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Unconstrained ETD Methods on the Diffuse-Interface Model with the Peng-Robinson Equation of State

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Abstract. In this study, we apply first-order exponential time differencing (ETD) methods to solve benchmark problems for the diffuse-interface model using the Peng-Robinson equation of state. We demonstrate the unconditional stability of the proposed algorithm within the ETD framework. Additionally, we analyzed the complexity of the algorithm, revealing that computations like matrix multiplications and inversions in each time step exhibit complexity strictly less than $\mathcal{O}(n^2)$, where n represents the number of variables or grid points. The main objective was to develop an algorithm with enhanced performance and robustness. To achieve this, we avoid iterative solutions (such as matrix inversion) in each time step, as they are sensitive to matrix properties. Instead, we adopted a hierarchical matrix ($\mathcal{H}$-matrix) approximation for the matrix inverse and matrix exponential used in each time step. By leveraging hierarchical matrices with a rank $k \ll n$, we achieve a complexity of $\mathcal{O}(kn\log(n))$ for their product with an n-vector, which outperforms the traditional $\mathcal{O}(n^2)$ complexity. Overall, our focus is on creating an unconditionally stable algorithm with improved computational efficiency and reliability.

AMS subject classifications: 65F60, 65M22, 68Q25
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1 Introduction

The Peng-Robinson equation of state (PR-EOS) has proven to be highly successful in accurately describing the thermodynamic properties of fluids and gases in various fields,

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including academia, industry, petroleum, and chemical engineering [1,2]. Compared to the Van der Waals equation, the cubic equation of state provides more precise predictions for a wide range of materials, such as nitrogen (N_2), carbon dioxide (CO_2), and hydrocarbons. Crucially, phase equilibrium calculations ([3–7]) and surface tension predictions [4,5,8–10] between gas and liquid play a vital role in simulating essential problems related to phase equilibrium. Recently, exponential time differencing (ETD) methods have garnered attention for solving numerical solutions in partial differential equations (PDEs) containing nonlinear terms, including Allen-Cahn and Cahn-Hilliard equations [11–13], as well as other dynamic interfacial problems. Although the ETD methods were once considered impractical [14], they demonstrated advantages, particularly when dealing with high stiffness linear operators present in the aforementioned PDEs. By overcoming the numerical instability issues arising from system stiffness, ETD methods have become a favorable choice for such problems.

This work is focused on applying first-order ETD methods to solve the model problem for the diffuse-interface model with the Peng-Robinson equation of state. The framework of the ETD is briefly discussed, and the unconditional stability of the algorithm is proven. The primary goal is to develop an algorithm that is more robust, unconditionally stable, and reduces the time required to construct the matrix exponentials. Additionally, the aim is to avoid iterative solutions, such as matrix inversion, in each time step during the online calculations. The performance of the algorithm depends heavily on matrix sparsity, and reducing the complexities of matrix-vector multiplications is a key objective.

To achieve these goals, the matrix inverse and matrix exponential used in each time step were constructed using hierarchical matrices ($\mathcal{H}$-matrix, [15,16]). Originally developed for linear algebra arithmetic involving dense matrices, a hierarchical matrix of size $n\times n$ consists of a tree of blocks with a certain rank $k\ll n$. The complexities for its product with an n-vector are shown to be $\mathcal{O}(kn\log(n))$, which is significantly better than the $\mathcal{O}(n^2)$ complexity observed with regular dense matrices. By employing hierarchical matrices, the algorithm ensures efficient computations, thereby contributing to the desired stability and performance objectives.

2 Model equations

For convenience, we adapt all the notations in [17] to describe the model equation as follows: The model equation was used to calculate the Helmholtz free energy density (denoted as f_b) of a bulk fluid as a function of molar density (denoted as c). This equation is derived from the PR-EoS (Peng-Robinson Equation of State) and is given by

$$f_b(c)=f_b^{ideal}(c)+f_b^{repulsion}(c)+f_b^{attraction}(c), \tag{2.1}$$

where the components of the Helmholtz free energy density are defined as follows:

1. Ideal Gas Contribution (f_b^{ideal}):

$$f_b^{ideal}(c)=c\vartheta_0+cRT\ln(c), \tag{2.2}$$