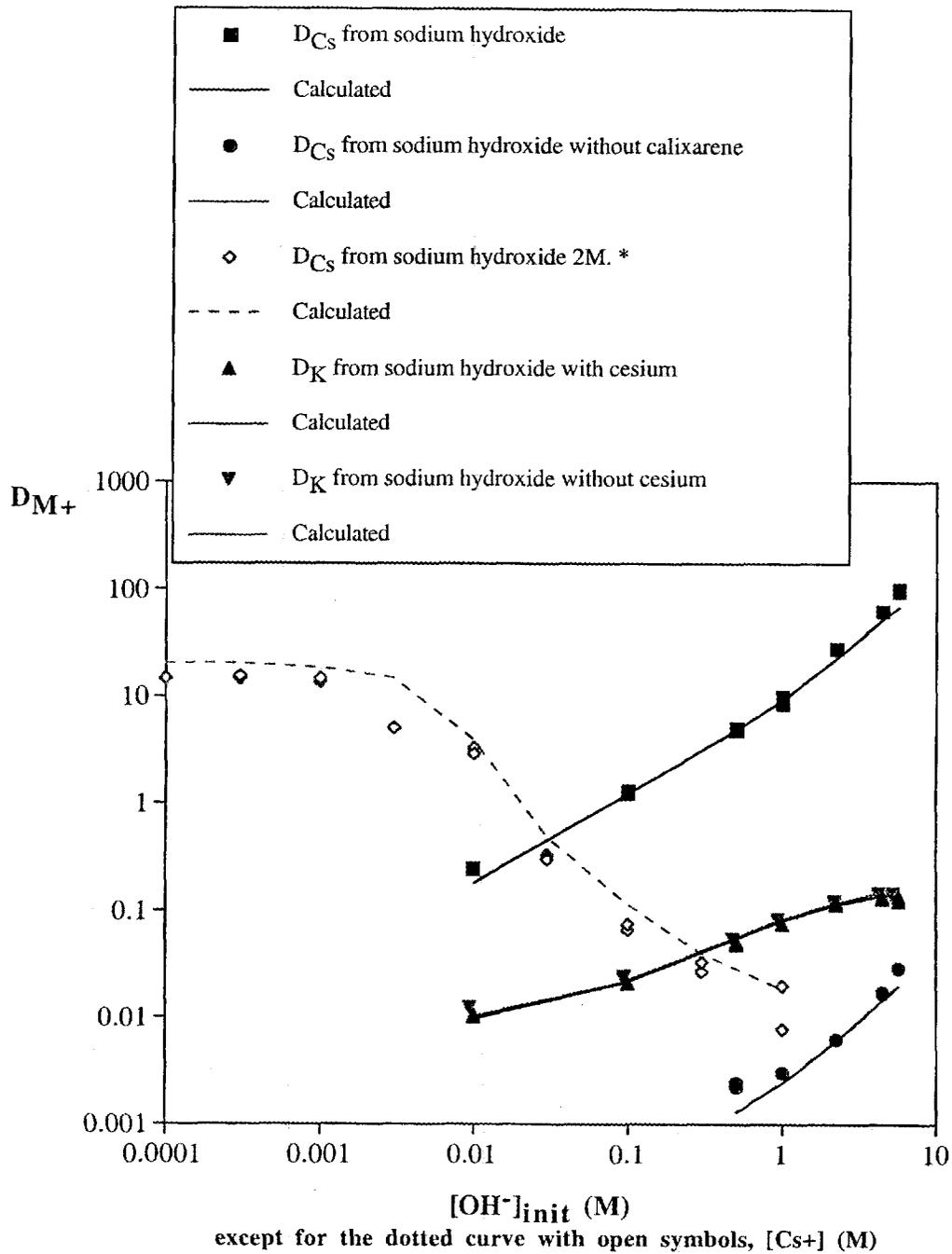


Fig. 3 . Fit of data points for hydroxide media

The fit for all the data is rather good with the exception of the curve for which the concentration of sodium hydroxide was held constant and the concentration of cesium nitrate varied (* dotted curve and open symbols = variable cesium nitrate concentration in NaOH 2 M). The reliability of these results was rather low since a third phase was observed for most of them. However, the results and the corresponding fit are presented to show that the overall trend is followed. Consider also, when a third phase is observed,

the distribution coefficients are usually lower than they would be without a third phase since some of the activity present in the organic phase that is subsampled for counting is present in the third phase. This is exactly what is observed in this case.

4.3. EXTRACTION MODELING FROM NITRITE OR CHLORIDE MEDIA

The inclusion of the data points containing nitrite salts led to the introduction of three more species in the model:

Table 6. Species and formation constants for the model derived for nitrite data

Species	Formation constant
CsNO ₂ Calix(o)	Log ₁₀ K = 3.152
KNO ₂ Calix(o)	Log ₁₀ K = 1.098
NaNO ₂ Calix(o)	Log ₁₀ K = -1.313

Among the anions in the study, chloride has the highest hydration energy, and therefore is not as extractable as the other three. The formation constants of the species involving this anion are expected to be lower than those found earlier. Extraction tests showed that sodium chloride is not extracted detectably when the calixarene is absent. The inclusion of the data points containing chloride salts led to comparable species:

Table 7. Species and formation constants for the model derived for chloride data

Species	Formation constant
CsClCalix(o)	Log ₁₀ K = 2.587
KClCalix(o)	Log ₁₀ K = 0.575
NaClCalix(o)	Log ₁₀ K = -1.455

Figures 4 and 5 present the fit obtained with the model for the systems containing chloride and nitrite, respectively.

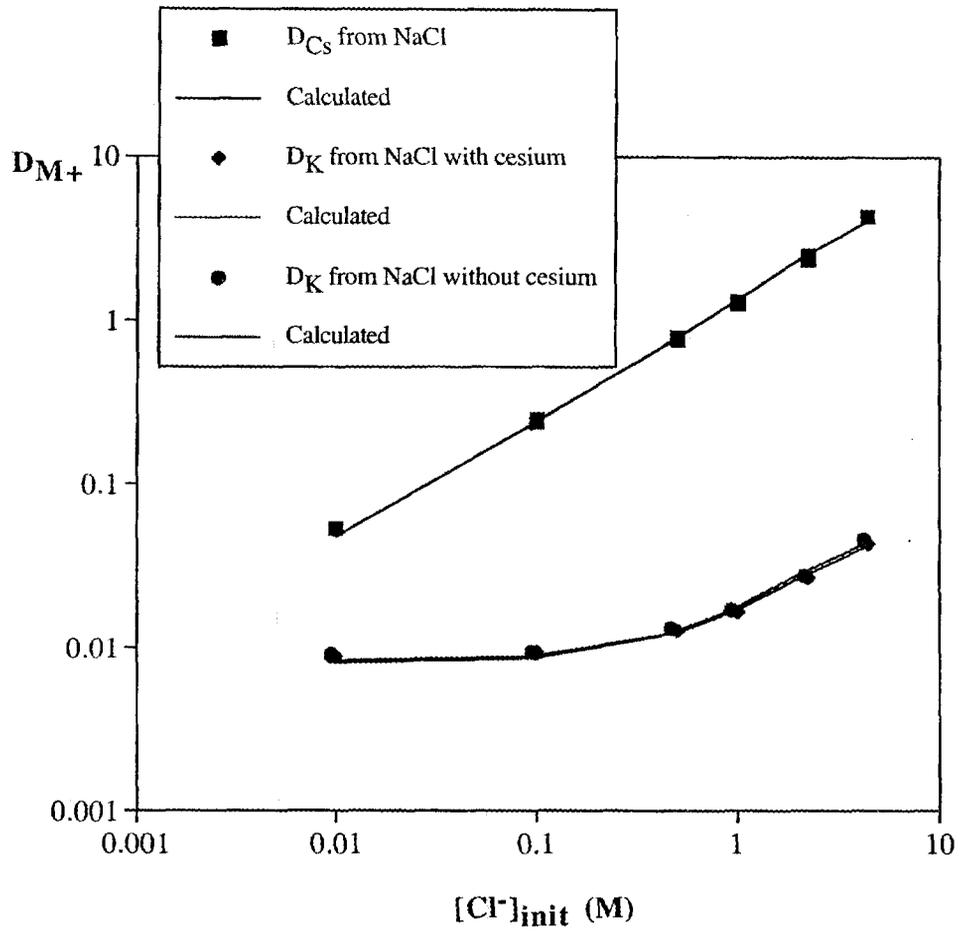


Fig. 4. Fit of cesium and potassium distribution ratios for chloride media

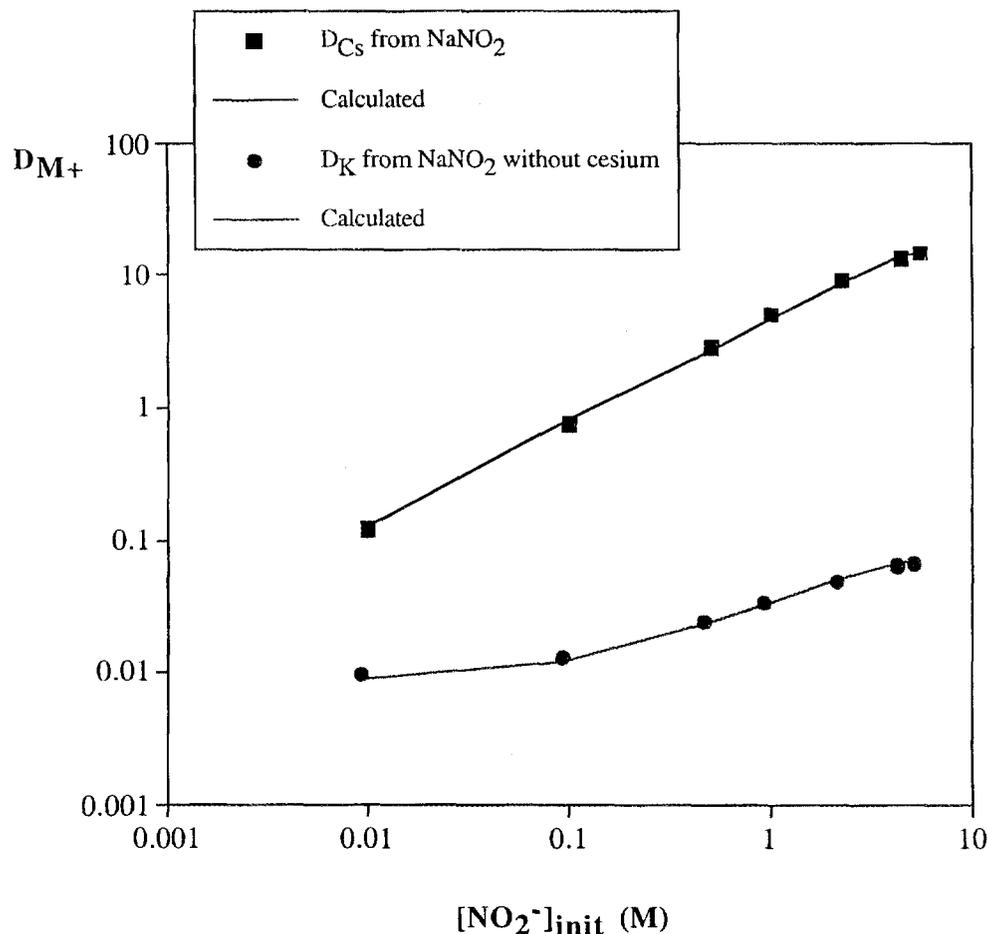


Fig. 5 Fit of cesium and potassium distribution ratios for nitrite media

These experiments conclude our tests from simple systems to determine the best model at 25 °C. The subsequent experiments attempted to validate the findings with more complicated systems.

4.4. VARIABLE TEMPERATURE TESTS

Two sets of experiments (extraction of cesium from sodium nitrate or sodium hydroxide) were carried out at two temperatures, 20 °C and 35 °C. It is shown that lower distribution ratios are obtained for higher temperatures, which agrees with the exothermic character of the extraction reaction.

The modeling of these data involved only the determination of the formation constants. Although the activity coefficients are also temperature dependent, the values for 25 °C were also used at 20 °C and 35 °C, since the change is small. Formation constants are mentioned for information purposes only. All

experiments carried out at 25 °C that allowed the determination of the model to this point would have been needed at other temperatures to determine an accurate model at these temperatures.

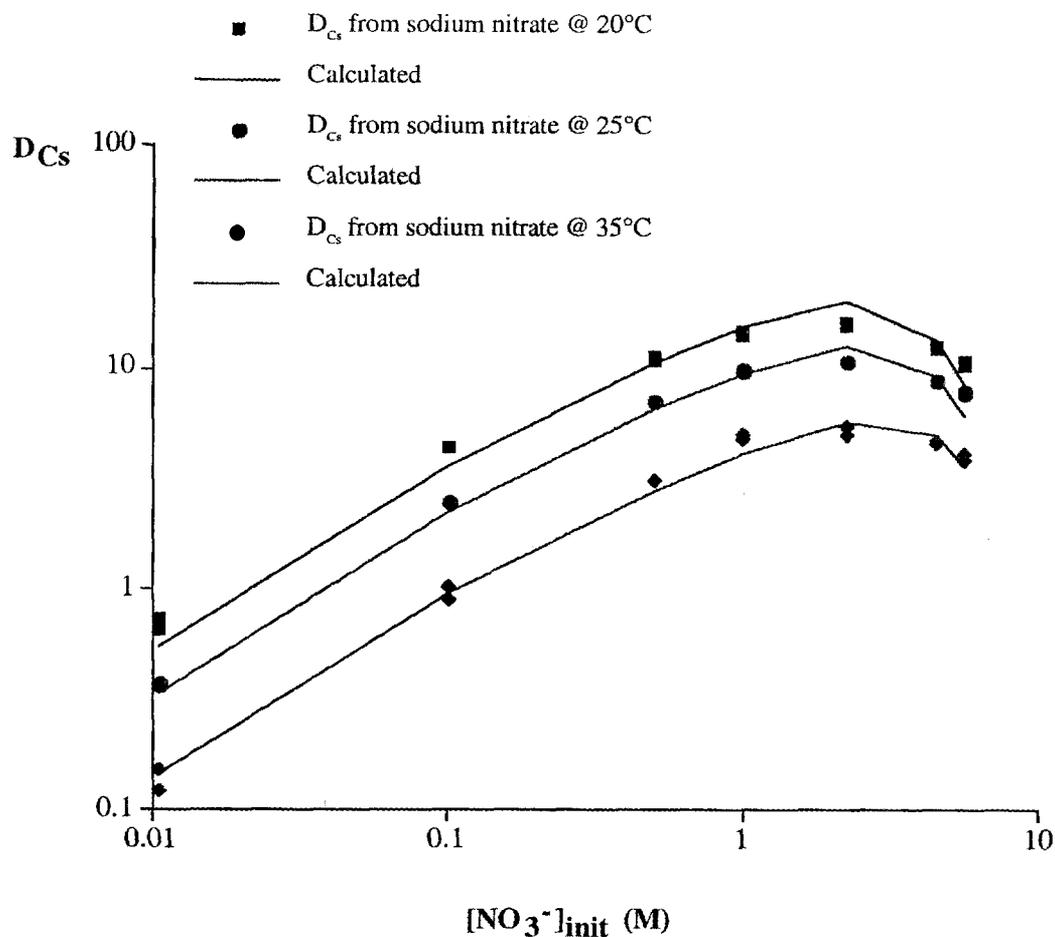


Fig. 6. Fit of cesium distribution ratios for nitrate media at different temperature

Table 8. Formation constant for the model including nitrate data at different temperatures

Species	Formation constant	Formation constant	Formation constant
	@ 20 °C	@ 25 °C	@ 35 °C
CsNO ₃ Calix(o)	Log ₁₀ K = 3.831	Log ₁₀ K = 3.615	Log ₁₀ K = 3.251
(CsNO ₃) ₂ Calix(o)	kept constant	Log ₁₀ K = 4.317	kept constant
NaNO ₃ Calix(o)	Log ₁₀ K = -0.851	Log ₁₀ K = -0.943	Log ₁₀ K = -1.152

The formation constant of the complex involving two cesium ions is held constant for the three temperatures since there are almost no data points supporting this species in the data sets collected at 20 °C or 35 °C. Formation constants of complexes containing cesium and sodium nitrate (Table 8) and cesium hydroxide (Table 9) were included in these models for the corresponding temperatures. Figures 6 and 7 present the fit obtained as a function of nitrate and hydroxide concentrations, respectively, at the different temperatures. The expected trend of the formation constant values follow very well the exothermic behavior observed for this system. The fit could be improved, particularly for the data at 35 °C, which suggests that some of the assumptions are not valid and the appropriate data sets need to be collected to reduce the uncertainties.

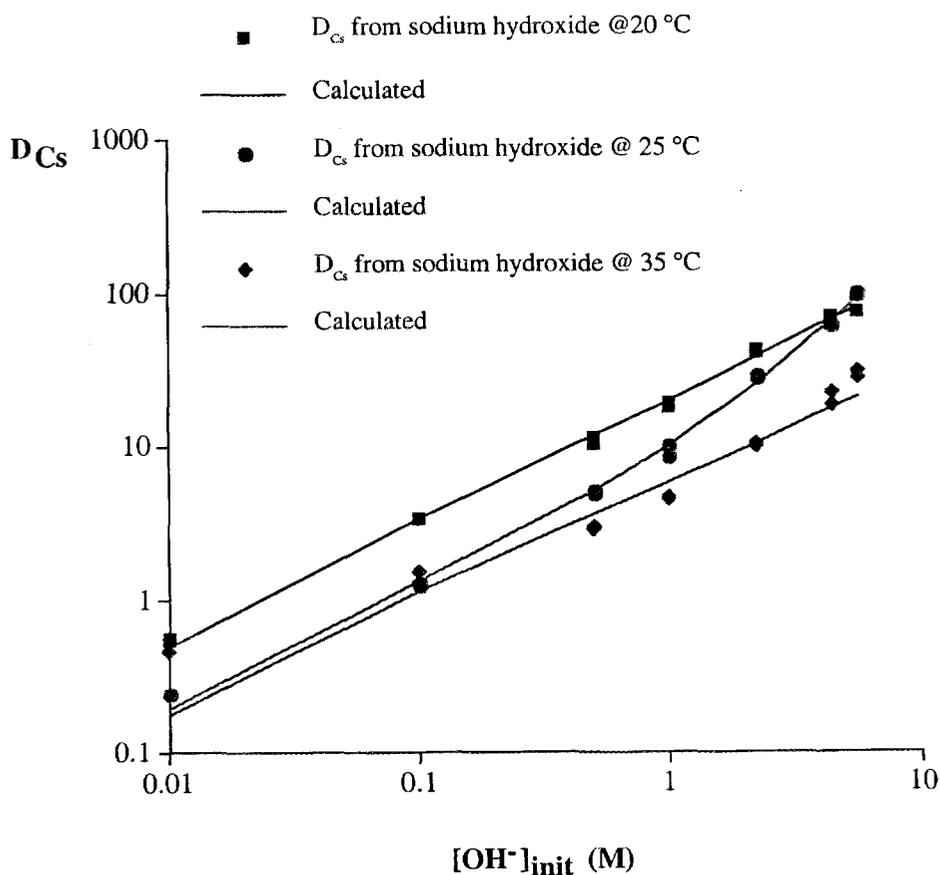


Fig. 7. Fit of cesium distribution ratios for hydroxide media at different temperature

Table 9. Formation constant for the model including hydroxide data at different temperatures

Species	Formation constant @ 20 °C	Formation constant @ 25 °C	Formation constant @ 35 °C
CsOHCalix(o)	Log ₁₀ K = 3.531	Log ₁₀ K = 3.332	Log ₁₀ K = 3.008

4.5. TESTS INVOLVING TANK SIMULANTS

Real waste batch tests conducted at the Savannah River Site provided the concentrations of major components (sodium, potassium, cesium, hydroxide, and nitrate) of the waste.¹ From these results, simulants were prepared using either nitrite or chloride. These initial concentrations were entered into a data file, and the program SXFIT predicted the cesium distribution ratio based on all the species and formation constants presented above. Table 10 presents the comparison between the measured distribution ratios and those that were predicted. The O/A volume ratios were all 1/3. All experiments were carried out at 25 °C.

Table 10. Comparison of measured and predicted distribution ratios for tank simulants

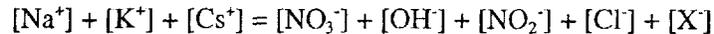
Tank	Composition	D _{Cs} measured	D _{Cs} predicted	error
Tank 13	[Na ⁺] = 5.6 M	8.38	9.75	16.3%
	[K ⁺] = 0.067 M			
	[Cs ⁺] = 0.512 mM			
	[OH ⁻] = 2.29 M			
	[NO ₃ ⁻] = 0.767 M			
	[NO ₂ ⁻] = 2.6 M			
Tank 13 (with chloride, substituted for nitrite)	[Na ⁺] = 5.6 M	6.68	6.88	3.0%
	[K ⁺] = 0.067 M			
	[Cs ⁺] = 0.512 mM			
	[OH ⁻] = 2.29 M			
	[NO ₃ ⁻] = 0.767 M			
	[Cl ⁻] = 2.6 M			
Tank 26	[Na ⁺] = 5.6 M	16.8	15.0	10.7%
	[K ⁺] = 0.041 M			
	[Cs ⁺] = 0.219 mM			
	[OH ⁻] = 4.71 M			
	[NO ₃ ⁻] = 0.956 M			
	[NO ₂ ⁻] = 0 M			

Tank	Composition	D_{Cs} measured	D_{Cs} predicted	error
Tank 33	[Na ⁺] = 3.3 M	16.3	20.5	25.7%
	[K ⁺] = 5 mM			
	[Cs ⁺] = 8.03 μ M			
	[OH ⁻] = 1.47 M			
	[NO ₃ ⁻] = 1.44 M			
	[NO ₂ ⁻] = 0.4 M			
Tank 35	[Na ⁺] = 5.6 M	22.8	23.9	4.6%
	[K ⁺] = 0.01 M			
	[Cs ⁺] = 0.188 mM			
	[OH ⁻] = 2.93 M			
	[NO ₃ ⁻] = 1.4 M			
	[NO ₂ ⁻] = 1.3 M			
Tank 46	[Na ⁺] = 5.6 M	17.6	17.3	1.7%
	[K ⁺] = 0.032 M			
	[Cs ⁺] = 0.378 mM			
	[OH ⁻] = 3.98 M			
	[NO ₃ ⁻] = 0.606 M			
	[NO ₂ ⁻] = 1.0 M			
Tank 46 (with chloride substituted for nitrite)	[Na ⁺] = 5.6 M	15.4	14.8	3.8%
	[K ⁺] = 0.032 M			
	[Cs ⁺] = 0.378 mM			
	[OH ⁻] = 3.98 M			
	[NO ₃ ⁻] = 0.606 M			
	[Cl ⁻] = 1.0 M			

The prediction is very good in all the cases, for any composition of the simulant. The only overestimation occurred for the simulant that contains a lower concentration of sodium. Otherwise, all the predictions are within an average of 7% of error, which is excellent for a model that contains a minimum number of product species. This comparison validates the model described previously.

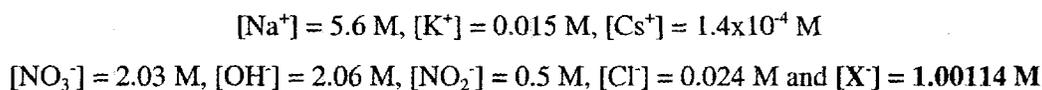
4.6. PREDICTION OF CESIUM EXTRACTION FROM THE FULL SIMULANT

Simplified tank simulants were prepared containing only those ions assumed to be playing major roles in the systems. Following this assumption, we decided to go further and test our model by creating a data file containing the initial concentrations representing the full simulant. In order to test our model with the full simulant,⁹ a “dummy non-extractable” anion (X⁻) was introduced to ensure the electroneutrality of the aqueous phase:



This “dummy” anion is intended to represent all the anions present in the simulant that are not extracted and assumed to play only minor roles in the extraction process. However, they are important from a global ionic strength standpoint.

Under the aqueous initial conditions:



and the species with their respective formation constants listed in Table 11, the predicted cesium distribution ratio is 16.8. The average of all cesium distribution ratios obtained in extraction conditions is 16.9.¹⁰ This comparison allows a still greater confidence in the reliability of the model, and confirms that the anions present in the simulant other than hydroxide, nitrate, nitrite, and chloride do not play a major role.

Table 11. Model used in this work

Species	Formation constant
CsNO ₃ Calix(o)	Log ₁₀ K = 3.615
(CsNO ₃) ₂ Calix(o)	Log ₁₀ K = 4.317
CsOH(o)	Log ₁₀ K = -2.263
CsOHCalix(o)	Log ₁₀ K = 3.332
CsNO ₂ Calix(o)	Log ₁₀ K = 3.152
CsClCalix(o)	Log ₁₀ K = 2.587
KNO ₃ Calix(o)	Log ₁₀ K = 1.387
KOHCalix(o)	Log ₁₀ K = 1.549
KNO ₂ Calix(o)	Log ₁₀ K = 1.098
KClCalix(o)	Log ₁₀ K = 0.575
NaOH(o)	Log ₁₀ K = -0.565
NaNO ₃ Calix(o)	Log ₁₀ K = -0.943
NaNO ₂ Calix(o)	Log ₁₀ K = -1.313
NaClCalix(o)	Log ₁₀ K = -1.455

5. CONCLUSION

The model gives a very good overall fit for a large number of data points (almost 300) obtained from simple systems. The overall agreement is adequate for such a large data set and the number of species assumed in the organic phase is very small. In addition, all the formation constants are consistent within themselves. They follow the values of the Gibbs energy of partitioning for the four anions. The definitive test to predict the cesium distribution ratios based on initial concentrations is extremely satisfactory. In addition, the prediction of the distribution coefficient obtained with the full simulant is very close to the value obtained experimentally. We can say that not only does the model fit the data very well, but it also includes the cations and anions that play major roles in more complicated mixtures.

6. REFERENCES

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7. APPENDIX

Data points obtained experimentally and organized for modeling with the program

D _{value}	Error	[Lig]	[Na ⁺]	[K ⁺]	[Cs ⁺]	[H ⁺]	[NO ₃ ⁻]	[OH ⁻]	[NO ₂ ⁻]	[Cl ⁻]
1.39E-02	6.95E-04	0.01	0.000E+00	0.00E+00	1.00E+00	0.00E+00	1.00000E+00	0.0E+00	0.0E+00	0.0E+00
1.34E-02	6.70E-04	0.01	0.000E+00	0.00E+00	1.00E+00	0.00E+00	1.00000E+00	0.0E+00	0.0E+00	0.0E+00
5.03E-02	2.52E-03	0.01	0.000E+00	0.00E+00	3.00E-01	0.00E+00	3.00000E-01	0.0E+00	0.0E+00	0.0E+00
5.05E-02	2.53E-03	0.01	0.000E+00	0.00E+00	3.00E-01	0.00E+00	3.00000E-01	0.0E+00	0.0E+00	0.0E+00
1.07E-01	5.35E-03	0.01	0.000E+00	0.00E+00	1.00E-01	0.00E+00	1.00000E-01	0.0E+00	0.0E+00	0.0E+00
1.07E-01	5.35E-03	0.01	0.000E+00	0.00E+00	1.00E-01	0.00E+00	1.00000E-01	0.0E+00	0.0E+00	0.0E+00
2.69E-01	1.35E-02	0.01	0.000E+00	0.00E+00	3.00E-02	0.00E+00	3.00000E-02	0.0E+00	0.0E+00	0.0E+00
2.34E-01	1.17E-02	0.01	0.000E+00	0.00E+00	3.00E-02	0.00E+00	3.00000E-02	0.0E+00	0.0E+00	0.0E+00
2.17E-01	1.09E-02	0.01	0.000E+00	0.00E+00	1.00E-02	0.00E+00	1.00000E-02	0.0E+00	0.0E+00	0.0E+00
1.96E-01	9.80E-03	0.01	0.000E+00	0.00E+00	1.00E-02	0.00E+00	1.00000E-02	0.0E+00	0.0E+00	0.0E+00
4.41E-02	2.21E-03	0.01	0.000E+00	0.00E+00	5.00E-04	1.00E-03	1.50000E-03	0.0E+00	0.0E+00	0.0E+00
4.44E-02	2.22E-03	0.01	0.000E+00	0.00E+00	5.00E-04	1.00E-03	1.50000E-03	0.0E+00	0.0E+00	0.0E+00
1.26E-01	6.30E-03	0.01	0.000E+00	0.00E+00	5.00E-04	3.00E-03	3.50000E-03	0.0E+00	0.0E+00	0.0E+00
1.24E-01	6.20E-03	0.01	0.000E+00	0.00E+00	5.00E-04	3.00E-03	3.50000E-03	0.0E+00	0.0E+00	0.0E+00
3.48E-01	1.74E-02	0.01	0.000E+00	0.00E+00	5.00E-04	1.00E-02	1.05000E-02	0.0E+00	0.0E+00	0.0E+00
2.85E-01	1.43E-02	0.01	0.000E+00	0.00E+00	5.00E-04	1.00E-02	1.05000E-02	0.0E+00	0.0E+00	0.0E+00
1.46E+00	7.30E-02	0.01	0.000E+00	0.00E+00	5.00E-04	5.00E-02	5.05000E-02	0.0E+00	0.0E+00	0.0E+00
1.46E+00	7.30E-02	0.01	0.000E+00	0.00E+00	5.00E-04	5.00E-02	5.05000E-02	0.0E+00	0.0E+00	0.0E+00
2.52E+00	1.26E-01	0.01	0.000E+00	0.00E+00	5.00E-04	1.00E-01	1.00500E-01	0.0E+00	0.0E+00	0.0E+00
2.47E+00	1.24E-01	0.01	0.000E+00	0.00E+00	5.00E-04	1.00E-01	1.00500E-01	0.0E+00	0.0E+00	0.0E+00
3.78E-01	1.89E-02	0.01	1.000E-02	0.00E+00	5.00E-04	0.00E+00	1.05000E-02	0.0E+00	0.0E+00	0.0E+00
3.68E-01	1.84E-02	0.01	1.000E-02	0.00E+00	5.00E-04	0.00E+00	1.05000E-02	0.0E+00	0.0E+00	0.0E+00
2.50E+00	1.25E-01	0.01	1.000E-01	0.00E+00	5.00E-04	0.00E+00	1.00500E-01	0.0E+00	0.0E+00	0.0E+00
2.50E+00	1.25E-01	0.01	1.000E-01	0.00E+00	5.00E-04	0.00E+00	1.00500E-01	0.0E+00	0.0E+00	0.0E+00
7.14E+00	3.57E-01	0.01	5.000E-01	0.00E+00	5.00E-04	0.00E+00	5.00500E-01	0.0E+00	0.0E+00	0.0E+00
7.23E+00	3.62E-01	0.01	5.000E-01	0.00E+00	5.00E-04	0.00E+00	5.00500E-01	0.0E+00	0.0E+00	0.0E+00
9.84E+00	4.92E-01	0.01	1.000E+00	0.00E+00	5.00E-04	0.00E+00	1.00050E+00	0.0E+00	0.0E+00	0.0E+00
9.87E+00	4.94E-01	0.01	1.000E+00	0.00E+00	5.00E-04	0.00E+00	1.00050E+00	0.0E+00	0.0E+00	0.0E+00
1.08E+01	5.40E-01	0.01	2.250E+00	0.00E+00	5.00E-04	0.00E+00	2.25050E+00	0.0E+00	0.0E+00	0.0E+00
1.09E+01	5.45E-01	0.01	2.250E+00	0.00E+00	5.00E-04	0.00E+00	2.25050E+00	0.0E+00	0.0E+00	0.0E+00
9.00E+00	4.50E-01	0.01	4.500E+00	0.00E+00	5.00E-04	0.00E+00	4.50050E+00	0.0E+00	0.0E+00	0.0E+00
8.95E+00	4.48E-01	0.01	4.500E+00	0.00E+00	5.00E-04	0.00E+00	4.50050E+00	0.0E+00	0.0E+00	0.0E+00
7.96E+00	3.98E-01	0.01	5.600E+00	0.00E+00	5.00E-04	0.00E+00	5.60050E+00	0.0E+00	0.0E+00	0.0E+00
7.84E+00	3.92E-01	0.01	5.600E+00	0.00E+00	5.00E-04	0.00E+00	5.60050E+00	0.0E+00	0.0E+00	0.0E+00
5.78E-03	2.89E-04	0.01	0.000E+00	1.51E+00	0.00E+00	0.00E+00	1.51000E+00	0.0E+00	0.0E+00	0.0E+00
5.66E-03	2.83E-04	0.01	0.000E+00	1.51E+00	0.00E+00	0.00E+00	1.51000E+00	0.0E+00	0.0E+00	0.0E+00
9.92E-03	4.96E-04	0.01	0.000E+00	7.55E-01	0.00E+00	0.00E+00	7.55000E-01	0.0E+00	0.0E+00	0.0E+00
9.95E-03	4.98E-04	0.01	0.000E+00	7.55E-01	0.00E+00	0.00E+00	7.55000E-01	0.0E+00	0.0E+00	0.0E+00
1.30E-02	6.49E-04	0.01	0.000E+00	3.00E-01	0.00E+00	0.00E+00	3.00000E-01	0.0E+00	0.0E+00	0.0E+00
1.33E-02	6.65E-04	0.01	0.000E+00	3.00E-01	0.00E+00	0.00E+00	3.00000E-01	0.0E+00	0.0E+00	0.0E+00
8.15E-03	4.07E-04	0.01	0.000E+00	1.00E-01	0.00E+00	0.00E+00	1.00000E-01	0.0E+00	0.0E+00	0.0E+00
8.36E-03	4.18E-04	0.01	0.000E+00	1.00E-01	0.00E+00	0.00E+00	1.00000E-01	0.0E+00	0.0E+00	0.0E+00
1.13E-02	5.66E-04	0.01	9.400E-03	6.00E-02	0.00E+00	0.00E+00	6.94000E-02	0.0E+00	0.0E+00	0.0E+00
1.12E-02	5.58E-04	0.01	9.400E-03	6.00E-02	0.00E+00	0.00E+00	6.94000E-02	0.0E+00	0.0E+00	0.0E+00
1.95E-02	9.77E-04	0.01	9.400E-02	6.00E-02	0.00E+00	0.00E+00	1.54000E-01	0.0E+00	0.0E+00	0.0E+00
1.97E-02	9.83E-04	0.01	9.400E-02	6.00E-02	0.00E+00	0.00E+00	1.54000E-01	0.0E+00	0.0E+00	0.0E+00
3.81E-02	1.91E-03	0.01	4.700E-01	6.00E-02	0.00E+00	0.00E+00	5.30000E-01	0.0E+00	0.0E+00	0.0E+00
3.81E-02	1.90E-03	0.01	4.700E-01	6.00E-02	0.00E+00	0.00E+00	5.30000E-01	0.0E+00	0.0E+00	0.0E+00
4.86E-02	2.43E-03	0.01	9.400E-01	6.00E-02	0.00E+00	0.00E+00	1.00000E+00	0.0E+00	0.0E+00	0.0E+00
5.48E-02	2.74E-03	0.01	2.162E+00	6.00E-02	0.00E+00	0.00E+00	2.22200E+00	0.0E+00	0.0E+00	0.0E+00
5.51E-02	2.75E-03	0.01	2.162E+00	6.00E-02	0.00E+00	0.00E+00	2.22200E+00	0.0E+00	0.0E+00	0.0E+00
5.01E-02	2.51E-03	0.01	4.230E+00	6.00E-02	0.00E+00	0.00E+00	4.29000E+00	0.0E+00	0.0E+00	0.0E+00
5.00E-02	2.50E-03	0.01	4.230E+00	6.00E-02	0.00E+00	0.00E+00	4.29000E+00	0.0E+00	0.0E+00	0.0E+00
4.41E-02	2.20E-03	0.01	5.264E+00	6.00E-02	0.00E+00	0.00E+00	5.32400E+00	0.0E+00	0.0E+00	0.0E+00

4.74E-02	2.37E-03	0.01	5.264E+00	6.00E-02	0.00E+00	0.00E+00	5.32400E+00	0.0E+00	0.0E+00	0.0E+00
9.33E-03	4.67E-04	0.01	1.000E-02	6.00E-02	5.00E-04	0.00E+00	7.05000E-02	0.0E+00	0.0E+00	0.0E+00
9.37E-03	4.69E-04	0.01	1.000E-02	6.00E-02	5.00E-04	0.00E+00	7.05000E-02	0.0E+00	0.0E+00	0.0E+00
1.66E-02	8.31E-04	0.01	1.000E-01	6.00E-02	5.00E-04	0.00E+00	1.60500E-01	0.0E+00	0.0E+00	0.0E+00
1.67E-02	8.36E-04	0.01	1.000E-01	6.00E-02	5.00E-04	0.00E+00	1.60500E-01	0.0E+00	0.0E+00	0.0E+00
3.13E-02	1.57E-03	0.01	5.000E-01	6.00E-02	5.00E-04	0.00E+00	5.60500E-01	0.0E+00	0.0E+00	0.0E+00
3.20E-02	1.60E-03	0.01	5.000E-01	6.00E-02	5.00E-04	0.00E+00	5.60500E-01	0.0E+00	0.0E+00	0.0E+00
4.04E-02	2.02E-03	0.01	1.000E+00	6.00E-02	5.00E-04	0.00E+00	1.06050E+00	0.0E+00	0.0E+00	0.0E+00
4.10E-02	2.05E-03	0.01	1.000E+00	6.00E-02	5.00E-04	0.00E+00	1.06050E+00	0.0E+00	0.0E+00	0.0E+00
4.78E-02	2.39E-03	0.01	2.250E+00	6.00E-02	5.00E-04	0.00E+00	2.31050E+00	0.0E+00	0.0E+00	0.0E+00
4.66E-02	2.33E-03	0.01	2.250E+00	6.00E-02	5.00E-04	0.00E+00	2.31050E+00	0.0E+00	0.0E+00	0.0E+00
4.21E-02	2.10E-03	0.01	4.500E+00	6.00E-02	5.00E-04	0.00E+00	4.56050E+00	0.0E+00	0.0E+00	0.0E+00
4.31E-02	2.16E-03	0.01	4.500E+00	6.00E-02	5.00E-04	0.00E+00	4.56050E+00	0.0E+00	0.0E+00	0.0E+00
4.04E-02	2.02E-03	0.01	5.600E+00	6.00E-02	5.00E-04	0.00E+00	5.66050E+00	0.0E+00	0.0E+00	0.0E+00
3.87E-02	1.94E-03	0.01	5.600E+00	6.00E-02	5.00E-04	0.00E+00	5.66050E+00	0.0E+00	0.0E+00	0.0E+00
8.82E-03	4.41E-04	0.01	0.000E+00	6.00E-02	5.00E-04	1.00E-03	6.15000E-02	0.0E+00	0.0E+00	0.0E+00
8.97E-03	4.49E-04	0.01	0.000E+00	6.00E-02	5.00E-04	1.00E-03	6.15000E-02	0.0E+00	0.0E+00	0.0E+00
8.98E-03	4.49E-04	0.01	0.000E+00	6.00E-02	5.00E-04	3.00E-03	6.35000E-02	0.0E+00	0.0E+00	0.0E+00
9.19E-03	4.59E-04	0.01	0.000E+00	6.00E-02	5.00E-04	3.00E-03	6.35000E-02	0.0E+00	0.0E+00	0.0E+00
9.78E-03	4.89E-04	0.01	0.000E+00	6.00E-02	5.00E-04	1.00E-02	7.05000E-02	0.0E+00	0.0E+00	0.0E+00
1.04E-02	5.21E-04	0.01	0.000E+00	6.00E-02	5.00E-04	1.00E-02	7.05000E-02	0.0E+00	0.0E+00	0.0E+00
1.36E-02	6.82E-04	0.01	0.000E+00	6.00E-02	5.00E-04	5.00E-02	1.10500E-01	0.0E+00	0.0E+00	0.0E+00
1.40E-02	6.99E-04	0.01	0.000E+00	6.00E-02	5.00E-04	5.00E-02	1.10500E-01	0.0E+00	0.0E+00	0.0E+00
1.78E-02	8.92E-04	0.01	0.000E+00	6.00E-02	5.00E-04	1.00E-01	1.60500E-01	0.0E+00	0.0E+00	0.0E+00
1.82E-02	9.08E-04	0.01	0.000E+00	6.00E-02	5.00E-04	1.00E-01	1.60500E-01	0.0E+00	0.0E+00	0.0E+00
2.94E-03	1.47E-04	0.01	0.000E+00	1.00E-02	0.00E+00	0.00E+00	1.00000E-02	0.0E+00	0.0E+00	0.0E+00
2.49E-03	1.25E-04	0.01	0.000E+00	1.00E-02	0.00E+00	0.00E+00	1.00000E-02	0.0E+00	0.0E+00	0.0E+00
1.23E-02	6.15E-04	0.01	0.000E+00	1.00E-01	0.00E+00	0.00E+00	1.00000E-01	0.0E+00	0.0E+00	0.0E+00
1.22E-02	6.11E-04	0.01	0.000E+00	1.00E-01	0.00E+00	0.00E+00	1.00000E-01	0.0E+00	0.0E+00	0.0E+00
1.59E-02	7.94E-04	0.01	0.000E+00	3.00E-01	0.00E+00	0.00E+00	3.00000E-01	0.0E+00	0.0E+00	0.0E+00
1.58E-02	7.92E-04	0.01	0.000E+00	3.00E-01	0.00E+00	0.00E+00	3.00000E-01	0.0E+00	0.0E+00	0.0E+00
8.98E-03	4.49E-04	0.01	0.000E+00	1.00E+00	0.00E+00	0.00E+00	1.00000E+00	0.0E+00	0.0E+00	0.0E+00
9.12E-03	4.56E-04	0.01	0.000E+00	1.00E+00	0.00E+00	0.00E+00	1.00000E+00	0.0E+00	0.0E+00	0.0E+00
2.47E-01	1.24E-02	0.01	1.00E-02	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	1.00E-02	0.00E+00	0.00E+00
2.51E-01	1.26E-02	0.01	1.00E-02	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	1.00E-02	0.00E+00	0.00E+00
1.32E+00	6.60E-02	0.01	1.00E-01	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	1.00E-01	0.00E+00	0.00E+00
1.26E+00	6.30E-02	0.01	1.00E-01	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	1.00E-01	0.00E+00	0.00E+00
5.18E+00	2.59E-01	0.01	5.00E-01	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.00E-01	0.00E+00	0.00E+00
4.96E+00	2.48E-01	0.01	5.00E-01	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.00E-01	0.00E+00	0.00E+00
1.02E+01	5.10E-01	0.01	1.00E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	1.00E+00	0.00E+00	0.00E+00
8.65E+00	4.33E-01	0.01	1.00E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	1.00E+00	0.00E+00	0.00E+00
2.83E+01	1.42E+00	0.01	2.25E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	2.25E+00	0.00E+00	0.00E+00
2.89E+01	1.45E+00	0.01	2.25E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	2.25E+00	0.00E+00	0.00E+00
6.29E+01	3.15E+00	0.01	4.50E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	4.50E+00	0.00E+00	0.00E+00
6.31E+01	3.16E+00	0.01	4.50E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	4.50E+00	0.00E+00	0.00E+00
1.02E+02	5.10E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.60E+00	0.00E+00	0.00E+00
9.88E+01	4.94E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.60E+00	0.00E+00	0.00E+00
2.25E-03	1.13E-04	0.00	5.00E-01	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.00E-01	0.00E+00	0.00E+00
2.54E-03	1.27E-04	0.00	5.00E-01	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.00E-01	0.00E+00	0.00E+00
3.04E-03	1.52E-04	0.00	1.00E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	1.00E+00	0.00E+00	0.00E+00
3.05E-03	1.53E-04	0.00	1.00E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	1.00E+00	0.00E+00	0.00E+00
6.32E-03	3.16E-04	0.00	2.25E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	2.25E+00	0.00E+00	0.00E+00
6.45E-03	3.23E-04	0.00	2.25E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	2.25E+00	0.00E+00	0.00E+00
1.75E-02	8.75E-04	0.00	4.50E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	4.50E+00	0.00E+00	0.00E+00
1.73E-02	8.65E-04	0.00	4.50E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	4.50E+00	0.00E+00	0.00E+00
2.99E-02	1.50E-03	0.00	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.60E+00	0.00E+00	0.00E+00
2.96E-02	1.48E-03	0.00	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.60E+00	0.00E+00	0.00E+00
2.03E-02	1.02E-03	0.01	2.00E+00	0.00E+00	1.00E+00	0.00E+00	1.0000E+00	2.00E+00	0.00E+00	0.00E+00
8.07E-03	4.04E-04	0.01	2.00E+00	0.00E+00	1.00E+00	0.00E+00	1.0000E+00	2.00E+00	0.00E+00	0.00E+00
2.75E-02	1.38E-03	0.01	2.00E+00	0.00E+00	3.00E-01	0.00E+00	3.0000E-01	2.00E+00	0.00E+00	0.00E+00
3.36E-02	1.68E-03	0.01	2.00E+00	0.00E+00	3.00E-01	0.00E+00	3.0000E-01	2.00E+00	0.00E+00	0.00E+00
6.63E-02	3.32E-03	0.01	2.00E+00	0.00E+00	1.00E-01	0.00E+00	1.0000E-01	2.00E+00	0.00E+00	0.00E+00
7.70E-02	3.85E-03	0.01	2.00E+00	0.00E+00	1.00E-01	0.00E+00	1.0000E-01	2.00E+00	0.00E+00	0.00E+00
3.44E-01	1.72E-02	0.01	2.00E+00	0.00E+00	3.00E-02	0.00E+00	3.0000E-02	2.00E+00	0.00E+00	0.00E+00

3.14E-01	1.57E-02	0.01	2.00E+00	0.00E+00	3.00E-02	0.00E+00	3.0000E-02	2.00E+00	0.00E+00	0.00E+00
3.35E+00	1.68E-01	0.01	2.00E+00	0.00E+00	1.00E-02	0.00E+00	1.0000E-02	2.00E+00	0.00E+00	0.00E+00
3.02E+00	1.51E-01	0.01	2.00E+00	0.00E+00	1.00E-02	0.00E+00	1.0000E-02	2.00E+00	0.00E+00	0.00E+00
5.07E+00	2.54E-01	0.01	2.00E+00	0.00E+00	3.00E-03	0.00E+00	3.0000E-03	2.00E+00	0.00E+00	0.00E+00
5.09E+00	2.55E-01	0.01	2.00E+00	0.00E+00	3.00E-03	0.00E+00	3.0000E-03	2.00E+00	0.00E+00	0.00E+00
1.36E+01	6.80E-01	0.01	2.00E+00	0.00E+00	1.00E-03	0.00E+00	1.0000E-03	2.00E+00	0.00E+00	0.00E+00
1.48E+01	7.40E-01	0.01	2.00E+00	0.00E+00	1.00E-03	0.00E+00	1.0000E-03	2.00E+00	0.00E+00	0.00E+00
1.48E+01	7.40E-01	0.01	2.00E+00	0.00E+00	3.00E-04	0.00E+00	3.0000E-04	2.00E+00	0.00E+00	0.00E+00
1.56E+01	7.80E-01	0.01	2.00E+00	0.00E+00	3.00E-04	0.00E+00	3.0000E-04	2.00E+00	0.00E+00	0.00E+00
1.49E+01	7.45E-01	0.01	2.00E+00	0.00E+00	1.00E-04	0.00E+00	1.0000E-04	2.00E+00	0.00E+00	0.00E+00
1.48E+01	7.40E-01	0.01	2.00E+00	0.00E+00	1.00E-04	0.00E+00	1.0000E-04	2.00E+00	0.00E+00	0.00E+00
7.96E+00	3.98E-01	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.6005E+00	0.00E+00	0.00E+00	0.00E+00
7.84E+00	3.92E-01	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.6005E+00	0.00E+00	0.00E+00	0.00E+00
9.98E+00	4.99E-01	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0405E+00	5.60E-01	0.00E+00	0.00E+00
9.88E+00	4.94E-01	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0405E+00	5.60E-01	0.00E+00	0.00E+00
1.33E+01	6.65E-01	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	4.2005E+00	1.40E+00	0.00E+00	0.00E+00
1.31E+01	6.55E-01	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	4.2005E+00	1.40E+00	0.00E+00	0.00E+00
2.51E+01	1.26E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	2.8005E+00	2.80E+00	0.00E+00	0.00E+00
2.31E+01	1.16E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	2.8005E+00	2.80E+00	0.00E+00	0.00E+00
4.71E+01	2.36E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	1.4005E+00	4.20E+00	0.00E+00	0.00E+00
4.74E+01	2.37E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	1.4005E+00	4.20E+00	0.00E+00	0.00E+00
1.02E+02	5.10E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.60E+00	0.00E+00	0.00E+00
9.88E+01	4.94E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.60E+00	0.00E+00	0.00E+00
1.07E-02	5.34E-04	0.01	1.00E-02	6E-02	5.00E-04	0.00E+00	6.05E-02	1.000E-02	0.00E+00	0.00E+00
1.03E-02	5.13E-04	0.01	1.00E-02	6E-02	5.00E-04	0.00E+00	6.05E-02	1.000E-02	0.00E+00	0.00E+00
2.07E-02	1.03E-03	0.01	1.00E-01	6E-02	5.00E-04	0.00E+00	6.05E-02	1.000E-01	0.00E+00	0.00E+00
2.08E-02	1.04E-03	0.01	1.00E-01	6E-02	5.00E-04	0.00E+00	6.05E-02	1.000E-01	0.00E+00	0.00E+00
4.90E-02	2.45E-03	0.01	5.00E-01	6E-02	5.00E-04	0.00E+00	6.05E-02	5.000E-01	0.00E+00	0.00E+00
5.11E-02	2.55E-03	0.01	5.00E-01	6E-02	5.00E-04	0.00E+00	6.05E-02	5.000E-01	0.00E+00	0.00E+00
7.44E-02	3.72E-03	0.01	1.00E+00	6E-02	5.00E-04	0.00E+00	6.05E-02	1.000E+00	0.00E+00	0.00E+00
7.67E-02	3.83E-03	0.01	1.00E+00	6E-02	5.00E-04	0.00E+00	6.05E-02	1.000E+00	0.00E+00	0.00E+00
1.18E-01	5.88E-03	0.01	2.25E+00	6E-02	5.00E-04	0.00E+00	6.05E-02	2.250E+00	0.00E+00	0.00E+00
1.22E-01	6.10E-03	0.01	2.25E+00	6E-02	5.00E-04	0.00E+00	6.05E-02	2.250E+00	0.00E+00	0.00E+00
1.31E-01	6.54E-03	0.01	4.50E+00	6E-02	5.00E-04	0.00E+00	6.05E-02	4.500E+00	0.00E+00	0.00E+00
1.36E-01	6.79E-03	0.01	4.50E+00	6E-02	5.00E-04	0.00E+00	6.05E-02	4.500E+00	0.00E+00	0.00E+00
1.26E-01	6.29E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	6.05E-02	5.600E+00	0.00E+00	0.00E+00
1.39E-01	6.94E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	6.05E-02	5.600E+00	0.00E+00	0.00E+00
1.28E-02	6.41E-04	0.01	9.400E-03	6E-02	0.00E+00	0.00E+00	6.00E-02	9.400E-03	0.00E+00	0.00E+00
1.27E-02	6.34E-04	0.01	9.400E-03	6E-02	0.00E+00	0.00E+00	6.00E-02	9.400E-03	0.00E+00	0.00E+00
2.37E-02	1.18E-03	0.01	9.400E-02	6E-02	0.00E+00	0.00E+00	6.00E-02	9.400E-02	0.00E+00	0.00E+00
2.51E-02	1.25E-03	0.01	9.400E-02	6E-02	0.00E+00	0.00E+00	6.00E-02	9.400E-02	0.00E+00	0.00E+00
5.74E-02	2.87E-03	0.01	4.700E-01	6E-02	0.00E+00	0.00E+00	6.00E-02	4.700E-01	0.00E+00	0.00E+00
5.72E-02	2.86E-03	0.01	4.700E-01	6E-02	0.00E+00	0.00E+00	6.00E-02	4.700E-01	0.00E+00	0.00E+00
8.59E-02	4.30E-03	0.01	9.400E-01	6E-02	0.00E+00	0.00E+00	6.00E-02	9.400E-01	0.00E+00	0.00E+00
8.76E-02	4.38E-03	0.01	9.400E-01	6E-02	0.00E+00	0.00E+00	6.00E-02	9.400E-01	0.00E+00	0.00E+00
1.19E-01	5.95E-03	0.01	2.162E+00	6E-02	0.00E+00	0.00E+00	6.00E-02	2.162E+00	0.00E+00	0.00E+00
1.24E-01	6.19E-03	0.01	2.162E+00	6E-02	0.00E+00	0.00E+00	6.00E-02	2.162E+00	0.00E+00	0.00E+00
1.51E-01	7.53E-03	0.01	4.230E+00	6E-02	0.00E+00	0.00E+00	6.00E-02	4.230E+00	0.00E+00	0.00E+00
1.53E-01	7.66E-03	0.01	4.230E+00	6E-02	0.00E+00	0.00E+00	6.00E-02	4.230E+00	0.00E+00	0.00E+00
1.46E-01	7.30E-03	0.01	5.152E+00	6E-02	0.00E+00	0.00E+00	6.00E-02	5.152E+00	0.00E+00	0.00E+00
1.50E-01	7.52E-03	0.01	5.152E+00	6E-02	0.00E+00	0.00E+00	6.00E-02	5.152E+00	0.00E+00	0.00E+00
4.04E-02	2.02E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	5.6605E+00	0.00E+00	0.00E+00	0.00E+00
3.87E-02	1.94E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	5.6605E+00	0.00E+00	0.00E+00	0.00E+00
5.19E-02	2.59E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	5.1005E+00	5.60E-01	0.00E+00	0.00E+00
5.31E-02	2.66E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	5.1005E+00	5.60E-01	0.00E+00	0.00E+00
6.95E-02	3.48E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	4.2605E+00	1.40E+00	0.00E+00	0.00E+00
7.06E-02	3.53E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	4.2605E+00	1.40E+00	0.00E+00	0.00E+00
1.07E-01	5.37E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	2.8605E+00	2.80E+00	0.00E+00	0.00E+00
1.10E-01	5.49E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	2.8605E+00	2.80E+00	0.00E+00	0.00E+00
1.57E-01	7.86E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	1.4605E+00	4.20E+00	0.00E+00	0.00E+00
1.48E-01	7.42E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	1.4605E+00	4.20E+00	0.00E+00	0.00E+00
1.39E-01	6.94E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	6.0500E-02	5.60E+00	0.00E+00	0.00E+00
1.39E-01	6.94E-03	0.01	5.60E+00	6E-02	5.00E-04	0.00E+00	6.0500E-02	5.60E+00	0.00E+00	0.00E+00
1.20E-01	6.02E-03	0.01	1.00E-02	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	0.00E+00	1.00E-02	0.00E+00

4.86E+01	2.43E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	4.20E+00	0.00E+00	1.40E+00
1.02E+02	5.12E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.60E+00	0.00E+00	0.00E+00
9.88E+01	4.94E+00	0.01	5.60E+00	0.00E+00	5.00E-04	0.00E+00	5.0000E-04	5.60E+00	0.00E+00	0.00E+00
8.91E-03	4.45E-04	0.01	1.000E-02	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	1.000E-02
8.92E-03	4.46E-04	0.01	1.000E-02	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	1.000E-02
9.37E-03	4.69E-04	0.01	1.000E-01	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	1.000E-01
9.54E-03	4.77E-04	0.01	1.000E-01	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	1.000E-01
1.27E-02	6.37E-04	0.01	5.000E-01	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	5.000E-01
1.30E-02	6.49E-04	0.01	5.000E-01	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	5.000E-01
1.69E-02	8.47E-04	0.01	1.000E+00	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	1.000E+00
1.69E-02	8.44E-04	0.01	1.000E+00	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	1.000E+00
2.73E-02	1.36E-03	0.01	2.250E+00	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	2.250E+00
2.69E-02	1.35E-03	0.01	2.250E+00	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	2.250E+00
4.44E-02	2.22E-03	0.01	4.500E+00	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	4.500E+00
4.45E-02	2.22E-03	0.01	4.500E+00	6.00E-02	5.00E-04	0.00E+00	6.05E-02	0.00E+00	0.00E+00	4.500E+00
9.18E-03	4.59E-04	0.01	9.400E-03	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	9.400E-03
9.12E-03	4.56E-04	0.01	9.400E-03	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	9.400E-03
9.66E-03	4.83E-04	0.01	9.400E-02	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	9.400E-02
9.67E-03	4.84E-04	0.01	9.400E-02	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	9.400E-02
1.33E-02	6.63E-04	0.01	4.700E-01	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	4.700E-01
1.33E-02	6.64E-04	0.01	4.700E-01	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	4.700E-01
1.76E-02	8.79E-04	0.01	9.400E-01	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	9.400E-01
1.76E-02	8.79E-04	0.01	9.400E-01	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	9.400E-01
2.80E-02	1.40E-03	0.01	2.162E+00	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	2.162E+00
2.78E-02	1.39E-03	0.01	2.162E+00	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	2.162E+00
4.64E-02	2.32E-03	0.01	4.230E+00	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	4.230E+00
4.64E-02	2.32E-03	0.01	4.230E+00	6.00E-02	0.00E+00	0.00E+00	6.00E-02	0.00E+00	0.00E+00	4.230E+00

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