A Two-Phase Flow Model for Aerogel in a Non-Equilibrium Process

†Dia Zeidan[1], Eric Goncalves[2] and Lucy T. Zhang[3]

[1]School of Basic Sciences and Humanities, German Jordanian University, Amman, Jordan
[2]ISAE-ENSMA, Institut Pprime, UPR 3346 CNRS, Poitiers, France
[3]Department of Mechanical, Aerospace and Nuclear Engineering, Rensselaer Polytechnic Institute, Troy, USA

†Corresponding and presenting author: dia.zeidan@gju.edu.jo

ABSTRACT

This paper presents the extension of hyperbolic and conservative two-phase flow model to a mixture of porous media containing nanofluids which is known as an aerogel. We focus mainly on the application of non-equilibrium mixture behaviour between phases in one space dimension. The governing equations are solved by finite volume techniques using Godunov methods of centred-type. Special emphasis is given to important and unsteady non-linear phenomena such as shock propagation in low densities aerogels. Simulation results are compared with other methods providing a remarkable agreement. Results show the good capabilities of this mixture formulation in the resolution of discontinuities in aerogel problems. This provides some insights into the fundamental properties of aerogels and helps to better understand some of the inherent difficulties in quantifying them using two-phase flow processes.

Keywords: Multiphase flows, Non-equilibrium, Nanofluids, Porous media, System of conservation laws, Riemann solution, Numerical simulation

Introduction

Aerogel is very common in industrial processes and its applications are already in use within the energy efficiency and pharmaceutical industries. In the meantime, many aerogels rely on light porous materials for which it may be densified into silica glasses by thermal processes (see, for example, [1, 2, 3, 16, 21]). During the last century, the nuclear and aerospace industries pushed strong research activity on the area. Their efforts have been aimed at the clarification of the mechanisms taking place during this complicated physical situation. In general, aerogels are solids with high void fractions, that is, porosities along with high surface areas, and possess very low densities and low conductivities. See, for example, [4, 5, 14] and references therein Typically, attention is given to the combination of high void fractions and very small pores to provide aerogels along with their extreme properties such as very low solid density and low thermal conductivity. These are mainly based on experimental analysis [6, 10, 11, 13]. However, there have been various gel-derived materials numerical simulations on the basis of simple models known as, for instance, reaction-limited cluster aggregation (RLCA) [18], diffusion-limited cluster aggregation (DLCA) [12] and diffusion-limited aggregation (DLA) [9]. Given such developments, it is of interest to investigate aerogels from mathematical and numerical point of views. Since aerogels belong to the family of nanoporous materials, it can be considered as a porous media containing nanofluids. Accordingly, one can use single or two-phase flow approach to study such phenomena. In the single-phase flow approach, the nanosolids can be simply fluidized where the relative velocity between phases is considered negligible. However, in the two-phase flow approach, this relative velocity may not be zero due to phase

interactions. In this regard, there exist a large number of articles concerning two-phase flow models where the nanofluids are treated as a simplified mixture of a base fluid and nanoparticles (see, for example, [7, 16, 20]). Most these articles are indicated that the two-phase mixture approach is more precise than single-phase approach. Further, these articles also have mainly examined the physical aspects of nanofluids rather than theoretical and numerical aspects of such models. As far it goes, only limited number of articles address the issues of the application of two-phase flow equations in nanofluids. These articles, however, used turbulent and laminar steady state two-phase flow models without taking care of the mathematical features of such models. This paper aims to study aerogels by considering a recently developed two-phase flow model (see, for example, [15, 19]). To this end, an unsteady system based on mixture formulations is presented. The aerogel is composed of nanofluids in which the dispersed phase is a gas. The governing partial differential equations (PDEs) are three mixture conservation of mass, momentum and energy. This is accomplished by three balance equations for gas void fraction, gas mass fraction and relative velocity between the gas and nanofluids. The single set of equations do not need any physical artificial stabilizing terms due to their conservativity and hyperbolicity features. Further, the deeply coupled equations are resolved on the basis of the Riemann problem using Godunov methods of centred type [17]. This solution becomes appropriate since it leads to making the relative velocity visible which is an advanced computational tool for aerogels. The model provides successful results at high and low phase velocities with low and high void fractions. It is found that the inclusion of nanofluids into the base fluid produce physically realistic solutions for strong relative motion between the nanofluids and gas phases. Results are compared with other available numerical methods producing accurate, efficient and free from numerical dissipation and dispersion computations. Simulation and test results show that the model equations can effectively simulate non-equilibrium aerogel, which may broaden the possible application areas of aerogel such as energy storage and biological tissues. The model equations and their numerical discretization for the non-equilibrium behaviour is given in the following section. Verification results are discussed in the section after followed by the conclusions section.

Governing Equations and Numerical Implementation

The aerogel considered here is composed of nanofluids in which the dispersed phase is a gas. The governing equations are formulated upon the conservation laws for mixture mass and mixture momentum along with a balance equation for the relative velocity between the two phase system in a single set of equations. For the investigation within the scope of this paper, the two-phase flow treatment is essentially a two-phase mixture model with isentropic conditions which significantly simplifies the mathematical formulation for the aerogel thereby reduces the computational costs. Within the context of mixture formulations, the total density, ρ, total and relative velocities, u and u_r, are discontinuous at the time-dependent interface with void fraction of the gas phase is always between 0 and 1. The time-dependent equations for mixture mass, mixture momentum and relative velocity are as follows:

$$\frac{\partial}{\partial t}(\rho) + \frac{\partial}{\partial x}(\rho u) = 0, \tag{1}$$

$$\frac{\partial}{\partial t}(\rho u) + \frac{\partial}{\partial x}(\rho u^2 + P + \rho c(1-c)u_r^2) = \mathcal{S}, \tag{2}$$

$$\frac{\partial}{\partial t}(u_r) + \frac{\partial}{\partial x}\left(u u_r + (1-2c)\frac{u_r^2}{2} + \psi(P)\right) = \pi, \tag{3}$$

where t is the time, x is the spatial coordinate, c is the gas mass void fraction, P represent the mixture pressure and $\psi(P)$ is a function that connect the different phases through the following relation

$$\psi(P) = e_g + \frac{P_g}{\rho_g} - e_{nf} - \frac{P_{nf}}{\rho_{nf}}. \tag{4}$$

The terms $\mathcal{S}$ and π in the mixture momentum and relative velocity equations approximate the interphase exchange processes. In addition to that, constituting relationships need to be added to system (1)-(3) so that one can predict the content of the aerogel. Closure laws account for the nanofluid physical properties and the thermodynamic behaviour of the two-phase system between gas and nanofluid phases. Under the interest that the two-phase system is considered as an aerogel, the density, velocity, relative velocity, pressure of the aerogel are calculated by the following mixture laws:

$$\begin{aligned} &\rho = \alpha_g\rho_g + (1-\alpha_g)\rho_{nf} \quad \text{and} \quad \rho u = \alpha_g\rho_g u_g + (1-\alpha_g)\rho_{nf}u_{nf}, \\ &P = \alpha_g P_g + (1-\alpha_g)P_{nf},\ c = \alpha_g\rho_g\,\rho^{-1} \quad \text{and} \quad u_r = u_g - u_{nf}, \end{aligned}$$

where subscripts (g) and (nf) denote gas and nanofluid, respectively. Further, the physical properties of the nanofluid are defined as follows:

$$\rho_{nf} = \alpha_s\rho_s + (1-\alpha_s)\rho_{bf} \quad \text{and} \quad u_{nf}\rho_{nf} = \alpha_s\rho_s u_s + (1-\alpha_s)\rho_{bf}u_{bf},$$

where indexes (s) and (bf) refer to the solid and base fluid, respectively, and the void fraction of solid and base fluid agree with $\alpha_s + \alpha_{bf} = 1$. For the system closure, the stiffened equation of state (EOS) is used for each phase as

$$P_j = K_j\left(\frac{\rho_j}{\bar{\rho}_j}\right)^{\gamma_j} - P_\infty.$$

The subscript $j = g$ is for the gas phase while $j = nf$ for the nanofluid phase and γ_j, K_j, P_∞ and $\bar{\rho}_j$ are constant parameters to be specified for each phase.
It should be noted that system (1)-(3) has been intensively studied and independent of the numerical methods being employed to resolve it. See, for example, [8, 15, 19], and the references therein. This is due to the fact that the governing equations of the system inherit conservative form as well as well-posedness forming an initial-boundary-value problem that describe different physical phenomena of interests. Furthermore, the aerogel equations (1), (2) and (3) are discretized herein by finite volume Godunov-type approach. Within such approach, the governing equations incorporate the resolution of the Riemann problem for computing the interface fluxes using the following time-marching formula [17]

$$\mathbb{U}_i^{n+1} = \mathbb{U}_i^n - \frac{\Delta t}{\Delta x}\left[\mathbb{F}_{i+\frac{1}{2}} - \mathbb{F}_{i-\frac{1}{2}}\right] + \Delta t\,\mathbb{S}_i, \tag{5}$$

which update the aerogel variables to a new time step. In (5), subscript (i) represents the cell index, the superscript (n) is the time level, Δx and Δt are the cell size and time step, respectively. $\mathbb{F}_{i\pm\frac{1}{2}}$ are the numerical fluxes through the left and right interfaces of cell (i) and $\mathbb{S}_i$ is the source terms that are evaluated at the cell centre. These fluxes are calculated by means of the solution of a Riemann problem with appropriate time-centred left and right input states. In the results shown below, Godunov methods of centred-type such as the second-order Slope Limiter

Centred (SLIC) scheme are employed in the numerical resolution of system (1)-(3). The SLIC scheme provides a high-resolution of large-gradient regions that are free from spurious oscillations. For the numerical background and details of the SLIC scheme, the readers may refer to [17].

Results and Discussion

The numerical simulations are performed by solving the governing equations (1)-(3) with two different test problems using finite volume approach within the aerogel. The first test problem, Test 1, involves the following initial data for the Riemann problem

$$\begin{aligned}
\left(\alpha_g, \rho_g, u_g\right)_L &= \left(0.5, 2.0, -0.1\right) \\
\left(\alpha_s, \rho_s, u_s, \rho_{bf}, u_{bf}\right)_L &= \left(0.7, 8.0, -2.0, 0.1, -2.0\right) \\
\text{if} \quad x &\leq 0, \\
\left(\alpha_g, \rho_g, u_g\right)_R &= \left(0.5, 2.0, 0.1\right) \\
\left(\alpha_s, \rho_s, u_s, \rho_{bf}, u_{bf}\right)_R &= \left(0.7, 8.0, 2.0, 0.1, 2.0\right) \\
\text{if} \quad x &> 0,
\end{aligned}$$

corresponding to rarefaction waves traveling in opposite direction separated by a contact discontinuity. In the second test problem, Test 2, we consider a collision of two symmetric shock waves, weak, and a trivial contact discontinuity with the following initial data

$$\begin{aligned}
\left(\alpha_g, \rho_g, u_g\right)_L &= \left(0.01, 3.0, 0.1\right) \\
\left(\alpha_s, \rho_s, u_s, \rho_{bf}, u_{bf}\right)_L &= \left(0.0001, 7.0, 0.4, 2.0, 0.4\right) \\
\text{if} \quad x &\leq 0, \\
\left(\alpha_g, \rho_g, u_g\right)_R &= \left(0.01, 3.0, -0.1\right) \\
\left(\alpha_s, \rho_s, u_s, \rho_{bf}, u_{bf}\right)_R &= \left(0.0001, 7.0, -0.4, 2.0, -0.4\right) \\
\text{if} \quad x &> 0.
\end{aligned}$$

The simulations results are shown in figures 1 and 2. In all results, it is not possible to solve the governing equations analytically due to the existence of several non-linear properties and closure relations involved for the current interest in aerogel. Thus, we produce a high-resolution numerical solution for the Riemann problem to calculate the reference solution on a very fine mesh of 10000 cells by using the total variation diminishing (TVD) slope limiter centre (SLIC) scheme in the computational domain $[-10, 10]$. Simulations are also carried out for the aerogel with transmissive boundary conditions along with a CFL number of 0.9 through the SUPERBEE limiter in the course of the SLIC scheme. The numerical resolutions (symbols) are compared with the reference solutions (solid lines) as well as with numerical solutions provided by other numerical methods available in the literature on a coarse mesh of 100 cells. In the test cases, $\gamma_g = 1.4$, $\bar{\rho}_g = 1.0$ kg/m^3, $K_g = 1.0$ Pa and $P_\infty = 0$, $\gamma_{nf} = 2.8$, $\bar{\rho}_{nf} = 1.0$ kg/m^3, $K_{nf} = 1.0$ Pa and $P_\infty = 1$ Pa. Results for Test 1 are shown in figure 1 for the mixture two-phase flow variables using three different numerical methods at a time $t = 0.8$ ms. The solution for this test problem consists of left and right rarefactions propagating in opposite direction. Figure 1 shows that the resolution for the relative velocity jump across the middle wave with lower densities whereas the mixture density, mixture velocity and mixture pressure are not for rarefaction

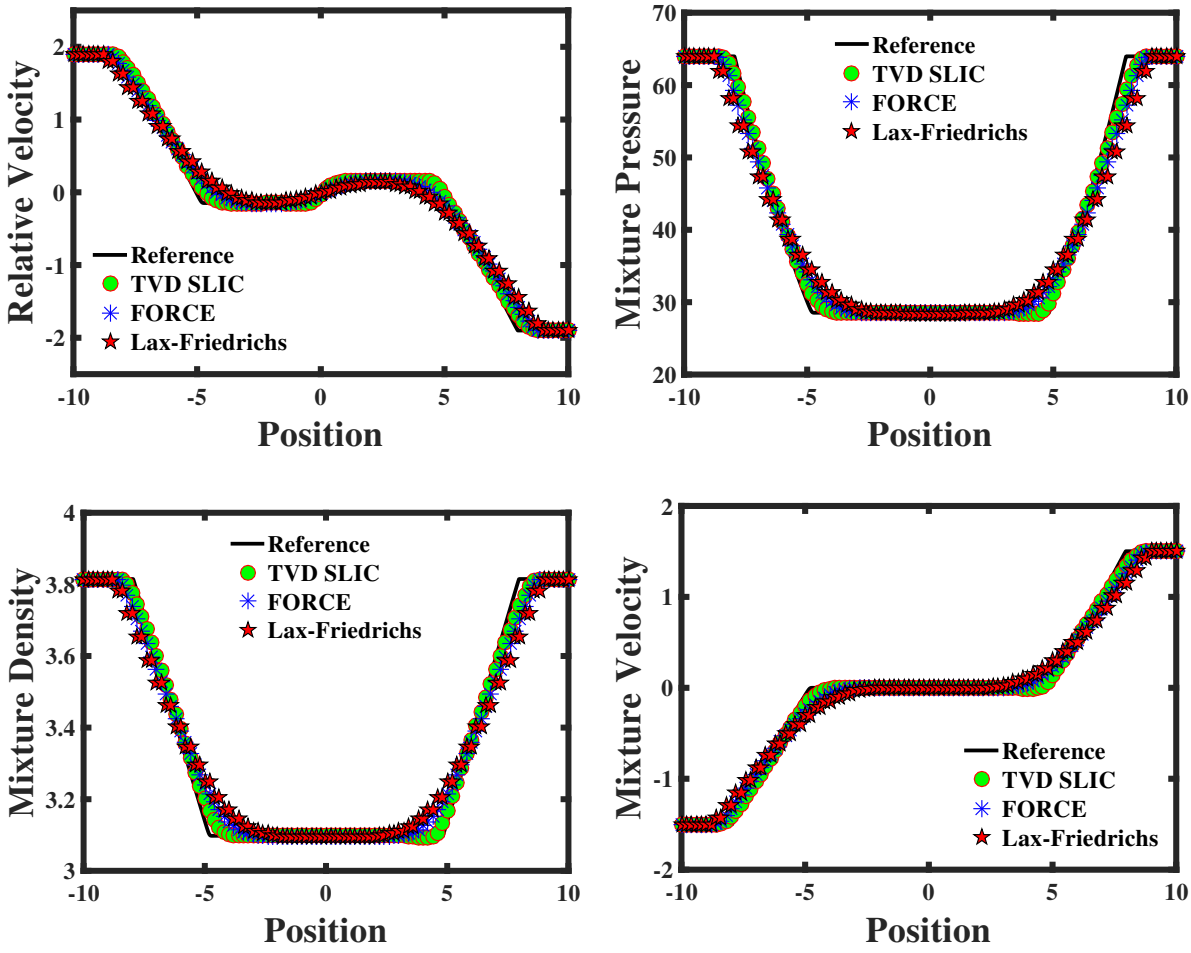

Figure 1: Test 1: Expansion tube problem in an aerogel at time $t = 0.8$ ms. The TVD SLIC, first-order centered (FORCE) and Lax-Friedrichs methods are compared with the reference solution results. Coarse meshes, symbols, are provided on 100 cells and very fine meshes of 10000 cells for the solid lines. The waves seen from left to right, a fast left rarefaction, a contact discontinuity and a fast right rarefaction.

waves propagating in an aerogel. The relative velocity jump indicates the possible sudden jump of gas and nanofluid velocities across the middle wave. It is also noted that a good agreement with the reference solutions for all the three methods and the model is able to deal with low density test case producing rarefaction waves. In figure 2, the results from a collision with an aerogel of low phase densities are displayed at a time $t = 2.3$ ms. The solution for the aerogel contains a left shock wave, a contact discontinuity and a right shock wave. It is observed that the mixture flow variables remain constant across the middle wave, however, the relative velocity jump discontinuously as in Test 1. This leads to the fact that lower phase densities slowly increase the relative motion between the gas and nanofluid phases during the collision. Again, the numerical results compared favourably with the reference solutions and capable of producing oscillation-free profiles at discontinuities. We conclude that the present model eqautions and the associated methods can automatically treat aerogel as a two-phase flow system even with different physical situations.

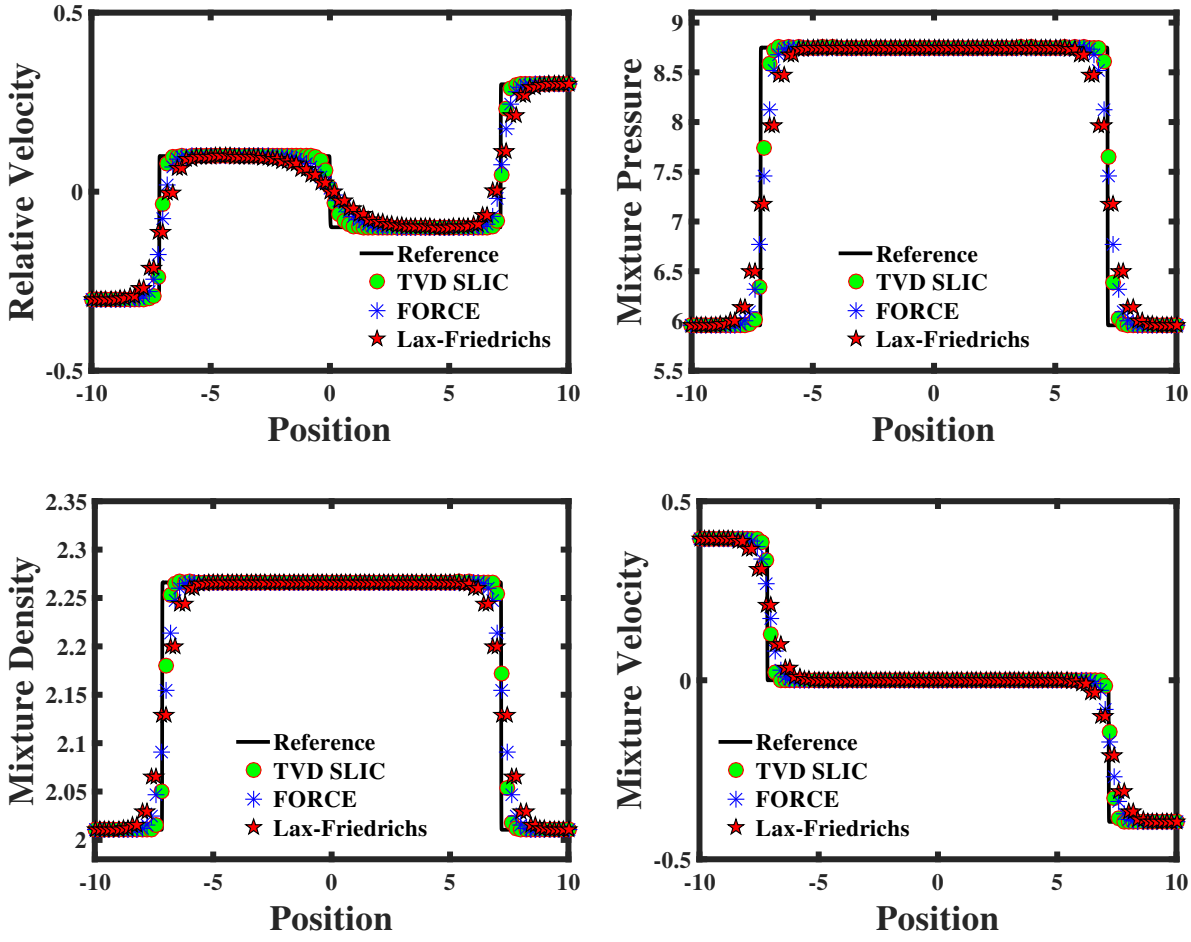

Figure 2: Test 2: The numerical results of the shock tube problem in an aerogel at time $t = 2.3$ ms. Comparison of the three different numerical methods using a coarse mesh of 100 cells (symbols) and the reference solution on a very fine mesh of 10000 cells (solid lines).

Concluding Remarks

A mixture two-phase flow model has been proposed for the simulation of porous media containing nanofluids used widely in aerogel production. This is motivated by the relative motion demanded to cope with the two-phase system arising in aerogels. Godunov methods with gradually growing levels of complexity in aerogels are employed to solve the governing equations in a one-dimensional domain representing the conceptual low-densities two-phase flow problems. This indicates that the extension of such mixture model to aerogel seems worthwhile as these type of hyperbolic problems are much better documented.

Acknowledgment

This work is supported by the German Jordanian University through the project SEED-SNRE 7-2014.

References

[1] Aegerter, M.A., Prassas, M. (2011) *Advances in Sol-Gel Derived Materials and Technologies*, Springer.

[2] Akimov, Y.K. (2003) Fields of application of aerogels (Review). *Instruments and Experimental Techniques* **46**, 287–299.

[3] Chaput, F., Lecomte, A., Dauger, A., Boilot, J.P. (1989) Preparation and structure of aluminosilicate aerogels. *Chem. Mater.* **1**, 199–201.

[4] Gurikov, P.A., Kolnoochenko, A.V., Menshutina, N.V. (2009) 3D reversible cellular automata for simulation of the drug release from aerogel-drug formulations. *Computer Aided Chemical Engineering* **26**, 943–947.

[5] Fricke, J. (1986) Thermal transport in porous superinsulations in aerogels. *Springer Proceedings in Physics* **6**, 94–103.

[6] Ibrahim, M., Biwole, P.H., Achard, P., Wurtz, E. (2015) Aerogel-based materials for improving the building envelopes thermal behavior: a brief review with a focus on a new aerogel-based rendering. *Energy Sustainability Through Green Energy*, 163–188.

[7] Kalteh, M., Abbassi, A., Saffar-Avval, M., Harting, J. (2011) Eulerian-Eulerian two-phase numerical simulation of nanofluid laminar forced convection in a microchannel. *International Journal of Heat and Fluid Flow* **32**, 107–116.

[8] La Spina, G., de'Michieli Vitturi, M., Clarke, A.B. (2017) Transient numerical model of magma ascent dynamics: application to the explosive eruptions at the Soufrière Hills Volcano. *Journal of Volcanology and Geothermal Research* **336**, 118–139.

[9] Ma, H.-S., Prévost, J.-H., Jullien, R., Scherer, G.W. (2001) Computer simulation of mechanical structureproperty relationship of aerogels. *J.Non-Cryst.Solids* **285**, 216–221.

[10] McNaught, A.D., Wilkinson, A. (2007) *Compendium of Chemical Terminology*, IUPAC Goldbook, PAC, 791801, Blackwell Science.

[11] Morales, R.V., da Cunha, C.R., Rambo, C.R. (2014) A complex network approach for the growth of aerogels. *Physica A: Statistical Mechanics and its Applications* **406**, 131–138.

[12] Paul, M. (1983) Formation of fractal clusters and networks by irreversible diffusion-limited aggregation. *Phys. Rev. Lett.* **51**, 1119–1122.

[13] Platzer, W.J., Bergkvist, M. (1993) Bulk and surface light scattering from transparent silica aerogel. *Solar Energy Mater Solar Cells* **31**, 243–251.

[14] Richter, K., Norris, P.M., Chang, C.L. (1995) Aerogels: applications, structure and heat transfer phenomena. *In Review on Heat Transfer, V. Prasad, Y. Jaluria, and G. Chen, Eds.* **6**, 61–114.

[15] Romenski, E., Resnyansky, A.D., Toro, E.F. (2007) Conservative hyperbolic formulation for compressible two-phase flow with different phase pressures and temperatures. *Quart. Appl. Math.* **65**, 259–279.

[16] Sehaqui, H., Zhou, Q., Berglund, L.A. (2011) High-porosity aerogels of high specific surface area prepared from nanofibrillated cellulose (NFC). *Composites Science and Technology* **71**, 1593–1599.

[17] Toro, E.F. (2009) *Riemann Solvers and Numerical Methods for Fluid Dynamics: A Practical Introduction*, Springer.

[18] Woignier, T., Phalippou, J., Pelous, J., Courtens, E. (1990) Different kinds of fractal structures in Si02 aerogels. *J. Non-Cryst. Solids* **121**, 198–204.

[19] Zeidan, D. (2016) Assessment of mixture two-phase flow equations for volcanic flows using Godunov-type methods. *Applied Mathematics and Computation* **272**, 707–719.

[20] Zhao, L., Yang, S., Bhatia, B., Strobach, E., Wang, E.N. (2016) Modeling silica aerogel optical performance by determining its radiative properties. *AIP Advances* **6**, 025123-1–8.

[21] Zhou, X., Torabi, M., Lu J., Shen, R., Zhang, K. (2014) Nanostructured energetic composites: synthesis, ignition/combustion modeling, and applications. *ACS Appl Mater Interfaces* **6**, 3058–74.