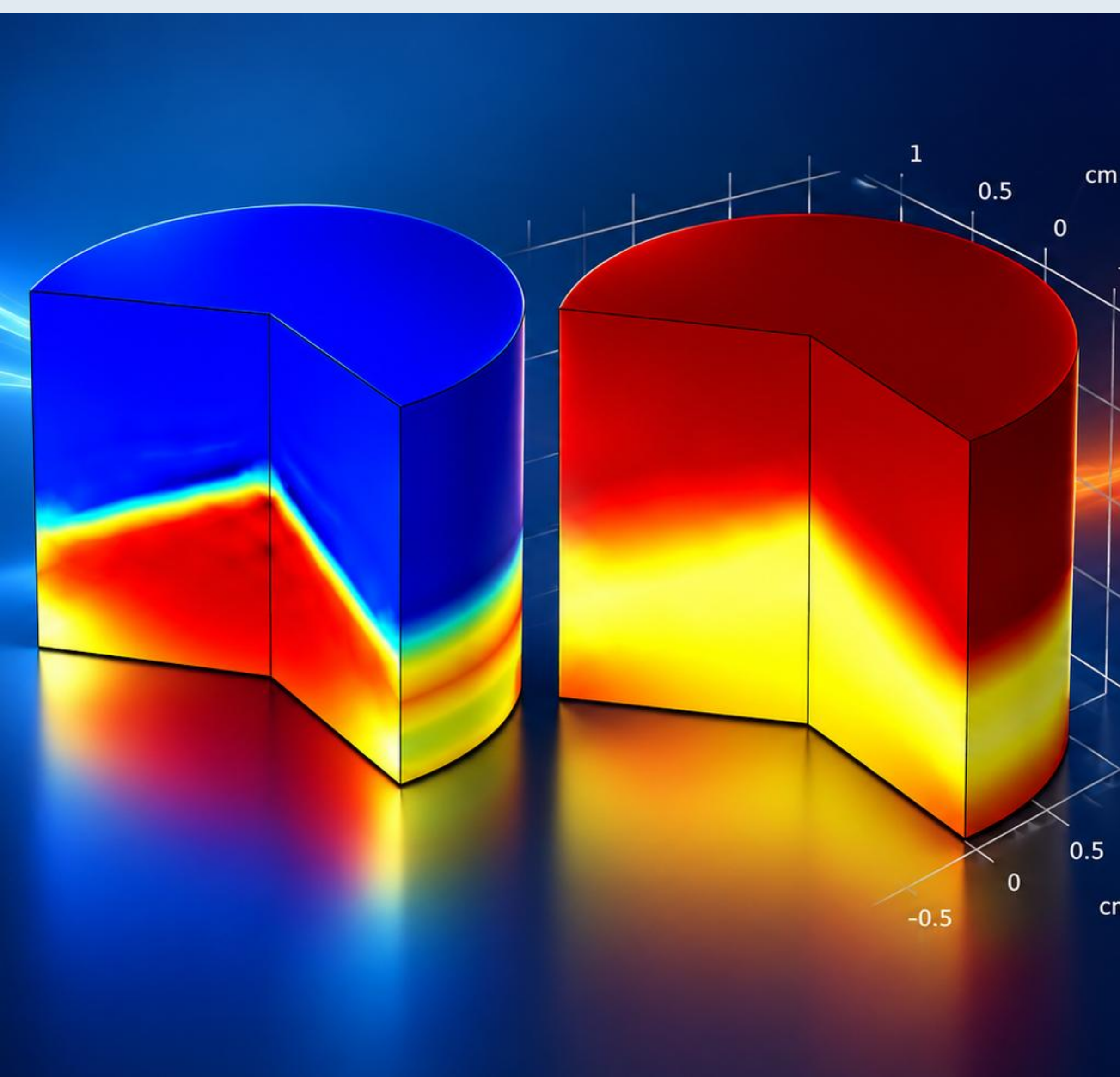




Optimization of SHS Process of Mg_2Si Thermoelectric Materials Synthesis

Promising thermoelectric materials used for waste energy recovery have been identified among semiconducting silicides, selenides and antimonides, which might be produced by Self-propagating High-temperature Synthesis (SHS). 3D modelling is used for rescaling and optimization of the SHS process. Optimization was used to find the solution of the inverse problem allowing for determination of unknown model parameters.

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Introduction & Goals

In modern technology of advanced materials, models play an essential role. They are crucial for understanding complex processes occurring during the synthesis of thermoelectric materials, such as Mg_2Si , used for waste energy recovery. Fabrication by SHS has several advantages: it is very quick; once initiated, the reaction goes by itself without any external source of heat; it produces no waste and overall can be classified as eco-friendly. Despite many advantages of SHS, there are still open questions. COMSOL Multiphysics® is used for modelling the SHS

process in 3D in vacuum. The model considers mass and energy equations, different reaction mechanisms and change of the sample morphology during the SHS process. Suitable boundary conditions correspond to complex, real sample geometry. The model was validated based on thermocouple readings. Sensitivity analysis of the model was performed. The inverse problem was used for determination of unknown values of some model parameters. The validated model can be used for rescaling and optimization of the SHS process.

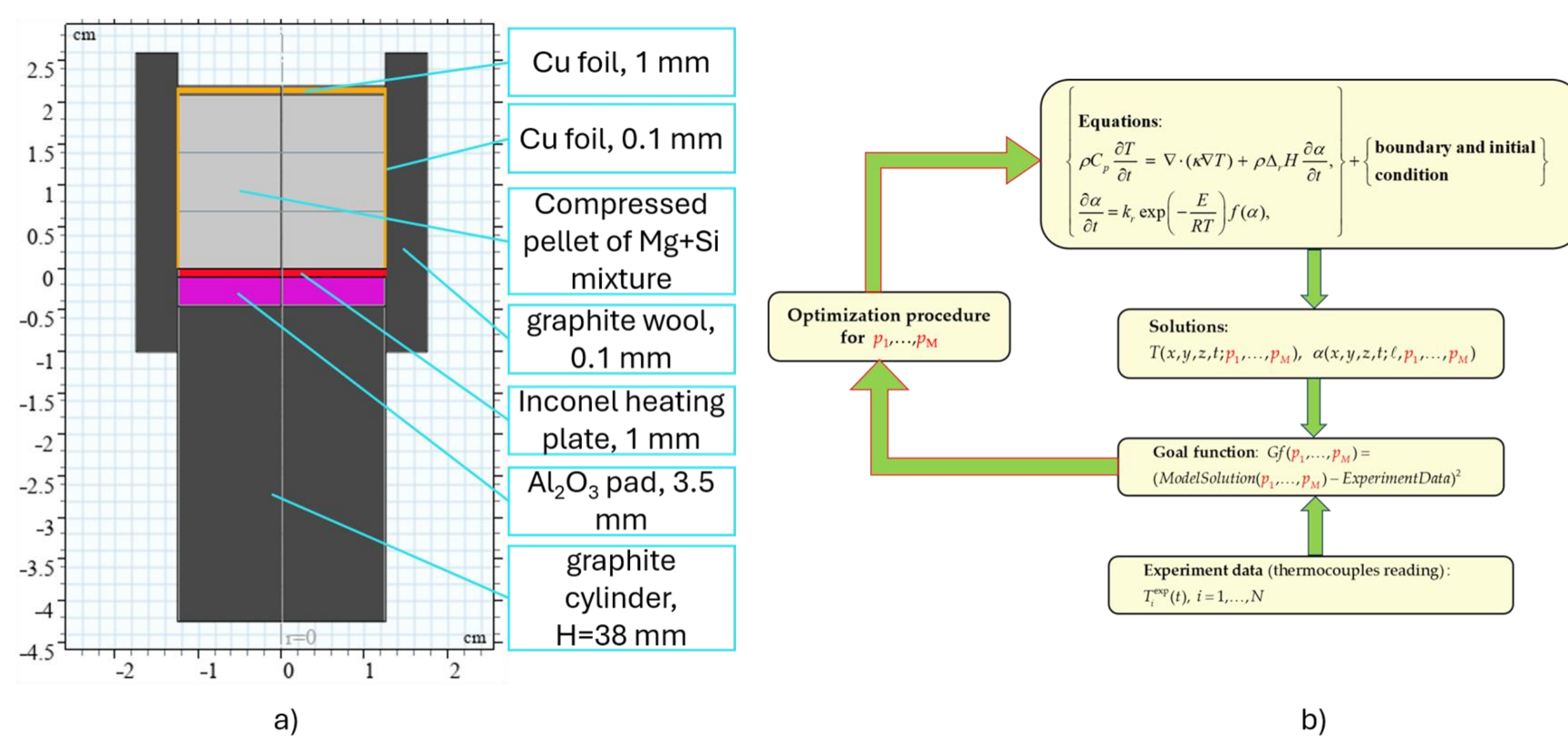


FIGURE 1. a) Detailed geometry and materials domains of the SHS model; b) Schematic of finding selected model parameters by the inverse method.

The Multiphysics SHS model

The SHS model considers a cylindrical sample located in vacuum. Energy transfer and the conversion factor are described by:

$$(1 - \varphi)\rho(\alpha, \varphi)C_p(\alpha, T)\frac{\partial T}{\partial t} = \nabla \cdot (\kappa(\alpha, \varphi, T)\nabla T) - \Delta_r H \frac{\partial \alpha}{\partial t} \rho(\alpha, \varphi)$$

$$\frac{\partial \alpha}{\partial t} = k(T)f(\alpha), k(T) = k_r \exp\left(-\frac{E_a}{RT}\right)$$

The initial sample is a mixture of Mg and Si powders which during SHS react to form Mg_2Si . Density $\rho(\alpha, \varphi)$, heat capacity $C_p(\alpha, T)$ and thermal conductivity $\kappa(\alpha, \varphi, T)$ are functions of conversion degree (α) and/or material's porosity (φ) and temperature (T). $f(\alpha)$ depends on the mechanism of SHS reaction, E_a is the activation energy and k_r is the pre-exponential kinetic coefficient. For n -th order reaction, $f(\alpha) = (1 - \alpha)^n$ and for Avrami–Erofeev kinetics: $f(\alpha) = 4(1 - \alpha)[- \ln(1 - \alpha)]^{3/4}$.

Results & conclusions

Locally ignited SHS in a custom designed reactor was successfully used to produce the Mg_2Si compound from the mixture of Mg and Si powders. Thermocouples placed in the sample measure temperature as a function of time. A 3D mathematical model of SHS in vacuum was formulated and solved in COMSOL Multiphysics®. The model takes into account the dependance of density, heat capacity and thermal conductivity on porosity, conversion degree and temperature. Measured time curves of temperature at three selected points in the reacting mixture Mg+Si were used as input data in the inverse method to obtain some physical parameters of the process (FIGURE 1). Temperature, reaction rate, and conversion degree fields were computed (FIGURE 2). Calculated temperature evolutions at selected points in the sample during SHS reaction were in good agreement with the experimental thermocouple readings.

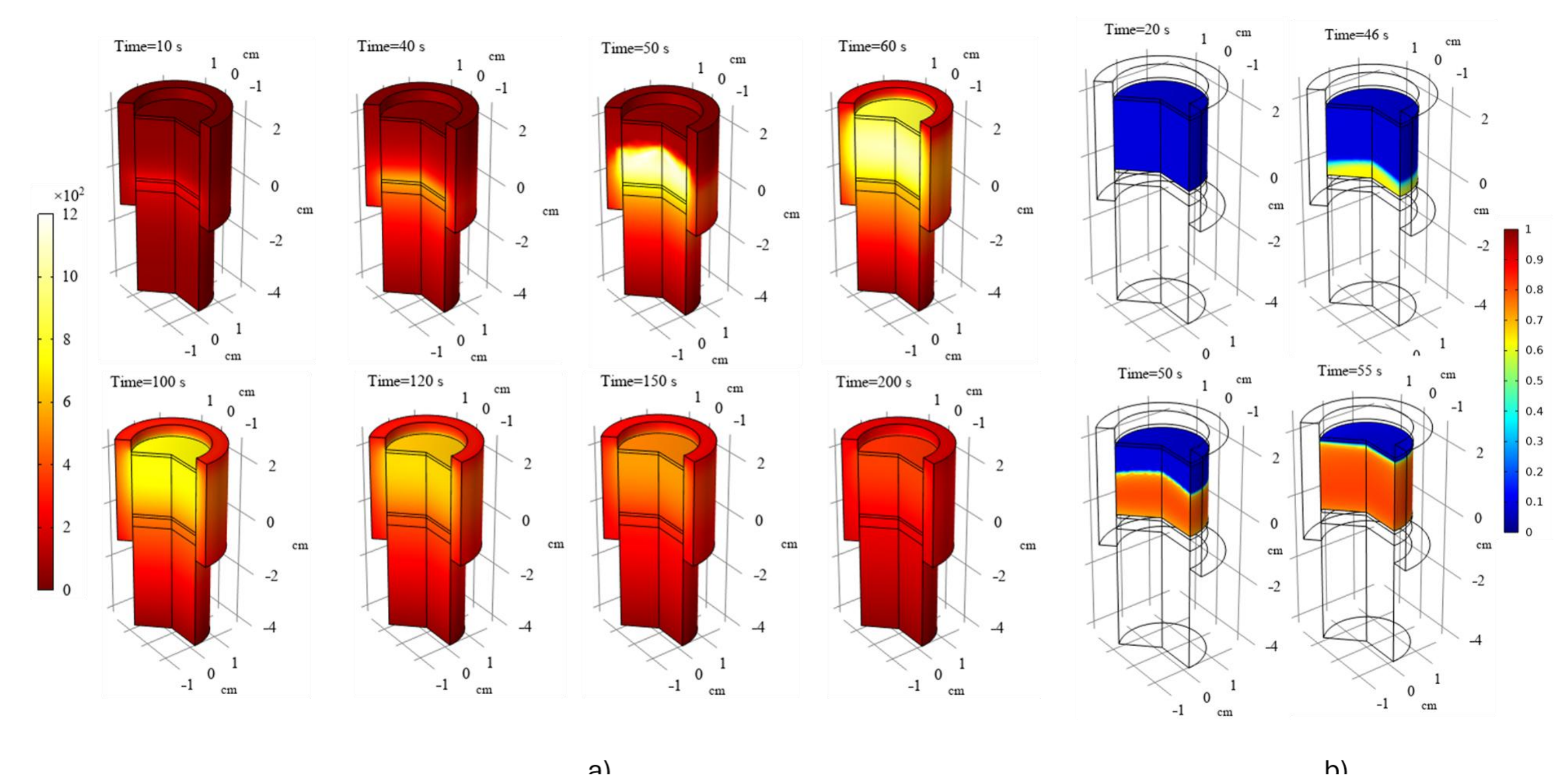


FIGURE 2. Numerical solution of the SHS model: a) temperature distribution (in K) and b) conversion factor in the sample for selected times.

REFERENCES

1. R. Filipek, "Modeling and inverse methods in materials engineering", Wydawnictwo Naukowe "Akapit", Kraków 2019.
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