Multiphysics Simulation of Seebeck Coefficient Measurement

K. Huang1, F. Edler1

Received: 27 August 2021 / Accepted: 7 March 2022 / Published online: 31 March 2022
© The Author(s) 2022

Abstract
Using the finite element method (FEM), we developed a simulation model based on our actual apparatus, which is used for measuring the Seebeck coefficient and the electrical conductivity of thermoelectric materials. In terms of geometry, materials, and physics, our simulation model corresponds to the essential details of the experimental device. This article describes the model in detail and highlights key technical characteristics. By comparing it with actual measurement results, the accuracy of the model has been verified. Based on this accurate model, a simulation comparison study on the changes of certain parameters was carried out. Finally, the results of these comparisons are discussed to gain a deeper understanding of the actual experimental process and to improve the accuracy of the measurement.

Keywords
Seebeck coefficient · finite element method (FEM) · cold finger effect (CFE) · dynamic and steady-state method

Introduction
The Physikalisch-Technische Bundesanstalt (PTB) uses its modified measuring system, SR3 (S = Seebeck coefficient; R = resistivity; 3 = version number), developed and constructed at Fraunhofer Institute for Physical Measurement Techniques (IPM),1 for the simultaneous measurement of the Seebeck coefficient S and the electrical conductivity σ of thermoelectric bulk materials and thin-film layers.2 With the construction and application of a new measuring insert/sample holder designed at PTB, measurements of the Seebeck coefficients up to approximately 750°C in inert gas atmospheres at pressures between 3000 Pa and 10000 Pa can now be carried out. This was previously only possible up to 600°C. The determination of S is ideally based on measurements of the voltage potential and the corresponding temperature difference over a sample at the same time and at the same positions. The electrical conductivity of the samples is measured using a collinear four-point probe technique.

In order to explore and reduce the uncertainty that affects the measurement of S, we have used COMSOL Multiphysics® software3 to develop a simulation model of the actual measurement arrangement. This paper introduces and validates the model in detail and shows and analyses some preliminary research results.

Construction of the Model
COMSOL Multiphysics® is a mathematical modelling software program used for modelling multiphysics systems. This software package has been successfully tested in the past by simulating thermoelectric legs, thus demonstrating its ability to accurately simulate thermoelectric devices.4,5 Furthermore, other modelling software has been used to perform simulation of multiphysics systems, for instance Ansys®6 and Autodesk Simulation,7 which were used to analyse Seebeck and electrical resistivity measurement systems.

One advantage of using simulation results to study the physical experiment process is that we can easily obtain more physical data at locations without installing many actual physical sensors. This might otherwise affect the original physical fields, for instance temperature and electrical potential. It also allows us to see the results on a time slice with more physics parameters, like S. In theory, it can be used with any physical parameter at any geometric location at any point in time.

K. Huang
kai.huang@ptb.de

1 Physikalisch-Technische Bundesanstalt, Abbestr. 2-12, 10587 Berlin, Germany

© Springer
Components of the Simulation

Geometry

The complete sample holder is shown in Fig. 1. A photo of the main parts of the sample holder of the SR3 measuring system is presented in Fig. 1a, while a plot of the model designed in COMSOL with a semi-perspective view of the sample holder is seen in Fig. 1b. By means of titan hexagon nuts (b), the sample (a) is fixed to two mounting bases (c) made of Inconel 600, which are screwed onto a ceramic plate (d). The ceramic plate (d) is mounted on a metal bracket made of heat-resistant steel 1.4845 (X8CrNi25-21). Two microheaters (e) are inserted in the left and right mounting base (c) in order to generate a variable temperature gradient between 0.5 K and 6 K over the sample (a). Two small replaceable type S thermocouples (f) usable as probes are inserted in the grooves of the ceramic holder (g). The ceramic holder (g) can move in the longitudinal direction on the ceramic plate (d). Two Inconel X750 coil springs (h), guided by the two microheaters, press the ceramic holder (g) with the two type S thermocouples (f) onto the sample (a) to ensure good thermal and electrical contact. The thermocouples are used to measure the temperatures $T_1$ and $T_2$. We can then calculate the average sample temperature $T_{\text{Sample}}$ and the temperature difference $\Delta T = T_1 - T_2$ over the sample. In addition, the two identical wires of the thermocouples are used to measure the voltages $V_{\text{PtRh}}$ and $V_{\text{Pt}}$, which are used to calculate the Seebeck coefficient $S_X$ of the sample together with $\Delta T$. The temperature of the reference junctions of the thermocouples $T_{\text{Ref}}$ is measured with a calibrated Pt-100 resistance thermometer (i). The reference junctions (j) of the type S thermocouples are in the hot zone of the sample holder (g) as well. The advantage of this arrangement is that the sample temperature and the temperature of the reference junctions are of the same order of magnitude. In this way, large temperature gradients along the thermocouple wires are avoided. Such an arrangement can therefore generally minimise the error-causing influences of unavoidable thermoelectric inhomogeneities in thermowires.
(when exposed to temperature gradients) on the sample temperature measurement. This enables a traceable determination of $T_{\text{Sample}}$ by means of the calibrated Pt-100. The measured voltages are transmitted from the heated zone via gold wires (k) and connectors to the room temperature zone.

The different measurement temperatures in the range between room temperature and 750°C are achieved by using a spiral tube heater (900 W) that surrounds the entire sample holder. The complete measurement insert of the SR3 device, which is inserted vertically into the measuring chamber, is shown in Fig. 2. It consists of the sample holder (covered by the main heater), the power supply lines, the signal lines, and the flange with the lead-throughs.

Large and complex three-dimensional (3D) geometries are often computationally expensive. The structure of the whole SR3 is very complicated and not easy to simulate for this reason. The size of the measuring chamber is 520 mm in height and 170 mm in diameter. In contrast, the minimum geometric size is the diameter of the thermocouple wires, which is only 0.08 mm. The huge geometric differences lead to large complex meshes. In order to perform the simulation in a reasonable time, the simulation model needs to be simplified wisely. For instance, ten titan screws, the two spiral springs, six titan bolts, and the parts of the two microheaters other than their heads are ignored, since they have only negligible influence, i.e. on the heat losses. In the simulation, the voltage potentials of the two thermocouples could be calculated directly. Therefore, the four gold signal lines and the related connectors do not have to be considered for the simulation. Moreover, they are outside the main heating area of the main heater and therefore also have little influence on the thermal performance of the device. The cross section of the thermocouple wire is a square instead of a circle to reduce the number of grids and improve the quality of the thermocouple wire’s mesh. All the other parts are designed according to the actual size of the SR3 device. The simplified part of the sample holder is given in Fig. 1b. Through these actions, the calculation time for solving the model is shortened considerably, and the memory requirements of the computer are reduced. Such reasonable simplifications of the geometry when building a simulation model, which do not significantly influence the correctness of the overall simulation, are generally useful and recommendable not only for our specific application example, but also for other measuring systems.

**Numerical Model**

In the simulation model, all three forms of heat transfer—convection, conduction, and radiation—need to be considered. Heat conduction occurs when the main heater is heated to transfer the converted heat from it to the measuring chamber. Convection drives a flow inside the vacuum chamber, transferring the heat from the main heater through the chamber via the movement of fluids (in our case, forming gas [95% nitrogen and 5% hydrogen]). According to the Stefan–Boltzmann law, the radiation output of a black body increases as the fourth power of the temperature increases. If the temperature is much higher than the ambient temperature, a large amount of net energy will be transferred from the surface by radiation. The most obvious manifestation of this phenomenon is that, when simulating the highest measured temperature, the control sensor reaches a temperature of about 600°C, while the air outside the measuring chamber is still at 25°C. A considerable part of the energy is transferred through radiation. All three heat transfer forms are modelled by using the heat transfer interface of COMSOL, coupling “heat transfer in solids and fluids” and “surface-to-surface radiation” according to Eqs. 1 and 2, respectively:

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p \vec{u} \cdot \nabla T + \nabla (-k \nabla T) = Q$$

(1)
where $\rho$ is the density, $C_p$ the specific heat capacity at constant pressure, $T$ the absolute temperature, $\vec{u}$ the velocity vector of translational motion, $k$ the thermal conductivity, and $Q$ the heat sources (main heater and microheaters).

\[
J = \varepsilon \sigma T^4 + \rho_d \left( G_m(J) + F_{amb} n^2 \sigma (T_{amb}^4 + G_{ext}) \right) \tag{2}
\]

where $J$ is the surface radiosity, $\varepsilon$ the surface emissivity, $n$ the refractive index of the transparent media, $\sigma$ the Stefan–Boltzmann constant, $\rho_d$ the diffuse reflectivity, $G_m$ the mutual surface irradiation, $F_{amb}$ the ambient view factor, and $G_{ext}$ the external irradiation.

To simulate the performance of the filling gas, the “laminar flow” interface must be coupled with the “heat transfer in solids and fluids” interface. As the forming gas is heated, changes in density and pressure will cause a flow in the chamber. The reference pressure level is set to the value of 5000 Pa. The “compressible flow (Ma < 0.3)” option in COMSOL is selected to simulate natural convection in the measuring chamber. The driving force of natural convection comes from the buoyancy caused by the temperature changes in density and pressure will cause a flow in the chamber. The reference pressure level is set to the value of 5000 Pa. The “compressible flow (Ma < 0.3)” option in COMSOL is selected to simulate natural convection in the measuring chamber. The driving force of natural convection comes from the buoyancy caused by the temperature gradient. The “gravity” feature and the non-isothermal flow interface need to be used at the same time. The combination of these two can simulate the coupled flow field and temperature field under the action of gravitational acceleration. Equation 3 governing non-isothermal flow is the Navier–Stokes equation with gravity. The main heater in the simulation is approximately a solid hollow cylinder, and this approximation ignores any internal influences inside the heating wire.

A “pressure point constraint” is added to avoid uncertain pressure fields, so as to solve the problem of model non-convergence in natural convection problems. It is selected at the top corner of the chamber, away from the area where we are concerned about the flow field. In a steady-state closed system, there is no clear inlet or outlet for the pressure level. In the absence of a reference pressure, Navier–Stokes equations have countless solutions to steady-state problems because they can only solve problems that vary with pressure gradients. Therefore, the pressure point constraint is required to specify the absolute pressure level of the fluid.

\[
\rho \left( \frac{\partial \vec{u}}{\partial t} + \vec{u} \cdot \nabla \vec{u} \right) = \nabla \cdot \left( -p \vec{I} + \mu \left( \nabla \vec{u} + \left( \nabla \vec{u} \right)^T \right) - \frac{2}{3} \mu \left( \nabla \cdot \vec{u} \right) \vec{I} \right) + \vec{F} + \rho \vec{g} \tag{3}
\]

Here, $\rho$ is the fluid pressure, $\vec{u}$ the velocity vector of translational motion, $\vec{I}$ the identity matrix, $\mu$ the fluid dynamic viscosity, $F$ the volume force vector, and $g$ the acceleration of gravity.

The above-mentioned physical conditions of the system, which are used for the initial to the steady state, correspond to “study step 1 – stationary” in COMSOL. They will continue to function in “study step 2 – time-dependent” when the system starts to simulate the process of measuring the Seebeck coefficient. Since our simulation purpose is to simulate the Seebeck coefficient measurement process, the “electric currents” interface must also be included to analyse the electric potential distribution according to Eqs. 4 and 5:

\[
\vec{J} = -\sigma \nabla V + \frac{\partial \vec{D}}{\partial t} + \vec{J} \tag{4}
\]

\[
\nabla \cdot \vec{J} = Q_{j,v} \tag{5}
\]

where $\vec{J}$ is the electric current density, $\sigma$ the conductivity, $V$ the electric potential, $\vec{D}$ the electric displacement field, $\vec{E}$ the externally applied current density, and $Q_{j,v}$ the volumetric source of current.

**Events Interface**

During the measurement process, the boundary condition of the microheater changes instantaneously according to the measurement time schedule, so we introduced the event interface to solve it. This interface models step changes to ensure that the solver accurately captures all step changes in the load. It balances the requirements of accuracy and low calculation cost. When the system reaches a steady state, the measurement process begins. The specific working schedule of the microheater in the simulation will be clarified in Sect. “Dynamic measuring method and steady-state method”. This can be applied to simulate similar hysteresis systems with low computational costs.

**Material Properties**

There are three sources of the attribute values of the various materials used in simulation: our previous research and experimental data, data sheets provided by other manufacturers, and COMSOL’s own library. These include the key attribute values of key materials, which are temperature-dependent material properties—for instance, the Seebeck coefficient of FeSi$_2$ and of Pt, the electrical conductivity of FeSi$_2$ and of Inconel 600, the thermal conductivity of Pt and of Inconel 600, and the surface emissivity of each material.

Different thermoelectric materials can be measured with the SR3 device. However, in this study, only FeSi$_2$ was used as a sample material to perform simulations, so that the influence of different simulation parameter changes on the
simulated and calculated Seebeck coefficient can be analysed well.

The properties of the forming gas in the chamber are only estimations. This is because there are no reliable values of the temperature-dependent heat capacity at constant pressure, density, or thermal conductivity, which are required for simulation, neither in the library of COMSOL, in the datasheets of the other manufacturers, nor in the literature. Therefore, in this study we used the existing nitrogen and hydrogen data in the COMSOL library as the basis and performed algebraic calculations on each required attribute according to the composition ratio of the mixed gas (forming gas). In addition to the simulation with the forming gas, we also ran two simulations as circumstantial evidence. Here we used pure nitrogen and pure hydrogen as a filling gas, instead of a forming gas, for simulations at 300°C. See Sect. “The cold finger effect (CFE)” for the results.

Mesh

The 3D global calculation model established is based on the actual size of a single unit prototype. The substantial difference in the model scales (i.e., the thermocouples, gas domain) requires additional attention during the discretization. The mesh volume is $1.345 \times 10^7$ mm$^3$. This statistic number remains unchanged because it equals the volume of the vacuum chamber. Other statistics will be slightly different according to the subtle changes in the internal geometry of the model. All these custom meshes consist of around 400,000 tetrahedra. The minimum element quality statistics are greater than 0.1, which is recommended by COMSOL. The meshes used for simulations balance element quality and the number of elements, while providing adequate resolution of the geometry.

The Study of the Simulation

One common measurement of the Seebeck coefficient when using the SR3 device starts with the heating of the main heater, so that the control sensor reaches the set temperature and then maintains that temperature. In the simulation, calculations of this process may take longer. A better solution is to set the temperature inside the chamber as the target temperature (600°C) and the outside temperature as the ambient temperature (25°C); see Fig. 3, “Study step 1”, initial state. Adding this stationary study step before starting the time-dependent study steps will first solve the fields under the steady state. This will provide a consistent initial value for the time-dependent steps. In this way, a temperature gradient space is generated, which equals the temperature gradient in the SR3 (Fig 3, “Study step 2”, “Time = 0 s”). This preparation stage includes the stationary step 1 and the first 600 s of step 2 (see Fig. 3). In this way, the computing effort is reduced and the computing time shortened.

There is a large temperature range (100–600)°C in Fig. 3; therefore, the small temperature difference of about 1 mK (see also Fig. 4) cannot be distinguished in the two plots “Time = 0 s” and “Time = 600 s” in “Study step 2”. Before continuing with the experimental stage which is of actual interest, the tolerance refinement study for the simulation model should be performed. This means resolving the model with tighter tolerances and comparing the results. This is because the relative tolerance is related to both the instantaneous error and the error accumulated.
over time. Since errors can accumulate and increase in magnitude, relative tolerances that are too loose will produce erroneous solutions. Resolving the model with a tighter relative tolerance will cause the solver to initially use a smaller time step, but the cumulative error will be smaller, or even negligible. This approach is not application-specific but can be used generally for time-dependent simulations.

The specific approach is as follows: During the first 600 s of the time-dependent study step in the simulation, the main heater maintains its operating power, and the two microheaters have not yet started to work. Theoretically, the model should reach a steady state, just like the SR3, after the control temperature (a type K thermocouple) reaches the set temperature value. Furthermore, the control temperature should fluctuate within a very small temperature range. We use this 600-s temperature stability value to evaluate whether the error of the model has reached what is expected. The stability of the temperature of the control thermocouple is used as an indicator of the achievement of steady-state conditions. The system reaches a steady state when the temperature is stable. This is consistent with the standard of judgment used in the actual experiment.

Figure 4 below shows the influence of different relative tolerance settings. The results of tolerance settings of 0.005 and 0.0005 show a good agreement, and the values are stable. The result with a tolerance setting of 0.05 shows the cumulative calculation error. This tolerance set-up is not suitable for subsequent research. To balance with the accuracy of the model and the saving of computing resources, a tolerance of 0.005 is the best choice for our simulation models (Table I).

### Results and Discussion

#### Seebeck Coefficient

Based on previous research, we used Eq. 6 to calculate the Seebeck coefficient:

\[
S_x = \frac{1}{2} \left( S_p - \frac{S_{TC}}{1 - \frac{\Delta V_{xp}}{\Delta V_{xp}}} + S_n + \frac{S_{TC}}{1 - \frac{\Delta V_{xn}}{\Delta V_{xn}}} \right) \tag{6}
\]

where \(S_x\) is the absolute Seebeck coefficients of the sample, \(S_p\) the absolute Seebeck coefficients of the thermocouple’s positive electrode, \(S_n\) the absolute Seebeck coefficients of the thermocouple’s negative electrode, \(S_{TC}\) the Seebeck coefficient of the thermocouple used, \(\Delta V_{xp}\) the voltage difference measured by using the positive electrodes of the thermocouples, and \(\Delta V_{xn}\) the voltage difference measured by the negative electrodes.

The same data-processing procedure based on this equation was also applied in the simulation. The voltage differences \(\Delta V_{xp}, \Delta V_{xn}\) in Eq. 6 can be directly evaluated by the post-processing function in COMSOL, the simulation software, instead of using a multimeter. The
calculation results and the Seebeck coefficients measured experimentally, as well as their uncertainties for \( k = 2 \), are shown in Table II.

Good agreement between the simulated and measured Seebeck coefficient of FeSi\(_2\) confirms the suitability of the simulation model constructed. Figure 5 shows default plots illustrating the temperature field and the electric potential distribution at 600°C after a simulation time of 700 s.

**Dynamic Measuring Method and Steady-State Method**

A dynamic measuring method\(^1\)\(^1\),\(^1\)\(^1\)\(^2\) is used to eliminate the effects of unavoidable offset voltages in the measuring system when determining the Seebeck coefficient.\(^1\)\(^3\) Since the voltages \( V_{xp} \) and \( V_{xn} \) are measured simultaneously in the simulation at the same temperature difference \( \Delta T \), they can be plotted against each other. The data are fitted by a straight line with the slopes \( dV_{xn}/dT \) or \( dV_{xp}/dT \). These are used together with the known Seebeck coefficients \( S_{typeS} \) of the thermocouple of the type S and the absolute Seebeck coefficients of the thermocouple wire to determine the Seebeck coefficient \( S_X \) of the sample to be calculated directly according to Eq. 6. Unless otherwise specified, the results mentioned in this article are calculated according to this method. The specific settings in the simulation are detailed below. The left microheater starts to work, and after the entire system reaches the thermal equilibrium state, the data recorded thereafter are used in Eqs. 7.1 and 7.2 to calculate \( S_X \). The microheater on the left is heated for 30 min and then shut down.

Figure 6 shows the Seebeck coefficient of FeSi\(_2\) at 300°C calculated from the collected voltage and temperature data every 10 s during the entire simulation. When the left microheater starts heating for 5 min (equivalent to after the 15th minute of the entire simulation process), the system basically reaches the thermal equilibrium state with constant temperature difference, \( \Delta T \) (green line). Within the interval between 15 min and 40 min, the steady-state method to calculate the Seebeck coefficient provided a constant value, \(-216.5 \, \mu\text{V/K}\) (blue line), which is consistent with the results of the
dynamic measurement method, $-216.8 \mu V/K$ (red dot). It is close to the Seebeck coefficient of the sample calculated by the post-processing tool (COMSOL) of the simulation result, $-217.8 \mu V/K$ (orange line). The conclusion is that the Seebeck coefficient obtained by using the dynamic measurement method is the same as the one obtained by using the steady-state method. The dynamic measuring method, however, significantly shortens the measurement time—by omitting the waiting time for the thermal equilibrium.

A test measurement by using the SR3 device confirms the results of the simulation. Three independent measurements were carried out to determine the Seebeck coefficient of a FeSi$_2$ sample at $(302.9 \pm 0.7)^\circ$C using the steady-state method (Eq. 7.1 and 7.2) on basis of the Pt and the Pt10%Rh wires, and the dynamic measuring method according to Eq. 6. Three different $\Delta T$ values with respective sign reversal (right and left miniature heater) were realised. The heating times were 15 min each. The Seebeck coefficients measured are summarised in Table III. The measurement uncertainty of $S$ for $k = 2$ amounts to a value of $\pm 7 \mu V/K$.

**The Cold Finger Effect (CFE)**

The contact of a temperature sensor with a temperature other than the surface temperature of the measurand to be measured inevitably leads to a disturbance of thermal processes at the interface. The resulting so-called CFE and its influence on the surface temperature measurement is very well described in a previous work. In addition, there is also an effect on the measurement of the voltage potential, which has a compensating influence on the measurement of the Seebeck coefficient.

Figure 7 shows the CFE of the sample at $25^\circ$C in the simulation. The temperature gradient (a) and electric potential (b) of the sample surface along the line connecting the two thermocouple beads are shown at 700 s, at the last second of the heating period of the left microheater. It can be seen that the two temperature sensors (type S thermocouples) reduced the temperature of the sample surface, causing the CFE. Furthermore, we found that this CFE also affects the voltage distribution at the sample surface.

![Image](image-url)
In this case, the CFE on temperature and voltage can be estimated using this method: Remove the data points affected by the CFE at 29.62°C, 26.38°C, 184.96 µV, and −404.63 µV, and calculate polynomials to interpolate the resulting gaps by using the undisturbed data points to restore the unaffected conditions (temporarily referred to as the “numerical correction method”). The temperature and electric potential values of these two points are respectively 31.17°C, 26.59°C, 448.13 µV, and −370.04 µV, which are supposed to be undisturbed by the CFE. When substituting these four recalculated temperature and voltage values into the steady-state equation, the Seebeck coefficient of −178.8 µV/K is obtained, which is the Seebeck coefficient value defined in the FeSi₂ material in the simulation at 25°C. Further calculations showed that the CFE has an influence on the temperature difference ΔT of −41.4% and on the electric potential difference ΔV of −38.8%. This resulted in a final experimental simulation error of +1.3% for the Seebeck coefficient in the steady-state method.

If the CFE on the temperature difference ΔT is almost equal to the effect on the voltage potential ΔV, then the calculated Seebeck coefficient is almost error-free. To mitigate the CFE, the microheater holders were equipped with additional sleeves which cover the thermocouples partly as shown in Fig. 8. In this way, additional heat is introduced from the microheaters to the two thermocouples at the same time. At the two positions of the thermocouple beads, the temperature field and the voltage field are still affected by the CFE, but to a lesser extent. The Seebeck coefficient calculated using the original two temperature data values and two voltage data values is −180.0 µV/K. A relative error of 0.1% then results. The Seebeck coefficient calculated by the “numerical correction method” is −179.6 µV/K. This is caused by the CFE which still has an influence on the temperature difference ΔT of −11.6% and on the electric potential difference ΔV of −11.3%.

The size of the sleeves is also important, as shown in Fig. 9. In the extreme case of 8-mm-long sleeves, the

---

**Fig. 8** Compensated CFE by using additional heating sleeves (width 1.5 mm) surrounding the thermocouple probes at 25°C, after 700 s of the simulation: (a) temperature gradient, (b) voltage potential.

**Fig. 9** Overcompensated CFE by using additional heating sleeves (width 8 mm) surrounding the thermocouple probes at 25°C, after 700 s of the simulation: (a) temperature gradient, (b) voltage potential.
Seebeck coefficient calculated by the steady-state method is $-176.2 \, \mu V/K$, which corresponds to a relative error of $-2.1\%$. When applying the “numerical correction method”, the value of $S$ is $-179.7 \, \mu V/K$, which corresponds to the reference value. The influence of the CFE on the temperature difference $\Delta T$ and the electric potential $\Delta V$ is $+17.0\%$ and $+15.3\%$, respectively. Therefore, the 8-mm-long sleeves caused an overcompensation of the CFE by a heat transfer that was too excessive to the probes via the sleeves.

**Different Filling Gases**

The influence of different filling gases on the temperature profile and on the electric potential above the sample was investigated by simulation methods at 300°C when using the optimum heating sleeves of 1.5-mm width. Pure

---

**Fig. 10** Results of simulation when using different filling gases at 300°C, at 700 s of the simulation: (a) temperature gradient in forming gas, (b) voltage potential in forming gas, (c) temperature gradient in H$_2$, (d) voltage potential in H$_2$, (e) temperature gradient in N$_2$, (f) voltage potential in N$_2$. 

$\text{Springer}$
nitrogen and pure hydrogen were chosen as filling gases to compare the results with those obtained when using forming gas as the filling gas that is used regularly when performing measurements with the SR3 device. The calculated temperature distributions and the electric potential curves are summarised in Fig. 10. Additionally, the errors of the Seebeck coefficients of FeSi$_2$ at 300°C resulting from the different gases are listed in Table IV.

The Seebeck coefficient simulation results obtained with H$_2$ and N$_2$ were basically consistent with the simulation results using the forming gas. This is in accordance with the experimental logic: when the thermal contacts between the thermocouple beads and the sample are good, only small amounts of different inert gases to fill the measuring chamber (about 5000 Pa) will not have a big impact on the Seebeck coefficient.

The CFE in pure hydrogen seems to be very small upon first examination of Fig. 10c and d. However, the error of the Seebeck coefficient is larger than the error obtained in both the other gases. This is because the influence of CFE on the temperature difference $\Delta T$ is +0.4%, but the influence on the electric potential difference $\Delta V$ is −0.1%, which results in an error of +1.4 µV/K of the Seebeck coefficient when applying the steady-state method.

The simulation results show that the algebraic method to describe the temperature-dependent properties of the forming gas by using the corresponding data of H$_2$ and N$_2$ is suitable, which can also be transferred to other measuring systems for the measurement of Seebeck coefficients.

Different gases have almost no effect on the final Seebeck coefficient measurement result, but the temperature gradient and electric potential distribution generated on the sample are different. Figure 10 shows these differences at 300°C and 5000 Pa.

The results of the temperature-dependent measurements of the Seebeck coefficient of a FeSi$_2$ sample in nitrogen and forming gas are shown in Fig. 11. Two measurements were carried out at rising and falling temperatures using the dynamic measurement method. The error bars of the individual values correspond to the expanded measurement uncertainty ($k = 2$). The results obtained in both filling gases agree within their measurement uncertainties and correspond to the simulated values.

### Summary and Conclusion

A 3D coupled thermal-fluid-electric FEM model is described. This model was developed to assess the performance of the measurement of Seebeck coefficients of

| Gas                  | Error (dynamic method) (µV/K) | Error (steady-state method) (µV/K) |
|----------------------|-------------------------------|-----------------------------------|
| Forming gas (95% N$_2$ + 5% H$_2$) | +0.7                          | +0.4                              |
| H$_2$                | +2.8                          | +1.4                              |
| N$_2$                | −0.1                          | +0.1                              |

Fig. 11 Measured and simulated values of the Seebeck coefficient of FeSi$_2$ in different filling gases.
FeSi₂ by using the SR3 device. All temperature-dependent material properties were taken into account. Actual measurement results were used to verify the simulation results, and some influence factors on the measurement of Seebeck coefficients were discussed. The following conclusions can be drawn:

(1) The general agreement of the experimental measured Seebeck coefficients of FeSi₂ at different temperatures with the values of S obtained by the simulations performed confirms the correctness and suitability of the COMSOL model constructed.

(2) No significant differences in calculating the Seebeck coefficient were found by using the dynamic measuring method (SR3 device) and the steady-state method, which emphasises the general equality of the two methods.

(3) The influence on the accuracy of Seebeck coefficient measurements of the CFE was shown. Importing heat from the microheater to the thermocouples is a practical solution not only for our specific measuring system SR3. These heat-conducting and heat-transferring sleeves which are optimally directly connected to the heating blocks need to be carefully designed to find a balance between reducing the CFE to an acceptable level and an overcompensating effect.

(4) The impact on the Seebeck coefficient measurements of the forming gas, H₂, and N₂ at pressures of about 5000 Pa is negligible.

Based on these simulation models, further investigations of various influences on the measurement of the Seebeck coefficient and of the electrical conductivity of different thermoelectric materials are planned. The influence of different filling gas pressures may, for instance, be studied.

Acknowledgments This work was accomplished in the course of the “TEST-HT” project (Thermoelectric Standardisation for High Temperatures) in cooperation between the Physikalisch-Technische Bundesanstalt (PTB) and the German Aerospace Center (DLR). The work was financed by the German Federal Ministry of Education and Research under the grant numbers 03VP04402 (PTB) and 03VP04401 (DLR). We would also like to thank Pawel Ziolkowski (DLR) for providing FeSi₂ samples and their physical properties data needed for simulations.

Funding Open Access funding enabled and organized by Projekt DEAL.

Conflict of interest The authors declare that they have no conflict of interest.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by/4.0/.

References

1. Fraunhofer Institute for Physical Measurement Techniques, https://www.ipm.fraunhofer.de/.
2. F. Edler and K. Huang, Analysis of the “cold Finger Effect” in measuring the Seebeck coefficient. Meas. Sci. Technol. (2020). https://doi.org/10.1088/1361-6501/abb38b.
3. COMSOL Multiphysics®, V. 5.4. https://www.comsol.com/. COMSOL AB, Stockholm, Sweden.
4. M. Jaegle, Multiphysics simulation of thermoelectric systems - Model of peltier-cooling and thermoelectric generation, Multiphysics Simul. Thermoelectr. Syst. - Model. Peltier-Cooling Thermoelectr. Gener. 7 (2008).
5. T. Forrister, Studying the Peltier and Seebeck Effects in Thermoelectric Devices, https://www.comsol.com/blogs/studying-the-peltier-and-seebeck-effects-in-thermoelectric-devices.
6. K. Zaborocki, P. Ziolkowski, T. Dasgupta, J. De Boor, and E. Müller, Simulations for the development of thermoelectric measurements. J. Electron. Mater. 42, 2402 (2013).
7. J. Mackey, F. Dynys, and A. Sehirlioglu, Uncertainty analysis for common Seebeck and electrical resistivity measurement. Rev. Sci. Instrum. 85, 085119 (2014).
8. Heraeus, Physical Properties of Pt Materials and Pt DPH Material, n.d.
9. S. M. Deutschland, Inconel Alloy 600, https://www.specialmetals.com/ (n.d.).
10. K. Huang, F. Edler, S. Haupt, P. Ziolkowski, C. Stiewe, and E. Müller, Modelling and uncertainty analysis of Seebeck coefficient measurements by using the finite element method. Mater. Today Proc. 44, 3500 (2019).
11. F. Nakama, A. T. Burkov, A. Heinrich, T. Oyoshi, and K. Yagasaki, Experimental set-up for thermopower and resistivity measurements at 100-1300 K. Int. Conf. Thermoelectr. ICT, Proc. 266 (1998).
12. O. Boffrou, A. Jacquot, A. Dauscher, B. Lenoir, and M. Stölzer, Experimental Setup for the measurement of the electrical resistivity and Thermopower of thin films and bulk materials. Rev. Sci. Instrum. 76, 053907 (2005).
13. F. Edler, E. Lenz, and S. Haupt, Reference material for Seebeck coefficients. Int. J. Thermophys. 36, 482 (2014).
14. J. Martin, T. Tritt, and C. Uher, High temperature Seebeck coefficient metrology. J. Appl. Phys. 108, 14 (2010).
15. J. Martin, Protocols for the high temperature measurement of the Seebeck coefficient in thermoelectric materials. Meas. Sci. Technol. 24, 085601 (2013).

Publisher’s Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.