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On the relation between dynamical density functional theory and Navier-Stokes equation

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HIGHLIGHTS

- The intrinsic relation between Navier-Stokes equations and DDFTs is revealed.
- Various DDFTs are derived from general Navier-Stokes equations with near-equilibrium assumptions.
- DDFT is extended to address coupling effect between fluid flow and adsorption dynamics.
- Pressure tensor is identified as the kernel for describing fluid dynamics.

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ABSTRACT

Navier-Stokes equations are widely applied to deal with non-equilibrium fluid dynamics such as the flow field on nanoscale. On the other category, dynamical density functional theory (DDFT) has recently been recognized as a robust tool to investigate the non-equilibrium processes such as molecular diffusion and adsorption dynamics. Both approaches have achieved great success and it's natural to wonder if there is any intrinsic relation inbetween. Herein, we prove that DDFT can be derived from the general Navier-Stokes equations with approximate evaluation of pressure tensor. Motivated by this procedure, we introduce the flow effect on pressure tensor, and then propose extensions of DDFT for addressing the coupling between dynamic adsorption and fluid flow. This work, revealing the relation between DDFT and Navier-Stokes equations, not only casts novel insights into the extension of DDFT, but also highlights a potential route to overcome the Navier-Stokes analytic solution problem.

1. Introduction

Most chemical engineering processes are in non-equilibrium involving molecular diffusion and mass transport (Wang et al., 2014; Krishna, 2012). To describe the dynamic characteristics of these processes, quite a few theoretical approaches have been developed. Among them, the Fick mass transport equation provides a traditional method for describing mass transport (Liu et al., 2012; Andreucci et al., 2019), and the basic idea lying in it is that the concentration gradient gives the driving force and drives the evolution of local concentration. In practical applications, the diffusion coefficients need to be determined in prior under most conditions, while the diffusion coefficients, often measured experimentally, do not reflect their dependence on local position (Kozlova et al., 2019; Chacón-Acosta et al., 2020). Such a dependence becomes key important when dealing molecular diffusion in nanopores (Liu et al., 2013a; Stopper et al., 2015). Another well-known approach is the Navier-Stokes equations, which establish the relations among the acceleration of fluid particle momentum, the exchange of liquid inside pressure, the dissipative viscous force and the gravity force. Navier-Stokes equations have been widely applied in engineering field to investigate non-equilibrium fluid dynamics (Łukaszewicz and Kalita, 2016). Due to the broad applications of Navier-Stokes equations, many physicists and even mathematicians have further investigated the compressible and incompressible Navier-Stokes equations and obtained a number of achievements (Bassi and Rebay, 1997; Xin, 1998; Kennedy et al., 2000; Brooks and Hughes, 1982; Ghia et al.,

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1982; Bell et al., 1989). From a mathematical perspective, the Navier-Stokes equations represent a set of partial differential equations, and its analytical solution can be hardly figured out due to its high non-linearity (Wolfram, 2002). Towards this end, various types of computational methods are proposed and developed. In addition, the simplifications of Navier-Stokes equations in specific circumstances are also reported for the convenience of calculation (Łukaszewicz and Kalita, 2016).

On the other category, the molecular diffusion equations, starting from molecular interaction, do not depend on the empirical mass transport parameters. In this regard, several theories have been developed, including non-equilibrium molecular dynamics (Tao et al., 2018), and kinetic Monte Carlo Simulation (Acharya et al., 2017; Chang et al., 2018). However, if one wants to obtain the macroscopic thermodynamic properties such as free energy and entropy, the detailed microscopic information of every molecule is needed in order to perform the ensemble average of observables at each time moment. Overall, the computation cost of this procedure is expensive. Besides, the time scale and length scale for the systems handled by the above theories are far too small compared with the real chemical systems. Hence, more advanced methods that can handle larger non-equilibrium systems and describe the local diffusion under different external fields are necessitated.

As one of the advanced liquid state theories, density functional theory (DFT) has recently been recognized as a robust tool to study the thermodynamic properties by predicting local density distributions of inhomogeneous liquids at equilibrium (Wu and Li, 2007; Hansen and McDonald, 2013), and it has been successfully applied to investigate solvation (Liu et al., 2011; Liu et al., 2013b; Zhao et al., 2011), adsorption (Schumacher et al., 2000; Liu et al., 2009; Ravikovitch et al., 2000), wetting (Meister and Kroll, 1985; Zeng et al., 2011), and freezing (Singh, 1991). Moreover, DFT has been extended to non-equilibrium systems, which is known as the dynamical density functional theory (DDFT). Till date, there have been a few formulation works on DDFT, providing important foundation for the engineering applications in the fields of green energy and novel materials (Qing et al., 2020; Qing et al., 2019; Liu, 2016). The first derivation of DDFT has been reported in the late 1990s when Marconi (Marconi and Tarazona, 1999) et al. presented a time-dependent density functional approach to study the relaxational dynamics interacting fluids subject to thermal noise. Afterwards, several modifications have been proposed by Marconi and his co-workers (Marconi and Tarazona, 2000; Marini Bettolo Marconi and Tarazona, 2006). Marconi et al. started from the Langevin stochastic equations and formulated a DDFT. In 2004, Archer and Evans (Archer and Evans, 2004) gave another derivation of DDFT, and they started from the Smoluchowski equation and employed near-equilibrium assumptions. Such a derivation elucidates the physical assumptions inherent in Marconi's work (Marconi and Tarazona, 1999). More recently, Español and Löwen (2009) derived a general version of DDFT for complex fluids which involves a generalized diffusion tensor. This approach assumes that the density evolves in time much slower than the current correlation function and it is in particular appropriate if there exist simultaneously fast and slow evolutions of observables.

Both DDFT and Navier-Stokes equations are versatile methods for dynamical processes in chemical engineering (Padmanabhan, 2011; Hansen et al., 2013), but the relation between them is not clear yet. In this work, we show that DDFT can be derived from the general Navier-Stokes equations. Our derivation, starting from the momentum balance equation of Navier-Stokes equations, involves a generalized pressure tensor as a kernel. By approximating the pressure tensor in different ways, we show that various versions of DDFT can be obtained.

The remainder of this work is laid out as follows: the classical DFT is recapitulated in the next section together with DDFT. Afterwards, a brief introduction of the Navier-Stokes equations is given in Section 3. The new derivation of DDFT from the Navier-Stokes equations is presented in Section 4. The extension of DDFT for addressing the coupling between dynamic adsorption and fluid flow is proposed in Section 5. Finally, a brief conclusion is given in Section 6.

2. Classical density functional theory

For interacting fluid subject to an external potential, there exists only one unique local density distribution, $\rho(\mathbf{r})$, and then the thermodynamic properties of this system can be determined by $\rho(\mathbf{r})$. This is the key idea in the Hohenberg-Kohn theorem (Hohenberg and Kohn, 1964; Evans, 1979). The major challenge in the practical application of DFT lies in the construction of the intrinsic Helmholtz free energy functional in term of local density, i.e., $F_{int}[\rho(\mathbf{r})]$. Within the grand canonical ensemble, the grand potential and the intrinsic free energy are related through:

$$\Omega[\rho(\mathbf{r})] = \mathrm{F}_{\mathrm{int}}[\rho(\mathbf{r})] + \int d\mathbf{r}[\rho(\mathbf{r})(\mathrm{V}_{\mathrm{ext}}(\mathbf{r}) - \mu)], \quad (1)$$

where $V_{\mathrm{ext}}(\mathbf{r})$ is the external potential and μ is the chemical potential of system component. In general, the intrinsic Helmholtz free energy includes an ideal-gas contribution and an excess one due to intermolecular interaction:

$$F_{int}[\rho(\mathbf{r})] = F^{\mathrm{id}}[\rho(\mathbf{r})] + F^{\mathrm{ex}}[\rho(\mathbf{r})]. \quad (2)$$

The ideal-gas contribution for inhomogeneous fluids can be formulated exactly as:

$$\beta F^{\mathrm{id}}[\rho(\mathbf{r})] = \int d\mathbf{r}\rho(\mathbf{r})\left[\ln(\rho(\mathbf{r})\Lambda^3) - 1\right], \quad (3)$$

where Λ is the thermal wavelength, $\beta = 1/k_BT$ with k_B the Boltzmann constant, and T is the absolute temperature.

For the system free of Coulombic interaction, the excess contribution can be generally divided into two terms, i.e., the one accounting for short-range repulsion and the one for perturbative attraction:

$$F^{\mathrm{ex}}[\rho(\mathbf{r})] = F^{\mathrm{ex}}_{\mathrm{rep}}[\rho(\mathbf{r})] + F^{\mathrm{ex}}_{\mathrm{att}}[\rho(\mathbf{r})]. \quad (4)$$

Both the repulsion term and attraction term can't be formulated exactly, and to tackle this problem, various approximate functionals have been developed. A feasible approach to address the repulsion contribution is the fundamental measure theory (FMT) originally developed by Rosenfeld (1989) and extended by several groups (Yu and Wu, 2002a,b). As for the attraction contribution, the first-order mean spherical approximation (FMSA) by Tang (2004) gives a convenient yet accurate solution, especially for the Lennard-Jones (LJ) systems. When the long-range Coulombic interaction is involved, additional contribution should be properly introduced (Zhao et al., 2011). The intrinsic Helmholtz free energy is related with the Helmholtz free energy through:

$$F[\rho(\mathbf{r})] = F_{int}[\rho(\mathbf{r})] + \int d\mathbf{r}\rho(\mathbf{r})V_{ext}(\mathbf{r}). \quad (5)$$

At thermodynamic equilibrium, the density profile, $\rho(\mathbf{r})$, can be determined by minimizing the Helmholtz free energy or grand potential functional:

$$\frac{\delta\Omega[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})} = \frac{\delta F[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})} - \mu = 0. \quad (6)$$

The minimization gives the following Euler-Lagrange equation (Zhao et al., 2015):

$$\rho(\mathbf{r}) = \rho_{\mathrm{b}}\exp\left(\beta\mu_{\mathrm{b}}^{\mathrm{ex}} - \beta\frac{\delta F^{\mathrm{ex}}[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})} - \beta V_{\mathrm{ext}}(\mathbf{r})\right), \tag{7}$$

where ρ_{b} and $\mu_{\mathrm{b}}^{\mathrm{ex}}$ are the number density and excess chemical potential of the corresponding bulk fluid, and $\delta F^{\mathrm{ex}}[\rho(\mathbf{r})]/\delta\rho(\mathbf{r}) = \mu^{\mathrm{ex}}(\mathbf{r})$ gives the local excess chemical potential.

The static density functional theory has been extended to non-equilibrium systems (Archer and Evans, 2004; Español and Löwen, 2009). The most prominent difference between DFT and DDFT is that DDFT redefines the density profile with a time limitation term t, which means the time evolution of local density profile, $\rho(\mathbf{r}, t)$, is associated with the time-dependent free energy functional. Currently, there are mainly three versions of DDFT for describing the temporal and spatial evolution of local density.

Starting from the Brownian dynamics equation, Marconi et al. used Langevin stochastic equations to derive a DDFT equation as follows (Marconi and Tarazona, 1999; Marconi and Tarazona, 2000) :

$$\frac{\partial\rho(\mathbf{r};t)}{\partial t} = \frac{1}{\gamma m}\nabla_{\mathbf{r}}\cdot\left[\rho(\mathbf{r};t)\nabla_{\mathbf{r}}\frac{\delta F[\rho(\mathbf{r};t)]}{\delta\rho(\mathbf{r};t)}\right], \tag{8}$$

where γ is the friction coefficient. In above equation, Marconi et al. assumed that the two-body correlations decay much faster to equilibrium than the one-body density, in other words, the above equation is therefore applicable for over-damped systems. This equation has been applied later to investigate solvation dynamics (Yoshimori, 2004) and crystal growth (Van Teeffelen et al., 2009). Archer and Evans used the Smoluchowski equation to derive the dynamic equation as follows (Archer and Evans, 2004; Archer, 2009) :

$$\frac{\partial^2}{\partial t^2}\rho(\mathbf{r};t) + \gamma\frac{\partial}{\partial t}\rho(\mathbf{r};t) = \frac{1}{m}\nabla_{\mathbf{r}}\cdot[\rho(\mathbf{r};t)\nabla_{\mathbf{r}}\frac{\delta F[\rho(\mathbf{r};t)]}{\delta\rho(\mathbf{r};t)}]. \tag{9}$$

Compared to Marconi's version, the second derivation of local density with respect to time is present. During this derivation it's easy to involve the many-body interactions among particles into consideration. The above equation has been employed to study the dynamics of fluid phase separation (Archer, 2005). It should be noted that the same DDFT equation was also derived from the Newton's equation of motion (Archer, 2006).

Recently, Español and Löwen (2009) derived the density evolution equation from the Liouville equation with the help of projection operator technique:

$$\frac{\partial}{\partial t}\rho(\mathbf{r};t) = \nabla_{\mathbf{r}}\int d\mathbf{r}'\mathbf{D}(\mathbf{r},\mathbf{r}';t)\nabla_{\mathbf{r}'}\frac{\delta F[\rho(\mathbf{r};t)]}{\delta\rho(\mathbf{r};t)}, \tag{10}$$

where $\mathbf{D}(\mathbf{r},\mathbf{r}';t)$ represents the diffusion tensor given by Green-Kubo relation. They showed that by using different approximations to treat the diffusion tensor, the two previous DDFT equations can be recovered. Therefore, the above equation can be seen as a general version of DDFT equation.

3. Navier-Stokes equations

Starting from Newton's constitutive equation, Navier and Stokes gave the set of Navier-Stokes equations, reading:

$$\frac{\partial}{\partial t}(\rho\mathbf{v}) + \nabla_{\mathbf{r}}\cdot(\rho\mathbf{v}\otimes\mathbf{v}) = Q + \frac{\Gamma}{m}, \tag{11}$$

where $\otimes$ represents the dyadic product of two vectors, $\mathbf{v}$ is the velocity of the fluid, $\mathbf{v}\otimes\mathbf{v}$is a special case of the tensor product, Q represents the sources and sinks in the fluid, and Γ is the total force. For classical fluid in which no chemical reaction is involved, the sources and sinks term vanishes, i.e., $Q = 0$. Expanding the derivatives completely we have:

$$\mathbf{v}\frac{\partial\rho}{\partial t} + \rho\frac{\partial\mathbf{v}}{\partial t} + \mathbf{v}\otimes\mathbf{v}\cdot\nabla_{\mathbf{r}}\rho + \rho\mathbf{v}\cdot\nabla_{\mathbf{r}}\mathbf{v} + \rho\mathbf{v}\nabla_{\mathbf{r}}\cdot\mathbf{v} = \frac{\Gamma}{m}. \tag{12}$$

Since $\mathbf{v}\otimes\mathbf{v}\cdot\nabla_{\mathbf{r}}\rho = \mathbf{v}(\mathbf{v}\cdot\nabla_{\mathbf{r}}\rho)$, Eq. (12) can be rewritten as:

$$\mathbf{v}\left(\frac{\partial\rho}{\partial t} + \nabla_{\mathbf{r}}\cdot(\rho\mathbf{v})\right) + \rho\left(\frac{\partial\mathbf{v}}{\partial t} + \mathbf{v}\cdot\nabla_{\mathbf{r}}\mathbf{v}\right) = \frac{\Gamma}{m}. \tag{13}$$

The leftmost term enclosed in parentheses is, by mass continuity, equal to zero. Noting that what remains on the left side of the equation is the convective derivative:

$$\rho\frac{D\mathbf{v}}{Dt} = \frac{\Gamma}{m}, \tag{14}$$

where $\frac{D}{Dt} \equiv \frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla_{\mathbf{r}}$ is the material derivative, which is a derivative taken along a path moving with velocity, $\mathbf{v}$. Then we multiply both sides of Eq. (14) by m, giving:

$$\rho\frac{D\mathbf{p}}{Dt} = \Gamma, \tag{15}$$

where $\mathbf{p} = m\mathbf{v}$ is the local momentum. The above equation represents a generalized expression of Newton's second law in terms of body forces instead of point forces (Łukaszewicz and Kalita, 2016).

4. From Navier-Stokes equations to DDFT

Now we specialize the above Navier-Stokes equations into the material derivative. We add the position and time variable to Eq. (15), yielding:

$$\rho(\mathbf{r};t)\frac{D\mathbf{p}(\mathbf{r};t)}{Dt} = \Gamma(\mathbf{r}), \tag{16}$$

where $D\mathbf{p}(\mathbf{r};t)/Dt$ is contributed by two terms: unsteady acceleration and convective acceleration (Łukaszewicz and Kalita, 2016), i.e.:

$$\frac{D\mathbf{p}(\mathbf{r};t)}{Dt} = \frac{\partial\mathbf{p}(\mathbf{r};t)}{\partial t} + \frac{1}{m}\mathbf{p}(\mathbf{r};t)\cdot\nabla_{\mathbf{r}}\mathbf{p}(\mathbf{r};t). \tag{17}$$

For colloidal systems, the body force, $\Gamma(\mathbf{r})$, contains three contributions: viscosity, pressure gradient and external force (Łukaszewicz and Kalita, 2016):

$$\Gamma(\mathbf{r}) = -\gamma\rho(\mathbf{r};t)\mathbf{p}(\mathbf{r};t) - \nabla_{\mathbf{r}}\cdot\hat{\mathbf{P}} + \rho(\mathbf{r};t)\mathbf{f}^{\mathrm{ex}}(\mathbf{r}), \tag{18}$$

where $\mathbf{f}^{\mathrm{ex}}(\mathbf{r}) \equiv -\nabla_{\mathbf{r}}V_{\mathrm{ext}}(\mathbf{r})$ is the one-body external force. Substituting Eqs. (17) and (18) back into eq.(16), giving:

$$\rho(\mathbf{r};t)\frac{\partial\mathbf{p}(\mathbf{r};t)}{\partial t} + \frac{1}{m}\rho(\mathbf{r};t)\mathbf{p}(\mathbf{r};t)\cdot\nabla_{\mathbf{r}}\mathbf{p}(\mathbf{r};t)$$
$$= -\gamma\rho(\mathbf{r};t)\mathbf{p}(\mathbf{r};t) - \nabla_{\mathbf{r}}\cdot\hat{\mathbf{P}} + \rho(\mathbf{r};t)\mathbf{f}^{\mathrm{ex}}(\mathbf{r}). \tag{19}$$

Eq. (19) is known as the equation of local momentum balance. Note that $\nabla_{\mathbf{r}}\cdot\hat{\mathbf{P}}$ represents the derivative of local pressure tensor. The pressure tensor, $\hat{\mathbf{P}}$, can be decomposed into two parts: the kinetic part $\hat{\mathbf{P}}^{(K)}$and the potential part $\hat{\mathbf{P}}^{(V)}$:

$$\hat{\mathbf{P}} = \hat{\mathbf{P}}^{(K)} + \hat{\mathbf{P}}^{(V)}. \tag{20}$$

The potential part of the pressure tensor originates from the molecular interaction, and it's related with the two-body density distribution (Keizer, 2012):

$$\nabla_{\mathbf{r}}\cdot\hat{\mathbf{P}}^{(V)}(\mathbf{r};t) = -\int\mathbf{f}_{1,2}(\mathbf{r}_1,\mathbf{r}_2)\rho(\mathbf{r}_1,\mathbf{r}_2;t)d\mathbf{r}_2, \tag{21}$$

where $\mathbf{f}_{1,2} \equiv -\nabla_{\mathbf{r}}\cdot\Gamma(\mathbf{r}_1,\mathbf{r}_2)$ denotes the force acting on particle 1 from particle 2 as $\Gamma(\mathbf{r}_1,\mathbf{r}_2)$ being the interaction potential in-between. $\rho(\mathbf{r}_1, \mathbf{r}_2; t)$ is the time-dependent two-body density distribution. One can interpret $\nabla_\mathbf{r} \cdot \hat{\mathbf{P}}^{(V)}(\mathbf{r}; t)$ as the time-dependent mean interaction force at position $\mathbf{r}$. The kinetic part of the pressure tensor is defined as:

$$\begin{aligned} \hat{\mathbf{P}}^{(K)}(\mathbf{r}; t) &= \int d\mathbf{p} \tfrac{[\mathbf{p}-\mathbf{p}(\mathbf{r};t)]\otimes[\mathbf{p}-\mathbf{p}(\mathbf{r};t)]}{m} f^{(1)}(\mathbf{r}, \mathbf{p}; t) \\ &= \int d\mathbf{p} \tfrac{\mathbf{p}\otimes\mathbf{p}}{m} f^{(1)}(\mathbf{r}, \mathbf{p}; t) - \tfrac{\rho(\mathbf{r};t)}{m} \mathbf{p}(\mathbf{r}; t) \otimes \mathbf{p}(\mathbf{r}; t), \end{aligned} \tag{22}$$

where $f^{(1)}(\mathbf{r}, \mathbf{p}; t)$ is the one-body probability function (Zhao and Wu, 2011). The kinetic pressure tensor $\hat{\mathbf{P}}^{(K)}$ originates from the molecular collision, and $\nabla_\mathbf{r} \cdot \hat{\mathbf{P}}^{(K)}(\mathbf{r})$ represents the entropic force due to the variation of local colloidal density.

Then we apply the following identity:

$$\begin{aligned} &\frac{\rho(\mathbf{r}; t)}{m} \mathbf{p}(\mathbf{r}; t) \cdot \nabla_\mathbf{r} \mathbf{p}(\mathbf{r}; t) - \nabla_\mathbf{r} \cdot [\frac{\rho(\mathbf{r}; t)}{m} \mathbf{p}(\mathbf{r}; t) \otimes \mathbf{p}(\mathbf{r}; t)] \\ &= -\mathbf{p}(\mathbf{r}; t) \nabla_\mathbf{r} \cdot [\frac{\rho(\mathbf{r}; t)}{m} \mathbf{p}(\mathbf{r}; t)], \end{aligned} \tag{23}$$

and meanwhile note that $\mathbf{j}(\mathbf{r}; t) = \frac{1}{m} \rho(\mathbf{r}; t) \mathbf{p}(\mathbf{r}; t)$ denotes the particle flux and the continuity equation reads (Zhao and Wu, 2011):

$$\frac{\partial}{\partial t} \rho(\mathbf{r}; t) = -\nabla_\mathbf{r} \cdot \mathbf{j}(\mathbf{r}; t), \tag{24}$$

and then we rewrite Eq. (19) as:

$$\begin{aligned} \frac{\partial \mathbf{j}(\mathbf{r}; t)}{\partial t} + \gamma \mathbf{j}(\mathbf{r}; t) = -\nabla_\mathbf{r} \cdot \int d\mathbf{p} \frac{\mathbf{p} \otimes \mathbf{p}}{m^2} f^{(1)}(\mathbf{r}, \mathbf{p}; t) + \frac{1}{m} \\ \times \int \mathbf{f}_{1,2}(\mathbf{r}_1, \mathbf{r}_2) \rho(\mathbf{r}_1, \mathbf{r}_2; t) d\mathbf{r}_2 \\ + \frac{\rho(\mathbf{r}; t)}{m} \mathbf{f}^{\mathrm{ex}}(\mathbf{r}). \end{aligned} \tag{25}$$

So far, we have made no approximations, and the above equations are exact. It is at this stage in the derivation that we make approximations to derive DDFT. First, we assume the system is in near-equilibrium state, and the one-body probability function satisfies the Maxwell-Boltzmann distribution:

$$f^{(1)}(\mathbf{r}, \mathbf{p}; t) \equiv f^{(1)}_{\mathrm{MB}}(\mathbf{r}, \mathbf{p}; t) = \rho(\mathbf{r}; t) \left(\frac{\beta}{2\pi m} \right)^{3/2} \exp\left(-\beta \frac{p^2}{2m} \right). \tag{26}$$

With this assumption, the first term on the right hand side of Eq. (25) can be evaluated as:

$$\begin{aligned} &\int d\mathbf{p} \tfrac{\mathbf{p}\otimes\mathbf{p}}{m^2} f^{(1)}_{\mathrm{MB}}(\mathbf{r}, \mathbf{p}; t) \\ &= \tfrac{\rho(\mathbf{r};t)}{m^2} \int d\mathbf{p} \begin{pmatrix} p_x p_x & p_y p_x & p_z p_x \\ p_x p_y & p_y p_y & p_z p_y \\ p_x p_z & p_y p_z & p_z p_z \end{pmatrix} \left(\tfrac{\beta}{2\pi m}\right)^{3/2} \exp\left[-\tfrac{\beta p^2}{2m}\right], \\ &= k_B T \tfrac{\rho(\mathbf{r};t)}{m} \widehat{\mathbf{1}} \end{aligned} \tag{27}$$

where $\hat{\mathbf{1}}$ is the three-dimension unit vector. Hence:

$$\begin{aligned} \nabla_\mathbf{r} \cdot \int d\mathbf{p} \tfrac{\mathbf{p}\otimes\mathbf{p}}{m} f^{(1)}(\mathbf{r}, \mathbf{p}; t) &= k_B T \nabla_\mathbf{r} \cdot \tfrac{\rho(\mathbf{r};t)}{m} \widehat{\mathbf{1}} \\ &= \tfrac{k_B T}{m} \nabla_\mathbf{r} \rho(\mathbf{r}; t) \\ &= \tfrac{\rho(\mathbf{r};t)}{m} \nabla_\mathbf{r} [k_\mathrm{B} T \ln \rho(\mathbf{r}; t)], \\ &= \tfrac{\rho(\mathbf{r};t)}{m} \nabla_\mathbf{r} \mu^{\mathrm{id}}(\mathbf{r}; t) \end{aligned} \tag{28}$$

where $\mu^{\mathrm{id}}(\mathbf{r}; t) = k_\mathrm{B} T \ln[\rho(\mathbf{r}; t) \Lambda^3]$ is the ideal part of local chemical potential.

Second, we assume that the two-body density distribution in a non-equilibrium system is the same as in the corresponding equilibrium fluid sharing the same one body density profile, i.e. we apply the following sum rule, which is exact for equilibrium fluids (Archer and Evans, 2004):

$$\frac{1}{m} \int \mathbf{f}_{1,2}(\mathbf{r}_1, \mathbf{r}_2) \rho(\mathbf{r}_1, \mathbf{r}_2; t) d\mathbf{r}_2 = k_\mathrm{B} T \frac{\rho(\mathbf{r}_1)}{m} \nabla_\mathbf{r} c(\mathbf{r}_1; t), \tag{29}$$

where $c(\mathbf{r}_1; t)$ is the one-body direct correlation function.

The chemical potential determined by Eq. (6) within the framework of DDFT contains three contributions:

$$\frac{\delta F[\rho(\mathbf{r}; t)]}{\delta \rho(\mathbf{r}; t)} = \mu^{\mathrm{id}}(\mathbf{r}; t) + \mu^{\mathrm{ex}}(\mathbf{r}; t) + V_{\mathrm{ext}}(\mathbf{r}; t). \tag{30}$$

We employ the familiar relation $\mu^{\mathrm{ex}}(\mathbf{r}; t) = -k_\mathrm{B} T c(\mathbf{r}; t)$, and combine Eqs. (28)–(30), we can rewrite Eq. (25) as:

$$\frac{\partial \mathbf{j}(\mathbf{r}; t)}{\partial t} + \gamma \mathbf{j}(\mathbf{r}; t) = -\frac{\rho(\mathbf{r}; t)}{m} \nabla_\mathbf{r} \frac{\delta F[\rho(\mathbf{r}, t)]}{\delta \rho(\mathbf{r}, t)}. \tag{31}$$

Eq. (31) and Eq. (24) form a closed set of dynamical equations for the local density, $\rho(\mathbf{r}; t)$ and the local flux, $\mathbf{j}(\mathbf{r}; t)$. As the density functional, $F[\rho(\mathbf{r}, t)]$, is approximated by that of the corresponding equilibrium system, Eq. (31) is known as the DDFT.

If we apply divergence on both sides of Eq. (31) and then substitute Eq. (24) into the resulting equation, we immediately recover Archer's formulation in Eq. (9). It's worth noting at this point that if we further assume the friction coefficient γ is very large so that the first term on the left hand side of Eq. (31) is negligible, Eq. (31) recoveries to Eq. (8), which is the original DDFT by Marconi and Tarazona (1999), Marconi and Tarazona (2000). For pure solvent system, we have $\gamma = 0$, Eq. (31) reduces to (setting $m = 1$):

$$\frac{\partial \mathbf{j}(\mathbf{r}; t)}{\partial t} = -\rho(\mathbf{r}; t) \nabla_\mathbf{r} \frac{\delta F[\rho(\mathbf{r}; t)]}{\delta \rho(\mathbf{r}; t)}. \tag{32}$$

The above equation coupled with Eq. (24) recovers to the dynamical equation proposed by Bagchi and his coworkers (Burghardt and Bagchi, 2006; Bagchi and Chandra, 1991; Bagchi and Bhattacharyya, 2001).

5. Extension of DDFT

The intrinsic relation between the Navier-Stokes equation and DDFT discussed above casts helpful insights for further extending the DDFT. Till date DDFT has been extensively employed to investigate molecular diffusion such as gas species in porous materials and ionic diffusion in electrochemical systems during the charge–discharge processes. In chemical engineering, molecular diffusion is often coupled with fluid flow (Yu et al., 2017), and the flow has non-neglectable effect on the diffusion dynamics when the system size reduces to nanoscale. For example, Mao and Sinnott (2000) utilized molecular dynamics simulations to investigate the flow of methane, ethane, and ethylene through carbon nanotubes, they found that the fluid–fluid interaction and fluid-wall interaction both had strong impact on molecule flow and diffusion processes. Ding et al. (2019) fabricated a novel electrochemical membrane reactor with a gas diffusion cathode for H_2O_2 generation, they found that the oxygen flow and gas diffusion near the catalytic layer have an enhancement on oxygen utilization efficiency and H_2O_2 productivity. Nevertheless the coupling between the fluid flow and molecular diffusion can't be properly addressed by the existing versions of DDFT.

Inspecting the derivation above, we notice that the approximate evaluations of pressure tensor lead to different versions of DDFT. Motivated by this treatment, we re-examine the calculation of pressure tensor by additionally considering the flow effect, and thus further extend DDFT for addressing the coupling between the fluid flow and molecular diffusion.

For a fluid flow system with $\mathbf{p}(\mathbf{r}; t) \neq 0$, we have derived the one-body probability distribution with the help of maximum information entropy principle in our previous work, and it reads (Zhao and Wu, 2011):

$$f^{(1)}(\mathbf{r},\mathbf{p};t)=\rho(\mathbf{r};t)\left(\frac{\beta}{2\pi m}\right)^{3/2}\exp\left(-\beta\frac{[\mathbf{p}-\mathbf{p}(\mathbf{r};t)]^{2}}{2m}\right), \tag{33}$$

and hence after some algebra we have:

$$\int d\mathbf{p}\frac{\mathbf{p}\otimes\mathbf{p}}{m^{2}}f^{(1)}(\mathbf{r},\mathbf{p};t)=k_{B}T\frac{\rho(\mathbf{r};t)}{m}\hat{\mathbf{1}}+\frac{\rho(\mathbf{r};t)}{m^{2}}\mathbf{p}(\mathbf{r};t)\otimes\mathbf{p}(\mathbf{r};t). \tag{34}$$

The divergence of above equation gives:

$$\nabla_{\mathbf{r}}\cdot\int d\mathbf{p}\frac{\mathbf{p}\otimes\mathbf{p}}{m^{2}}f^{(1)}(\mathbf{r},\mathbf{p};t)=\frac{\rho(\mathbf{r};t)}{m}\nabla_{\mathbf{r}}\mu^{\mathrm{id}}(\mathbf{r};t)+\nabla_{\mathbf{r}}\cdot[\frac{\rho(\mathbf{r};t)}{m^{2}}\mathbf{p}(\mathbf{r};t)\otimes\mathbf{p}(\mathbf{r};t)]. \tag{35}$$

Substituting the above equation into Eq. (25), and meanwhile applying the approximation in Eq. (29), we reach:

$$\frac{\partial\mathbf{j}(\mathbf{r};t)}{\partial t}+\gamma\mathbf{j}(\mathbf{r};t)=-\frac{\rho(\mathbf{r};t)}{m}\nabla_{\mathbf{r}}\frac{\delta F[\rho(\mathbf{r};t)]}{\delta\rho(\mathbf{r};t)}+\nabla_{\mathbf{r}}\cdot[\frac{\rho(\mathbf{r};t)}{m^{2}}\mathbf{p}(\mathbf{r};t)\otimes\mathbf{p}(\mathbf{r};t)]. \tag{36}$$

Apparently, if we further assume $\mathbf{p}(\mathbf{r};t)\otimes\mathbf{p}(\mathbf{r};t)=0$, the above equation immediately recovers to Eq. (31), i.e., the DDFT equation for pure diffusion process. The above equation with the combination of Eq. (24) gives a closed set of governing equations for addressing the coupling between fluid flow and diffusion dynamics.

Another extension of DDFT can be obtained if we take divergence on the both sides of above equation and then substitute the continuity equation, which reads:

$$\frac{\partial^{2}\rho(\mathbf{r};t)}{\partial t^{2}}+\gamma\frac{\partial\rho(\mathbf{r};t)}{\partial t}=-\nabla_{\mathbf{r}}\cdot\{\frac{\rho(\mathbf{r};t)}{m}\nabla_{\mathbf{r}}\frac{\delta F[\rho(\mathbf{r};t)]}{\delta\rho(\mathbf{r};t)}\}+\nabla_{\mathbf{r}}\cdot\{\nabla_{\mathbf{r}}\cdot[\rho(\mathbf{r};t)\mathbf{u}(\mathbf{r};t)\otimes\mathbf{u}(\mathbf{r};t)]\}. \tag{37}$$

Similarly, if the diffusion coefficient γ is large so that $\frac{\partial^{2}\rho(\mathbf{r};t)}{\partial t^{2}}$ is ignorable, the above equation can be further simplified as:

$$\frac{\partial\rho(\mathbf{r};t)}{\partial t}=-\nabla_{\mathbf{r}}\cdot\{\frac{\rho(\mathbf{r};t)}{\gamma m}\nabla_{\mathbf{r}}\frac{\delta F[\rho(\mathbf{r};t)]}{\delta\rho(\mathbf{r};t)}\}+\nabla_{\mathbf{r}}\cdot\frac{1}{\gamma}\{\nabla_{\mathbf{r}}\cdot[\rho(\mathbf{r};t)\mathbf{u}(\mathbf{r};t)\otimes\mathbf{u}(\mathbf{r};t)]\}. \tag{38}$$

Note that the above equation indicates that the evolution of local density is composed of two parts, namely the molecular diffusion (the first term on the right hand side) and the flow effect (the second term on the right hand side). If we ignored the flow effect, then Eq. (38) recovers to the original DDFT equation in Eq. (10). Eq. (36), Eq. (37) and Eq. (38) represent three extended versions of DDFT, and they provide theoretical foundation for describing the molecular diffusion, e.g., adsorptive diffusion onto solids surfaces, affected by fluid flow.

It should be noted that many chemical processes are in stationary states, in which the fluid density is irrelevant with time, i.e., $\partial\rho(\mathbf{r})/\partial t=0$. In this circumstance, Eq. (38) simplifies to:

$$0=-\nabla_{\mathbf{r}}\cdot\left\{\frac{\rho(\mathbf{r})}{\gamma m}\nabla_{\mathbf{r}}\frac{\delta F[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})}\right\}+\nabla_{\mathbf{r}}\cdot\frac{1}{\gamma}\{\nabla_{\mathbf{r}}\cdot[\rho(\mathbf{r})\mathbf{u}(\mathbf{r})\otimes\mathbf{u}(\mathbf{r})]\}. \tag{39}$$

By integrating both sides of the above equation, and considering the mass conservation equation, we have:

$$\frac{\rho(\mathbf{r})}{\gamma m}\nabla_{\mathbf{r}}\frac{\delta F[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})}=\frac{1}{\gamma}\{\nabla_{\mathbf{r}}\cdot[\rho(\mathbf{r})\mathbf{u}(\mathbf{r})\otimes\mathbf{u}(\mathbf{r})]\}, \tag{40}$$

which can be rewritten as:

$$\nabla_{\mathbf{r}}\frac{\delta F[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})}=\frac{m}{\rho(\mathbf{r})}\{\nabla_{\mathbf{r}}\cdot[\rho(\mathbf{r})\mathbf{u}(\mathbf{r})\otimes\mathbf{u}(\mathbf{r})]\}. \tag{41}$$

We assume this system is in contact with a bulk reservoir, and then integrate above equation from position $\mathbf{r}$ to the bulk system, we have:

$$\frac{\delta F[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})}-\mu_{\mathrm{bulk}}=\int_{\mathbf{r}}^{\mathrm{bulk}}\frac{m}{\rho(\mathbf{r}')}\{\nabla_{\mathbf{r}}\cdot[\rho(\mathbf{r}')\mathbf{u}(\mathbf{r}')\otimes\mathbf{u}(\mathbf{r}')]\}d\mathbf{r}', \tag{42}$$

where μ_{bulk} is the chemical potential of the fluid component in the bulk system. The integral on the right hand side of above equation can be considered as a driving potential originating from the hydrodynamic effect, which can be represented by an effective potential, $\phi_{\mathrm{f}}(\mathbf{r})$, satisfying:

$$\rho(\mathbf{r})\nabla_{\mathbf{r}}\phi_{\mathrm{f}}(\mathbf{r})=-m\nabla_{\mathbf{r}}\cdot[\rho(\mathbf{r})\mathbf{u}(\mathbf{r})\otimes\mathbf{u}(\mathbf{r})], \tag{43}$$

and then Eq. (42) can be rewritten as:

$$\frac{\delta F[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})}=\mu_{\mathrm{bulk}}-\phi_{\mathrm{f}}(\mathbf{r}). \tag{44}$$

The above equation represents the governing equation for local density profile in steady systems. By following the similar procedure as in the static density functional theory, we can obtain the generalized Euler-Lagrange equation from eq.(44), namely:

$$\rho(\mathbf{r})=\rho_{\mathrm{b}}\exp\left[\beta\mu_{\mathrm{bulk}}^{\mathrm{ex}}-\beta\phi_{\mathrm{f}}(\mathbf{r})-\beta V_{\mathrm{ext}}(\mathbf{r})-\frac{\delta\beta F^{\mathrm{ex}}[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r})}\right]. \tag{45}$$

Under hydrostatic condition, one naturally has $\phi_{\mathrm{f}}(\mathbf{r})=0$, and then the above equation recovers to the standard Euler-Lagrange equation (Hansen and McDonald, 2013; Zhao et al., 2015). It should be noted that the standard Euler-Lagrange equation can be derived from the existing three versions of DDFT, i.e., Eqs. (8), (9), and (10), under equilibrium condition. The generalized Euler-Lagrange equation beyond the standard one highlights the significant improvement of our extended DDFT, and more importantly, the generalized Euler-Lagrange equation indicates that the local density profile can be regulated not only by external potential but also by fluid flow, and this interesting feature stimulates us with a novel path to intensify and regulate relevant engineering processes including interface reaction and mass separation.

6. Conclusion

In this work, we prove that DDFT, a molecular dynamical theory, can be derived from the general Naiver-Stokes equations with near-equilibrium assumptions. Our derivation, starting from the momentum balance equation of Navier-Stokes equations, involves a generalized pressure tensor as a kernel. By employing various approximations to simplify the pressure tensor, various versions of DDFT can be deduced. This derivation casts helpful insights into the extension of DDFT. Stimulated by this procedure, we further extend DDFT to the dynamics systems in which molecular diffusions are closely coupled with fluid flow. To this end, we re-evaluate the pressure tensor by additionally considering flow effect. The extended DDFT provides a feasible approach to address the flow effect on both molecular diffusion and local density profile.

Finally, it's worth noting that the developed methods of FMT and FMSA within the framework of density functional theory can be, respectively, regarded as individual semi-analytical solutions to the free energy contributions from repulsive interaction and the attractive interaction. This physical concept that the free energy can be divided into two different contributions may provide a potential route to overcome the Navier-Stokes analytical solution problem.

CRediT authorship contribution statement

Chongzhi Teng Qiao Zhao: Conceptualization, Methodology, Writing - original draft, Conceptualization, Methodology, Writing - review & editing. **Xiaochen Yu:** Methodology, Validation, Writing - review & editing. **Leying Qing:** Validation, Writing - review & editing. **Bo Bao:** Validation, Resources, Writing - review & editing. **Shuangliang Zhao:** Resources, Writing - review & editing, Supervision. **Honglai Liu:** Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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