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Physical properties as indicators of liquid compositions: Derivation of the composition for Titan's surface liquids from the *Huygens* SSP measurements

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ABSTRACT

We present a method for inferring the relative molar abundance of constituents of a liquid mixture, in this case methane, ethane, nitrogen and argon, from a measurement of a set of physical properties of the mixture. This problem is of interest in the context of the *Huygens* Surface Science Package, SSP, equipped to measure several physical properties of a liquid in case of a liquid landing on Saturn's moon Titan. While previous models emphasized the possibility of *verifying* a certain model proposed by atmospheric composition and equations of state, we use an inverse approach to the problem, i.e. we will infer the liquid composition strictly from our measurements of density, refractive index, permittivity, thermal conductivity and speed of sound. Other a priori information can later be used to improve (or reject) the model obtained from these measurements.

Key words: instrumentation: miscellaneous – methods: numerical – techniques: miscellaneous – planets and satellites: individual: Titan.

1 INTRODUCTION

The *Huygens* probe, released from the *Cassini* orbiter to descend into the atmosphere of Saturn's large moon Titan on 2005 January 14, had the objective to analyse the atmospheric constituents, physical properties and dynamics before impacting the surface. During the descent, the instruments on board *Huygens* performed optical and IR imaging and spectrometry and also recorded atmospheric composition (including aerosols), temperature, pressure, winds and electrical properties.

1.1 Liquid surfaces on Titan

The nature of Titan's surface remains an open question until the arrival of the *Huygens* probe because Titan's distance and the haze in the atmosphere limit remote observations. A deep, global ocean of hydrocarbons was proposed by Flasar (1983) and Lunine, Stevenson & Yung (1983), based on photochemical models and the abundance of methane in the atmosphere. This view clearly influenced the design of the *Cassini–Huygens* mission. More than a decade later, a global ocean is ruled out by ground-based maps (e.g. Smith et al. 1996), but Lara, Lorenz & Rodrigo (1994) stress the need for a photochemical reservoir – either in the form of subsurface storage in regolith, as presented by Kossacki & Lorenz (1996), or in lakes and seas. Recently, specular radar reflections from 12 out of 16 sites examined by Campbell et al. (2003) have indicated smooth surfaces on Titan, consistent with hydrocarbon seas. Imaging of Titan during recent flybys shows a very diverse surface, which does not rule out liquids.

1.2 Scientific objectives of *Huygens* SSP

The *Huygens* Surface Science Package (SSP) consists of nine independent sensors. Its primary objective is to characterize the surface *Huygens* impacts after its descent. Seven of the sensors are mounted in a cavity in the foredome of the *Huygens* probe. The main objectives, given by Zarnecki et al. (2002), are to:

(i) determine the physical nature and condition of Titan's surface at the landing site;
(ii) determine the abundances of the major ocean constituents, placing bounds on atmospheric and ocean evolution;
(iii) measure the thermal, optical, acoustic and electrical properties and density of any ocean, providing data to validate physical and chemical models;
(iv) determine wave properties and ocean/atmosphere interactions;
(v) provide ground truth for interpreting large-scale Orbiter Radar Mapper and other experimental data.

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1.3 Aim of this work

The method in this work was developed with the *Huygens* SSP experiment in mind, which is why we start with the assumption that the *Huygens* probe will land in a liquid that is a mixture of methane, ethane, nitrogen (and argon). The applicability of our method, however, is not limited to this experiment, nor is it limited to this particular mixture of constituents. It can be seen as a general method to determine quantitatively the composition of a liquid from measurements of its physical properties, given constraints on its potential components. We will show how, under certain conditions, a reliable model of constituents composing the liquid can be derived from the physical properties, for example as measured by SSP's sensors. For this purpose, we approximate equations of state and present an algorithm to find the desired solution. The robustness of our method will be demonstrated in a few test cases.

2 PHYSICAL PROPERTIES OF MIXTURES

We need to specify which physical properties are measured before we elaborate the mixing rules used.

2.1 Measuring physical properties: the SSP sensors

SSP measures five physical properties of its environment: speed of sound, density, permittivity, refractive index and thermal conductivity. Apart from that, two accelerometers and a tilt sensor record the dynamics of the descent and impact, and a sounder will determine the depth of any surface liquid. Table 1 shows a listing of all sensors.

2.2 Linking physical properties and composition: mixing rules

In the following we shall present equations to approximate the properties of a liquid mixture of n components. The quantities to be determined are the mole fractions $\boldsymbol{x} = [x_1, \ldots, x_n]$ of the n constituents of the mixture. The volumetric fractions of the individual components are denoted by $\phi = [\phi_1, \ldots, \phi_n]$. The relation between mole fractions and volumetric fractions is approximated by Amagat's law

$$\phi_i = \frac{x_i V_i}{\sum_j x_j V_j}, \tag{1}$$

where V_i is the molar volume of component i at the pressure and temperature of the measurement and x_i is the mole fraction of component i.

2.2.1 Density

In a first approximation, the bulk density ρ of our mixture is given by

$$d_1(\phi) = \rho = \sum_i \rho_i \phi_i, \tag{2}$$

where ρ_i is the density of the pure substance i. We assume pressure and temperature to be known and constant.

2.2.2 Refractive index

Badoz et al. (1992) assumed a linear relationship between the refractive index n of the mixture and those of the pure components n_i such that $n = \sum_i n_i x_i$. An expression in terms of volume fractions

$$d_2(\phi) = n = \sum_i n_i \phi_i \tag{3}$$

is used in our case. This approximation is in slightly better agreement with the data of Badoz et al. (1992) than their model, provided the right expressions are used for density of dissolved nitrogen (cf. appendix A).

2.2.3 Thermal conductivity

Reid, Prausnitz & Sherwood (1977) present the Li equation as a possible way of approximating the bulk thermal conductivity of a mixture of liquids. According to this, the bulk thermal conductivity λ can be expressed in terms of the thermal conductivity of the pure components $\lambda_{i,j}$ by

$$d_3(\phi) = \lambda = \sum_i \sum_j \phi_i \phi_j \lambda_{ij}, \tag{4}$$

with

$$\lambda_{ij} = \frac{2}{1/\lambda_i + 1/\lambda_j}.$$

2.2.4 Electrical permittivity

To date, there are only very few measurements of electrical permittivity available at low temperatures. On the other hand, permittivity data for ethane, methane, nitrogen and argon under normal conditions are abundant. Using the Clausius–Mosotti equation, these values can be inserted into a density-dependent equation of the form

$$d_4(\phi) = \epsilon = \frac{-3}{\left(\sum_i \phi_i E_i^{\mathrm{NB}} P_i\right) - 1} - 2, \tag{5}$$

Table 1. SSP sensor subsystems.

Acronym	Sensor	Description
	Physical property related	
API–V	Acoustic properties – speed of sound	Pair of piezoelectric transducers
DEN	Density	Archimedes principle float
PER	Permittivity	Parallel plate capacitor to determine electrical permittivity and conductivity
REF	Refractometer	Critical angle refractometer
THP	Thermal properties	Transient hot-wire sensor to determine temperature and thermal conductivity
	Dynamics related	
ACC–E	External accelerometer	Force transducer to characterize impact
ACC–I	Internal accelerometer	Accelerometer to characterize descent and impact
API–S	Acoustic properties – sounder	Array of piezoelectric sensors, 'sonar' principle
TIL	Tilt	Electrolytic tilt sensor (2 axis)

with

$$E_i^{\mathrm{NB}} = \left.\frac{\epsilon_i - 1}{\epsilon_i + 2}\right|_{293\,\mathrm{K},\ 1013\,\mathrm{mbar}}$$

and

$$P_i = \frac{\left.\rho_i^{\mathrm{liquid}}\right|_{T,p}}{\left.\rho_i^{\mathrm{gas}}\right|_{293\,\mathrm{K},\ 1013\,\mathrm{mbar}}},$$

where T and p are the temperature and pressure at the point where the measurement takes place. As all components considered in our model consist of non-polar molecules, this is a valid approximation, as pointed out by Sen, Anicich & Arakelian (1992). It should also be pointed out that the electrical permittivity of non-polar substances ideally equals the square of its refractive index.

2.2.5 Speed of sound

No previous work on the speed of sound of liquid mixtures could be found. We derived a linear mixing rule by comparing its value for c_{s} against the values given by the equations of state of Peng & Robinson (1976), using the National Institute of Standards and Technology (1993) physical property data base. Values were in reasonably good agreement, within a few per cent, if the following equation was used:

$$d_5(\phi) = c_{\mathrm{s}} = \sum_i c_i \phi_i. \tag{6}$$

2.2.6 Mixture of n components

As an additional constraint, we assume that the volume fraction of substances other than the n components whose physical properties are known is negligible, i.e.

$$d_6(\phi) = 1 = \sum_i \phi_i. \tag{7}$$

3 OBTAINING COMPOSITIONS FROM PROPERTIES

Having set up rules that link physical properties and composition, the following method can be used to infer a composition from a set of measurements.

3.1 Algorithm

For the sake of simplicity, we ignore errors for the time being; we will come back to this topic at the end of the section. Assume we have the k measurements

$$\boldsymbol{d}_{\mathrm{m}} = (\hat{d}_1, \hat{d}_2, \ldots, \hat{d}_k) \tag{8}$$

of the physical properties of our liquid. We seek the root of

$$\boldsymbol{F} := \boldsymbol{d}_{\mathrm{p}} - \boldsymbol{d}_{\mathrm{m}} = \boldsymbol{0}, \tag{9}$$

where

$$\boldsymbol{d}_{\mathrm{p}} = (d_1(\boldsymbol{x}, p, T), d_2(\boldsymbol{x}, p, T), \ldots, d_k(\boldsymbol{x}, p, T)) \tag{10}$$

are physical properties that are predicted based on an assumed composition of molar fractions $\boldsymbol{x}$. These properties are of course valid for a certain pressure p and temperature T, but these are measured independently and can be taken as constants, provided that neither is changed by the measurements (and that all measurements are performed in the same ambient conditions). We can solve equation (9) iteratively. Assume we start at a 'first guess' composition of $\boldsymbol{x}_0$ and we want to improve our first guess. Then with

$$\boldsymbol{x} = \boldsymbol{x}_0 + \Delta\boldsymbol{x} \tag{11}$$

we obtain

$$\boldsymbol{F}(\boldsymbol{x}) = \boldsymbol{F}(\boldsymbol{x}_0) + \frac{\partial \boldsymbol{F}}{\partial \boldsymbol{x}}(\boldsymbol{x}_0) \cdot \Delta\boldsymbol{x}. \tag{12}$$

The residual between our property measurements and the predicted properties from our first-guess composition is

$$\boldsymbol{e} = -\boldsymbol{F}(\boldsymbol{x}_0) = \boldsymbol{d}_{\mathrm{m}} - \boldsymbol{d}_{\mathrm{p}}(\boldsymbol{x}_0). \tag{13}$$

If the functional shape of $\boldsymbol{d}_{\mathrm{p}}$ as presented by the equations in Section 2.2 is used, the original (non-linear) problem is reduced to a linear problem where the Jacobian

$$\boldsymbol{J} := \frac{\partial \boldsymbol{F}}{\partial \boldsymbol{x}} = \frac{\partial \boldsymbol{d}_{\mathrm{p}}}{\partial \boldsymbol{x}} \tag{14}$$

can in fact be derived analytically, as we shall show below. The operation in equation (11) can be repeated. We arrive at the iterative solution

$$\boldsymbol{x}^{(i+1)} = \boldsymbol{x}^{(i)} + \Delta\boldsymbol{x}^{(i)}, \tag{15}$$

where, in the simple case of exact measurements without errors, we have

$$\Delta\boldsymbol{x}^{(i)} = (\boldsymbol{J}^{\mathrm{T}}\boldsymbol{J})^{-1}\boldsymbol{J}^{\mathrm{T}}\boldsymbol{e}. \tag{16}$$

Equation (16) demands that $\boldsymbol{J}$ exists and its rank be at least equal to n, which will generally be the case unless the physical properties of the pure components are very similar. Otherwise a regularized solution will have to be sought. The above solution can be extended to include a weighting matrix $\boldsymbol{W}$ for the individual measurements (in fact, the whole iterative procedure can be derived assuming noisy data, as Tarantola & Valette (1982) have shown). It is quite common to use the covariance matrix as weighting matrix $\boldsymbol{W}$. If we ignore the quadratic correlation between dielectric permittivity and refractive index, we can use $\boldsymbol{W}$ to account for the variances σ_i of the individual measurements. With

$$W_{ij} = \delta_{ij}\sigma_i^{-2}, \tag{17}$$

where δ_{ij} is the Kronecker delta and σ_i is the variance of measurement $\hat{d}_i$, we can replace (16) by

$$\Delta\boldsymbol{x}^{(i)} = (\boldsymbol{J}^{\mathrm{T}}\boldsymbol{W}\boldsymbol{J})^{-1}\boldsymbol{J}^{\mathrm{T}}\boldsymbol{W}\boldsymbol{e}. \tag{18}$$

3.2 Derivatives of mixing rules and the Jacobian

As we have seen in Section 2.2, most physical properties of liquid mixtures are expressed as functions of volume fraction rather than mole fraction. We shall account for this by putting

$$\boldsymbol{J} = \frac{\partial \boldsymbol{d}_{\mathrm{p}}}{\partial \boldsymbol{x}} = \frac{\partial \boldsymbol{d}_{\mathrm{p}}}{\partial \phi} \cdot \frac{\partial \phi}{\partial \boldsymbol{x}}. \tag{19}$$

Inserting the equations of Section 2.2, we get

$$\frac{\partial d_1}{\partial \phi_i} = \rho_i, \tag{20}$$

$$\frac{\partial d_2}{\partial \phi_i} = n_i, \tag{21}$$

$$\frac{\partial d_3}{\partial \phi_i} = \sum_j \phi_j \lambda_{ij}, \tag{22}$$

$$\frac{\partial d_4}{\partial \phi_i} = \frac{3E_i^{\mathrm{NB}} P_i}{\left[\left(\sum_i \phi_i E_i^{\mathrm{NB}} P_i\right) - 1\right]^2}, \tag{23}$$

$$\frac{\partial d_5}{\partial \phi_i} = c_i, \tag{24}$$

and

$$\frac{\partial d_6}{\partial \phi_i} = 1 \tag{25}$$

for the first factor of the Jacobian, and with Amagat's law (1), we have

$$\frac{\partial \phi_i}{\partial x_j} = \frac{\delta_{ij} V_j \sum_\ell x_\ell V_\ell - x_i V_i V_j}{\left(\sum_\ell x_\ell V_\ell\right)^2} \tag{26}$$

for the second factor, where δ_{ij} is the Kronecker delta. Once we have inserted the derivatives (20)–(25) as well as (26) into the Jacobian (19), we can use (18) and (15) to find our solution.

3.3 Sensitivity and errors

As can be seen from (19), the sensitivity of our solution, i.e. the dependence of our final derived composition on the physical properties measured, can be expressed in a straightforward way: if our data vector is changed by an amount $\delta\boldsymbol{d}$, then, according to (18), this affects our solution $\boldsymbol{x}$ by

$$\delta\boldsymbol{x} = (\boldsymbol{J}^{\mathrm{T}}\boldsymbol{W}\boldsymbol{J})^{-1}\boldsymbol{J}^{\mathrm{T}}\boldsymbol{W}\,\delta\boldsymbol{d}. \tag{27}$$

Similarly, $\delta\boldsymbol{d}$ can be conceived as an error. The Jacobian allows for a mapping of measurement errors into errors in derived composition. With the help of (27), one can predict the impact of measurement errors upon the inferred composition reliably.

The robustness of our method was checked in Monte Carlo simulations, feeding the algorithm with measurements containing Gaussian errors (see Section 4.3). The error distribution of the derived composition also turned out to be Gaussian, as one would expect from a robust optimization procedure.

4 TEST CASES

We will present a few test cases to demonstrate the applicability and robustness of our method.

4.1 Convergence

To demonstrate the convergence of our algorithm, we show the result of a simulation in which the composition of a ternary mixture is derived in Fig. 1. We used a mixture of 40 per cent methane, 50 per cent ethane and 10 per cent nitrogen at 94 K and 1.5 bar. Physical properties are simulated and error-free. Our first-guess composition is that of equal fractions of all components. We arrive at an acceptably low residual in the 1 per cent range within a few dozen iterations.

4.2 Ternary mixture with measurement errors

Of course, one needs to test the robustness of the method when measurements contain errors. The concept of errors can be applied not only to the measurements, but also to predictions of physical properties from equations of state, because even sophisticated equations of state can contain considerable errors, as Reid et al. (1977) have pointed out. To test the stability of our method, we tested it with a combination of experimental data (where available) and data given by the National Institute of Standards and Technology (1993) physical properties data base. Both types of input into our algorithm almost certainly do not follow the simple mixing rules pointed out earlier, which is why we can consider these data as realistic simulants of measurements with a combination of systematic and random errors. We give the results of some of these experiments in Table 2. Note that the actual and derived composition agree quite well. We need to point out that the iterative procedure is essentially unbounded; therefore it allows fractions of more than 100 per cent as well as negative fractions. One could impose a strict boundary, but we found the unbounded approach very practical as it easily allows the identification of conflicts in our measurements or assumptions. Such conflicts may include the presence (to a significant level) in the mixture of a component not catered for in the solution, the presence in the solution of a component not in the mixture, or unforeseen offsets in the measurements. The identity of the anomalous component(s) and the magnitude and sign of exo-boundary solutions may also serve to indicate how the set of components available to the solution may be modified to give a physical answer. Here, the objective would be to find an alternative to component *j* with greater/smaller property P_j. This is more informative than a 'forced' zero, which would be ambiguous – it may indicate either a real solution, a need for a modified set of components, or a spurious set of measurements. The cause for the negative nitrogen abundance in the first line of Table 2 belongs to the category of unforeseen offsets in the measurements. Here, both refractive index and electrical permittivity fed into our algorithm were higher than expressed by the mixing rules above. Among the components, nitrogen has the lowest refractive index and electrical permittivity, and ethane the highest, which explains the final solution.

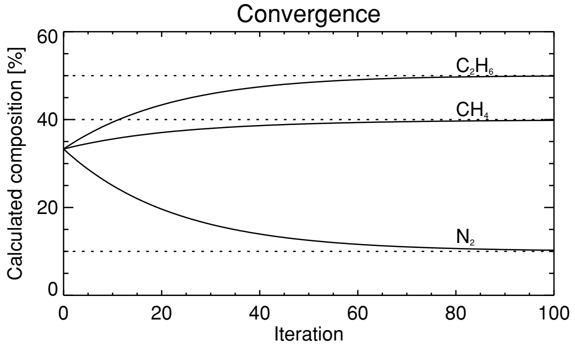

Figure 1. Convergence of the iterative procedure to recover the composition of a ternary mixture. The starting point assumes equal fractions for the three components.

4.3 Mixture of four components

With a total of 5(+1) measurements one would assume that more than three components of a mixture can be identified, provided that the physical properties of the components differ sufficiently for equation (16) to be not too ill-posed. In the context of the *Huygens* mission, argon has been proposed as a possible fourth component of the liquid on Titan. We can include argon in our model and solve for a four-component mixture. Because there are no experimental data of such a mix available, we performed Monte Carlo simulations by adding Gaussian errors to simulated property measurements. Fig. 2 shows the relative distribution of inferred compositions based on measurements with Gaussian errors with a standard deviation of

Table 2. Test cases to infer the composition of a mixture of methane, ethane and nitrogen at 94 K and 1.5 bar. Physical properties are either taken from the NIST chemical property data base or experimental values determined by Badoz et al. (1992)[1] and Birchley (1992)[2]. Note the occurrence of negative fractions that point at minor inconsistencies in our measurements (or our assumptions).

Actual composition			Physical properties				Derived composition			
C_2H_6	CH_4 [vol. per cent]	N_2	n (at 670 nm)	ρ [g cm^{-3}]	λ [W Km^{-1}]	ϵ	c_s [km s^{-1}]	C_2H_6	CH_4 [vol. per cent]	N_2
53	47	0	1.3538[1]	0.5686	0.2478	1.8328	1.8113	59.5	45.0	−4.6
0	100	0	1.2872[1]	0.4474	0.215[2]	1.6569	1.5077	−1.0	99.8	1.2
96	0	4	1.3848[1]	0.6530	0.2478	1.9177	1.9065	102.6	−3.2	0.6
50	44	6	1.3445[1]	0.5807	0.2398	1.8077	1.7476	56.7	41.4	1.9
0	83	17	1.2724[1]	0.4998	0.1955	1.6190	1.3107	−0.4	82.6	17.7

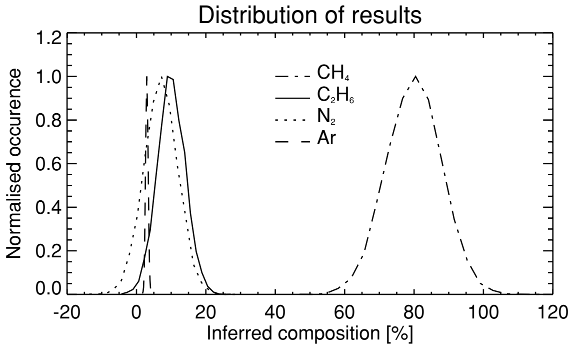

Figure 2. Normalized distribution of inferred results from simulated measurements containing Gaussian errors. Our input composition contains 80 per cent CH_4, 10 per cent C_2H_6, 7 per cent N_2 and 3 per cent Ar.

3 per cent of the total spread (cf. Table 3). The inferred compositions have near-Gaussian distributions, which proves the robustness of our method. The large variances, however, indicate that we are dealing with an ill-posed problem in that uncertainties in the measurement domain are inflated in the composition domain. The small variance of the argon fraction can be explained by its favourable combination of physical properties: it has a relatively high density while all other properties are quite similar to those of the carbohydrates, making an unambiguous identification easy.

4.4 Identification of poor solutions

In Section 4.2 we have shown how, in some cases, poor solutions – meaning solutions with a large residual – are obtained. If the 'real' composition is not known, the individual values of the residuals will point at unreliable measurements. A threshold needs to be set for an inferred composition that is considered a 'good' fit. In line with the chi-squared approach, we propose

$$q = \sum_{i=1}^{k} \frac{e_i^2}{\sigma_i^2} \tag{28}$$

as a measure for reliability of the result, where e_i is the residual between the final solution and the i-th measurement. Usually, values of $q < 1$ indicate a reasonably good agreement of the measurements and the inferred composition of the liquid.

Table 3. Range of physical properties of the pure constituents of our mixture and resulting variances.

Sensor	Max.	Min.	Std. deviation	
DEN	1.3527	0.4474	0.027	(g cm^{-3})
REF	1.3887	1.1985	0.006	—
THP	0.2517	0.1147	0.041	(W Km^{-1})
PER	1.9285	1.4364	0.015	—
API–V	1.9760	0.6511	0.04	(km s^{-1})

5 CONCLUSIONS

We have presented a method of recovering the composition of a liquid mixture from measurements of its physical properties, a problem which emerged in the development of the *Huygens* Surface Science Package. Our method is based on a few analytically simple approximations of physical properties and an assumption as to what the potential components are. We have tested the method using both experimental and simulated data. This method was to be used for SSP data analysis in case of a liquid landing on Titan, but it is applicable to other experimental scenarios, too. In the case of Titan, equilibrium models of atmosphere–liquid interactions can be used to constrain our model further or pinpoint measurements with large errors. Considering that most of the sensors required for the physical property measurements described in this work are comparatively inexpensive, application in other fields where chemical analyses are not feasible could be considered. A possible application could be the measurement of the atmospheric composition above the Martian polar caps which changes due to the seasonal condensation of CO_2, resulting in an enrichment in argon and nitrogen (Lorenz 2000). A combination of measurements of speed of sound and thermal conductivity could identify any argon or nitrogen enrichments without the need for a large instrument like a mass spectrometer.

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APPENDIX A: A NOT SO RIGOROUS TREATMENT OF PHYSICAL PROPERTIES

At the time of writing, a pressure of approximately 1.5 bar and a temperature of 94 K appear to be the best assumptions for the ambient conditions the *Huygens* probe will encounter.

As has been mentioned in Section 1.1, probable compositions of a liquid mixture comprise a certain quantity of argon and nitrogen. However, both are gaseous in their pure form at Titan's ambient conditions, therefore their physical properties cannot simply be inserted into our equations. For our purposes, we found it satisfactory to extrapolate the physical properties of N_2 or Ar from the properties found at the same pressure below the boiling point. This is admittedly only a first-order approximation, but in our tests this approximation turned out to be sufficiently precise. This is not surprising, as the mole fractions of argon and nitrogen are not expected to exceed a few per cent. In the absence of laboratory data we derived the following empirical equations for the physical properties at a pressure of 1.5 bar from the NIST data base (National Institute of Standards and Technology 1993).

A1 Nitrogen

$$\rho(T)|_{1.5\,\mathrm{bar}} \approx 1.143 - 0.004\,32T\ [\mathrm{g\,cm^{-3}}] \tag{A1}$$

$$c(T)|_{1.5\,\mathrm{bar}} \approx \exp(8.551 - 0.022T)\ [\mathrm{m\,s^{-1}}] \tag{A2}$$

$$\lambda(T)|_{1.5\,\mathrm{bar}} \approx 0.229 - 0.001\,214T\ [\mathrm{W\,Km^{-1}}] \tag{A3}$$

A2 Argon

$$\rho(T)|_{1.5\,\mathrm{bar}} \approx 1.929 - 0.006\,131T\ [\mathrm{g\,cm^{-3}}] \tag{A4}$$

$$c(T)|_{1.5\,\mathrm{bar}} \approx 1060.8907 - 2.835\,35T\ [\mathrm{m\,s^{-1}}] \tag{A5}$$

$$\lambda(T)|_{1.5\,\mathrm{bar}} \approx 0.275\,4836 - 0.001\,685\,05T\ [\mathrm{W\,Km^{-1}}] \tag{A6}$$

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