Investigation of Isotropic Laplacian Operators by Computer Simulation for A-B Diblock Copolymers

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Summary
The cell dynamics simulation (CDS) technique is elaborated for the lamellar system of A-B diblock copolymer. In which, the averaging operator (Laplacian) plays a key role for ensuring the isotropy. To achieve this objective, the original Laplacian used in CDS has been replaced by various other Laplacian schemes to observe more isotropic results. Generally, the CDS is used to evaluate the Time–Dependent Ginzburg–Landau (TDGL) equations where analysis remains inconclusive for optimal isotropy. In this paper, the stencil size with focus on its behavior for two dimensional Laplacian operators are considered. In simulations, the short stencil 5-point 5P was found anisotropic produced anisotropic results while the 9-point PK, BV1, BV2 and BV3 were found isotropic.

Key words:
Laplacian Operators, CDS, Isotropy, Diblock copolymers

1. Introduction

In mathematics, the differential or integral operators have been applied to several numerical solutions. The Laplacian (Δ) operator that comes from the divergence of the gradient and the form of this operator is given as follows [1]:

$$\nabla.(\nabla\psi) = \Delta\psi = \sum_{i=0}^{n} \frac{\partial^2\psi}{\partial x_i^2},$$
$$for\ x_i = x, y, z \ldots . dimensions \qquad (1)$$

where ∇ is called Nabla. For numerical solutions or approxiamtions, the discretizations of these operators are carried out to satisfy certain conditions using several methods by maintaining stability, accuracy, conservation and consistency. Usually, the discretizations of operators are made free from anisotropies. In fact, the anisptropic operators are containing error terms (or artefacts) in the any numerical approximation of partial differential equations (PDEs) [1]–[3]. The cell dynamics simulation method, for diblock polymers, involves PDEs and the discretizaiton of mathematical operator, such as Laplacian operator, is considered an essential feature. The use of isotropic Laplacians help to relieve anisotropies in describing the phenomenon of physical system, i.e., diblock copolymers. If the discretization of a Laplacian operator lacks isotropy or is not optimally isotropic, then the different values are generated from two identical edges formed at different angles. In numerical approximation, such type of anisotropic descretization of operator produces artefacts (error terms) due to which outlining edges remain inexplicit [4]. Therefore, the study of isotropic Laplacians is considered an important part and in this work two-dimensional Laplacian operators have been thoroughly investigated for cell dynamics simulation (CDS) method which has been used for the modelling of A-B forming system in diblock polymers. Mainly, the stability and isotropy of the CDS depend on these Laplacians. The Forward Euler's method as a finite difference approach had been used in original CDS method [5]. Several researchers have talked about employing isotropic Laplacians for different methods, i.e., Thampi et al. [1] suggested two- and three-dimensional Laplacians for Hybrid Lattice Boltzman simulation method. Various discretizations techniques have been carried out for approximating Laplacians to handle anistropic terms and to put best possible isotropic Laplacians in stencils. The isotropy of a two-dimensionl isotropic 9-point stencil and many 27-point stencil formulas have been discussed completely based on spatial discretizations. The quantification of the Laplacian schemes has been given by a rigorous comparison by using Fourier transformations at different planes. For a 9-point Laplacian, two methods are used for a 9-point Laplacian: the finite difference and finite volume method. The 9-point stencil was observed less isotropic in the spatial discretization through finite volume method. To measure isotropy, a comparison has been done for a 9-point Laplacian with an optimal isotropic 9-point Laplacian which was used by Shinozaki and Oono in a study of spindol decomposition based on CDS [5], [6]. It must be noted that in this work, the comparisons of stencils will

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also be done with Shinozaki and Oono's 9-point isotropic Laplacian [5].

Various studies have been done regarding the isotropic Laplacians. A finite difference time domain (FDTD) method called the Yee algorithm was developed to approximate Maxwell's equations where two-dimensional 5-point, 9-point and 25-point stencils were put to practice to avoid anistorpic Laplacians [7]. A better version of the Yee algorithm was introduced by applying FDTD to approximate partial differential operator (Laplacian) for Maxwell's equations. These schemes of finite difference methods were based on high-dimensional isotropic operators which reduced the anisotropies to larger extent [8]. Kumar [9] also worked on isoropic finite difference for Laplacians, he explains that anisotropy produces due to the directional bias of artefacts in the finite difference approximations. He has proposed a finite difference scheme with lowest error order terms with exception of directional bias.

In this work, the isotropic Laplacian schemes have been discussed and the numerical results have presented. These Laplacian operators are from CDS equations and the numerical results are obtained from simulations of A-B diblock copolymer lamellae forming system. In section 2 a brief introduction of A-B diblock copolymer for lamellae forming system is given along with the CDS frame work. In section 3, the isotropic Laplacian operator are discussed and derived. Section 4 discusses simulation results based on different Laplacian schemes and section 5 gives conclusion.

2. Cell Dynamics Simulation (CDS) method used for A-B diblock copolymers

Block copolymer is basically the two or more chemical blocks formed in a long chain of molecules of polymers. This chain comes into formation due to the molecular reaction. These molecules are called monomers. The block copolymer structures are found in melts, solutions, blends or solvents with different ranges [10]. The size of these chemical blocks range from nanometre to millimetre on the scale [11]. The polymer blocks in a chain of molecules are covalently bonded into one macromolecule and the blocks of two different monomers A and B are called A-B diblock, A-B-A triblock or multiblock copolymers [12]. The block copolymers, due to their innate ability can shape of nanostructures like lamellae, spheres, hexagonal-packed cylinders, gyroid, body-centred cubic structures, etc. [13], [14].

There are many applications of block copolymers in various fields, these are mainly applied by creating templates in soft nanotechnology and nanoelectronics. Furthermore, these are applicable for mechanical flow fields, nanoparticle synthesis, catalyst materials, electric fields, separation nonporous membranes, photonic crystals [15]–[19]. The applications of block copolymers are also found in medical science, i.e. drug delivery, and in commercial products such as bottle stoppers and jelly candles [20].

In science laboratories, the chemical bonding in polymers are observed through microscopies and further theories and interpretations of these complex systems are made based on mathematical models for their use in various applications. These processes are very long and time-consuming which require a lot effort. Therefore, the computer technology is an aided advantage and is inevitable to use. Thus, the study and the analysis of these complex systems become more convenient and take less time for the orientation in a broader view through simulations [13].

The block copolymers have been studied through various simulation techniques which include Molecular Dynamics (MD) [11], Lattice Boltzman (LB) method [11], [21], Dissipative Particle Dynamics (DPD) [22], Brownian Dynamics (BD) [23], Monte Carlo (MC) [24], Self-Consistent Field Theory (SCFT) [25] and Time Dependent Ginzburg Landau (TDGL) via CDS method [11], [13], [14], [26]. The cellular automaton CDS technique of Ginzburg Landau type is commonly used for modelling block copolymers due to its better efficiency, more accuracy and fast speed. The CDS is employed to study the evolution of an order parameter in microphase sepration in binary blends, alloys and diffusive structures in block copolymers [5], [6], [27]–[29].

The frame work of CDS for modelling lamellae forming system of A-B diblock copolymer is presented here and the numerical results obtained from simulations using isotropic Laplacians are presented in Results section. An order parameter $\psi(t,i)$ of block copolymer with time variable t evaluated at cell i in a grid can be defined as follows [14]:

$$\psi = \phi_A - \phi_B + (1-2f) \tag{2}$$

where ϕ_A and ϕ_B are local volume fractions of A and B. Total volume fraction of monomer A is defined as $f = N_A/(N_A + N_B)$ and the evolution of an order parameter in a single cell:

$$\psi(t+1,i) = g(\psi(t,i)) \tag{3}$$

where $g(\psi)$ is the map function. In a nonconserved case for diffusive dynamics, the time evolution of an order parameter is given as:

$$\psi(t+1,i) = g\big(\psi(t,i)\big) + D[\langle\langle\psi(t,i)\rangle\rangle - \psi(t,i)]$$

$$= \mathcal{T}[\psi(t,i)] \quad (4)$$

In equation (4), D is taken as phenomenological diffusion constant in diffusive dynamics term and the $\langle\langle X\rangle\rangle - X$ is the isotropized discrete Laplacian. The two-dimensional form of this Laplacians in a square lattice is:

$$\langle\langle\psi(t,i)\rangle\rangle = W_1 \sum_{NN} \psi(t,i) + W_2 \sum_{NNN} \psi(t,i) \quad (5)$$

where Ws are considered as wegiths, e.g., $W_1 = 1/6$ and $W_2 = 1/12$ and NN and NNN stand for nearest neighbours and next nearest neighbours [13], [14]. The artefacts which generate due to the interrelation between its neighbouring cells, the conserve case is considered to maintain isotropy and equilibrium in the system. After a net change in the cells, the order parameter in a conserved case is given as follows:

$$\mathcal{T}[\psi(t,i)] - \psi(t,i) \quad (6)$$

and the CDS for order parameter evolution becomes:

$$\psi(t+1,i) = \mathcal{T}[\psi(t,i)] - \langle\langle\mathcal{T}[\psi(t,i)] - \psi(t,i)\rangle\rangle. \quad (7)$$

The equation (7) with an additional term $-B\psi(t,i)$ becomes:

$$\psi(t+1,i) = \mathcal{T}[\psi(t,i)]$$

$$-\langle\langle\mathcal{T}[\psi(t,i)] - \psi(t,i)\rangle\rangle - B\psi(t,i). \quad (8)$$

Equation (7) is finite difference approximation of the Cahn-Hilliard-Cook (CHC) equation [13]:

$$\frac{\partial\psi}{\partial t} = \mathrm{K}\nabla^2\left[\frac{\delta F[\psi]}{\delta\psi}\right], \quad (9)$$

where is $F[\psi]$ a free energy functional and K is the phenomenological mobility constant. Equation (3) and (8) together can be written as [13]:

$$\mathcal{T}(t+1,i) = g\big(\psi(t,i)\big) - B\psi(t,i)$$

$$+D[\langle\langle\psi(t,i)\rangle\rangle + \psi(t,i)] \quad (10)$$

and the map function $g\big(\psi(t,i)\big)$ in equation (10) is given as:

$$g(\psi) = [1 + \tau - A(1-2f)^2\,]\psi$$

$$-v(1-2f)\psi^2 - u\psi \quad (11)$$

where τ denotes temperature and A,v,u are constants [13], [14].

3. Laplacian Schemes

Two-dimensional Laplacian schemes and their properties are discussed and in a form, that these Laplacian schemes are used in CDS method is also elaborated. The Laplacian in equation (1) in the Fourier space can be written as:

$$-\nabla.\nabla \Leftrightarrow \mathrm{k}^2 \quad (12)$$

where k is wave factor and the finite difference approximation of equation (12) is:

$$\Delta = S + O(h^n) \quad (13)$$

where S denotes stencils (grid cells) and $O(h^n)$ denotes truncation error up to order n.

Basically, the use of mathematical operators is essential in describing physical phenomena. Therefore, isotropy in numerical approximations of these operators must be ensured for better results. Nevertheless, the Laplacian operator is rotationally invariant but all the discretizations of this operator are not isotropic. The isotropy in natural phenomena can be defined as an identical appearance or behaviour of an object when it is measured or viewed from any direction. In other words, the isotropy is considered as homogeneousness in all positions [30]. The discretization of any Laplacian scheme is considered isotropic if it is without directional bias or preferences otherwise anisotropic due to the presences of directional bias [9].

The Laplacian schemes are formulated depending the size of the stencils, e.g., two-dimensional 5-point stencil, 9-point stencil or 25-point stencil formula and three-dimensional 7-point stencil or 27-point stencil formula. We are discussing here only the two-dimensional 9-point Laplacian schemes. In Figure 1, a two-dimensional 9-point stencil can be seen and we use DmQn notation for a model Laplacian to observe the number of dimensions m and the number of points n used for numerical calcualtion.

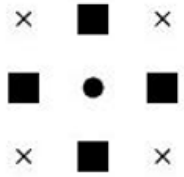

Fig. 1 The 9–point stencil shape of Laplacian

The first scheme can be formulated including its central cell (see dark circle in Figure 1) and the nearest neighbouring points to central cell (see dark square boxes). These be in total 5 points and a 5-point stencil is formed, that is D2Q5 [4], [31]. The Laplacian operator for D2Q5 is given in equation (14) and the Laplacian operators for D2Q9 are given in equations (15) and (16).

$$\Delta(\psi)_{P5} = \frac{1}{h^2}\left[\sum_{i=1}^{4}\psi_i^{(1)} - 4\psi^{(0)}\right] \quad (14)$$

$$\Delta(\psi)_{PK} = \frac{1}{6h^2}\left[4\sum_{i=1}^{4}\psi_i^{(1)} + 4\sum_{i=1}^{4}\psi_i^{(2)} - 20\psi_i^{(0)}\right] \quad (15)$$

$$\Delta(\psi)_{OP} = \frac{1}{2h^2}\left[\sum_{i=1}^{4}\psi_i^{(1)} + \frac{1}{2}\sum_{i=1}^{4}\psi_i^{(2)} - 6\psi_i^{(0)}\right] \quad (16)$$

The subscript P5 in equation (14) is meant to show that the Laplacian operator is based on 5-point stencil formula and the subscripts PK and OP in equations (15) and (16) stand for Patra-Karttunen and Oono-Puri respectively. Patra and Karttunen [32] have mentioned equation (14) to be anisotropic and Equation (15) to be isotropic. Puri and Oono employed equation (16) for simulation of spindol decomposition via CDS and they obtained optimal isotropic results of simulation images. Tomita [33] also mentioned Equation (16) to be the optimal choice for isotropy. The Equations (14), (15) and (16) for their corresponding Fourier transforms are given as follows:

$$\Gamma(\mathrm{k})_{P5} = \frac{2}{(\Delta x)^2}\left[\cos(\mathrm{k}_x\Delta x) + \cos(\mathrm{k}_y\Delta x) - 2\right] \quad (17)$$

$$\Gamma(\mathrm{k})_{PK} = \frac{2}{3(\Delta x)^2}\left[2\cos(\mathrm{k}_x\Delta\mathrm{x}) + 2\cos(\mathrm{k}_y\Delta\mathrm{x}) + \frac{1}{2}\left[\cos[(\mathrm{k}_x + \mathrm{k}_y)\Delta x] + \cos[(\mathrm{k}_x - \mathrm{k}_y)\Delta x]\right] - 5\right] \quad (18)$$

$$\Gamma(\mathrm{k})_{OP} = \frac{1}{2(\Delta x)^2}\left[2\cos(\mathrm{k}_x\Delta\mathrm{x}) + 2\cos(\mathrm{k}_y\Delta\mathrm{x}) + \cos[(\mathrm{k}_x + \mathrm{k}_y)\Delta x] + \cos[(\mathrm{k}_x - \mathrm{k}_y)\Delta x] - 6\right] \quad (19)$$

Expanding $\cos(x)$ in the equations (17), (18) and (19) at x = 0 gives respectively:

$$\Gamma(\mathrm{k})_{5P} = -(\mathrm{k}_x^2 + \mathrm{k}_y^2) + \frac{(\Delta x)^2}{12}(\mathrm{k}_x^2 + \mathrm{k}_y^2)^2 - \frac{(\Delta x)^2}{6}\mathrm{k}_x^2\mathrm{k}_y^2 + O(\mathrm{k}^6) \quad (20)$$

$$\Gamma(\mathrm{k})_{PK} = -(\mathrm{k}_x^2 + \mathrm{k}_y^2) + \frac{(\Delta x)^2}{12}(\mathrm{k}_x^2 + \mathrm{k}_y^2)^2 + \frac{(\Delta x)^2}{3}\mathrm{k}_x^2\mathrm{k}_y^2 + O(\mathrm{k}^6) \quad (21)$$

$$\Gamma(\mathrm{k})_{OP} = -(\mathrm{k}_x^2 + \mathrm{k}_y^2) + \frac{(\Delta x)^2}{12}(\mathrm{k}_x^2 + \mathrm{k}_y^2)^2 + \frac{(\Delta x)^2}{12}\mathrm{k}_x^2\mathrm{k}_y^2 + O(\mathrm{k}^6) \quad (22)$$

The Laplacian operators have been studied since long time for their isotropic properties in various two- and three-dimensional stencil types. The work in [34] proposed a different technique to evaluate a three-dimensional isotropic 27-point stencil operator. This technique and stencil operator was also used by Fraaije et al. [35] to enhance the efficiency and accuracy of a linkage operator in a standard lattice model. In this model, they achieved the optimal isotropy by employing proper scaling conditions for the stencil operator. In this article, the same technique has been applied for a two-dimensional 9-point family stencil operators (Laplacians). Likewise, we calculate the weights d_α for a 9-point stencil operator and the method is following.

Consider a half point finite difference:

$$D_\alpha(f)(x) = \sum_{\alpha=1}^{m=9} d_\alpha \frac{f\left(x+\frac{h}{2}r_\alpha\right) - f\left(x-\frac{h}{2}r_\alpha\right)}{\|hr_\alpha\|} \quad (23)$$

In Fourier space, the discrete half point derivative operator D_α in direction α is:

$$D_\alpha = \frac{2i}{\|hr_\alpha\|}\sin\left(\frac{h\mathrm{k}r_\alpha}{2}\right) \quad (24)$$

where r_α is a grid direction in positive half–space:

$$r_\alpha = \begin{Bmatrix}(1,0)\\(0,1)\end{Bmatrix} d_{10}.$$

$$r_\alpha = \begin{Bmatrix}(1,1)\\(1,-1)\end{Bmatrix} d_{11}. \quad (25)$$

A vector length |k| is taken as positive half space where $\mathrm{k} = \pi$ and The directions are fixed for vectors as $(|\mathrm{k}|, 0)$ and $(|\mathrm{k}|/\sqrt{2}, |\mathrm{k}|/\sqrt{2})$. The symbol $S(\mathrm{k})$ is used as $\nabla^2 = -\mathrm{k}.\,\mathrm{k}$.

$$-q^2 \to S(\mathrm{k}) = \sum_\alpha d_a D_\alpha D_\alpha \quad (26)$$

The values of weights d_a are evaluated by employing following two scaling and isotropy conditions given respectively in equations (27) and (28):

$$\frac{\partial s^2}{\partial x_i^2} = -2, \text{ where } x_i = x, y \quad (27)$$

$$S(\pi, 0) = S\left(\frac{\pi}{\sqrt{2}}, \frac{\pi}{\sqrt{2}}\right) \quad (28)$$

From the above method of [34] (B.A.C van Vlimmeren), the stencil is obtained with weights $d_{10} = 0.53015$ and $d_{10} = 0.53015$ and $d_{11} = 0.469849$, The stencil BV1 is given as follows:

$$\mathrm{S}(\mathrm{k})_{BV1} = 0.53015\left(-4sin\left[\frac{\mathrm{k}_x\Delta x}{2}\right]^2 - 4sin\left[\frac{\mathrm{k}_y\Delta x}{2}\right]^2\right) + 0.469849\left(-2sin\left[\frac{(\mathrm{k}_x+\mathrm{k}_y)\Delta x}{2}\right]^2 - 2sin\left[\frac{(\mathrm{k}_x+\mathrm{k}_y)\Delta x}{2}\right]^2\right). \quad (29)$$

For $BV1$ case the vector choice is taken as $S(\pi, 0) = S(\pi/\sqrt{2}, \pi/\sqrt{2})$. There are two other cases $BV2$ and $BV3$ where the vector choices vary. For $BV2$ case, the vector choice is fixed as $S(\pi/2{,}0) = S(\pi/2\sqrt{2}, \pi/2\sqrt{2})$ and for $BV3$ case, the vector choice is taken as $S(3\pi/4{,}0) = S(3\pi/4\sqrt{2}, 3\pi/4\sqrt{2})$. The stencils for $BV2$ and $BV3$ case are given as follows:

$$\mathrm{S}(\mathrm{k})_{BV2} = 0.63778\left(-4sin\left[\frac{\mathrm{k}_x\Delta x}{2}\right]^2 - 4sin\left[\frac{\mathrm{k}_y\Delta x}{2}\right]^2\right) + 0.362218\left(-2sin\left[\frac{(\mathrm{k}_x+\mathrm{k}_y)\Delta x}{2}\right]^2 - 2sin\left[\frac{(\mathrm{k}_x+\mathrm{k}_y)\Delta x}{2}\right]^2\right). \quad (30)$$

$$S(k)_{BV3} = 0.597131\left(-4sin\left[\frac{k_x\Delta x}{2}\right]^2 - 4sin\left[\frac{k_y\Delta x}{2}\right]^2\right) + 0.402869\left(-2sin\left[\frac{(k_x+k_y)\Delta x}{2}\right]^2 - 2sin\left[\frac{(k_x+k_y)\Delta x}{2}\right]^2\right). \quad (31)$$

The stencils in equations (29)-(31) are in k domain and for employing in CDS technique for simulation these need to be transformed to real analogues. The corresponding real analogues of equations (29)-(31) are given as follows:

$$S(k)_{BV1} = \frac{1}{(\Delta x)^2}\{0.53015(2\cos(k_x\Delta x) + 2\cos(k_y\Delta x)) + 0.234925(2\cos[(k_x+k_y)\Delta x] + 2\cos[(k_x-k_y)\Delta x])\} \quad (32)$$

$$S(k)_{BV2} = \frac{1}{(\Delta x)^2}\{0.63778(2\cos(k_x\Delta x) + 2\cos(k_y\Delta x)) + 0.362218(2\cos[(k_x+k_y)\Delta x] + 2\cos[(k_x-k_y)\Delta x])\} \quad (33)$$

$$S(k)_{BV3} = \frac{1}{(\Delta x)^2}\{0.597131(2\cos(k_x\Delta x) + 2\cos(k_y\Delta x)) + 0.402869(2\cos[(k_x+k_y)\Delta x] + 2\cos[(k_x-k_y)\Delta x])\} \quad (34)$$

In CDS the form of the Laplacian is used as $\ll X \gg -X$ and correspondingly the we need modify the Laplacian operators in the form $\ll X \gg$ to be used in simulations, the modifications for Laplacian operators (14)-(16) and (29)-(31) are given as follows:

$$\ll \psi \gg_{5P} = \frac{1}{4}\sum_{NN}\psi \quad (35)$$

$$\ll \psi \gg_{PK} = \frac{1}{5}\sum_{NN}\psi + \frac{1}{20}\sum_{NNN}\psi \quad (36)$$

$$\ll \psi \gg_{OP} = \frac{1}{6}\sum_{NN}\psi + \frac{1}{12}\sum_{NNN}\psi \quad (37)$$

$$\ll \psi \gg_{BV1} = 0.173235\sum_{NN}\psi + 0.076765\sum_{NNN}\psi \quad (38)$$

$$\ll \psi \gg_{BV2} = 0.63778\sum_{NN}\psi + 0.362218\sum_{NNN}\psi \quad (38)$$

$$\ll \psi \gg_{BV3} = 0.597131\sum_{NN}\psi + 0.402869\sum_{NNN}\psi \quad (38)$$

In simulation results section, we refer Laplacian operators with their subscripts used. For equations (14)-(16) and (29)-(31), the subscripts are referred as 5P, PK, OP, BV1, BV2 and BV3 respectively.

4. Simulation Results

A coarse–grained discretization CDS technique is used to model the A–B diblock copolymers [13] where various two-dimensional Laplacian operators are employed to investigate isotropic simulation results. For this purpose, a detailed discussion has been given to understand the evolution of order parameter in microphase separation and front propagation for A–B dibblock copolymer systems via CDS method. Fortran 90 programming language is used for CDS code. The CDS code was run on the machine with Linux 3.7 desktop Opensuse 12.3.

Following are the steps in CDS algorithm by considering the discrete equations (10) and (11):

1. Setting random initial values for order parameter ψ;
2. Setup of periodic boundary conditions in both dimensions, i.e. x and y
3. Evaluation of first (inner) Laplacian for order parameter ψ, i.e. $\ll \psi \gg -\psi$;
4. Mutiplying step 3 by diffusion constant D, i.e. $D[\ll \psi \gg -\psi]$;
5. Evaluation of the map function (11) and assigning parameter to specific constants;
6. Combining steps 4 and 5; see equation (10);
7. Evaluation of second (outer) discrete Laplacian for the result of step 6, as follows:
   $\ll \Gamma(t,i) \gg -\Gamma(t,i)$;
8. Computing new values for an order parameter;

A morphology defines shapes which are found during the evolution process in microphase separation of A–B diblock copolymers at different times. Different morphologies were discovered: lamellae, bicontinuous, cylinders and spheres [14]. The constant values in map function (see equation (11)) decide any certain morphology. The simulation details are presented Table 1 [14].

Table 1: Simulation parameters

τ	f	u	v	B	D	A	Morphology
0.36	0.48	0.38	2.3	0.02	0.7	1.5	Lamellae
0.33	0.44	0.38	2.3	0.02	0.5	1.5	Bicontinuous
0.30	0.40	0.38	2.3	0.02	0.4	1.5	Cylinders
0.20	0.40	0.38	2.3	0.01	0.5	1.5	Spheres

We choose lamellae morphology for the simulations. For all simulations, the grid size was chosen to be 128×128 with grid spacing $h = \Delta x = \Delta y = 1$ and at the initial state, the parameter was set randomly by $\psi = \pm 0.3$. Simulations were performed without any specific time scale to generate numerical values of ψ.

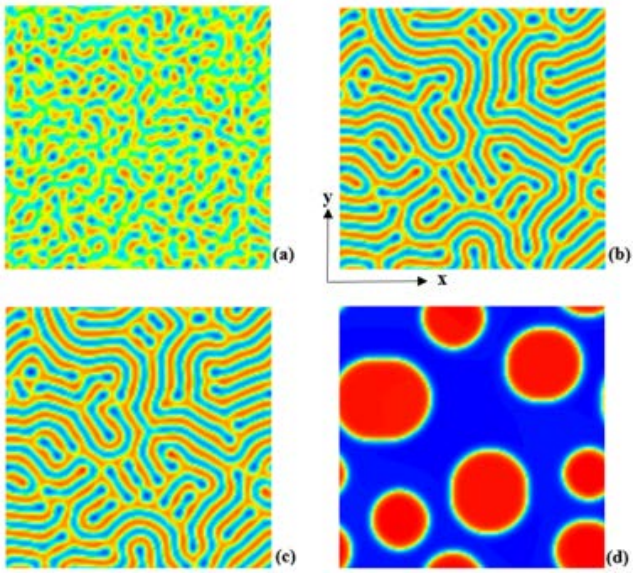

Fig. 2 Results of CDS based on OP (9–point stencil), equation (16).

Real space simulation snapshots in (a), (b) and (c) are for 100th, 10000th and 100000th time steps by following Table 2. Real space simulation snapshot in (d) is for 100000th by following Table 3.

Table 2: System parameters for Lamellae morphology

CDS Parameters	τ	f	u	v	B	D	A	Initial random values
Lamellae Morphology	0.36	0.48	0.38	2.3	0.02	0.7	1.5	$\psi_i = \pm 0.3$

Table 3: System parameters used in cell dynamical method for binary blend

CDS Parameters	τ	f	u	v	B	D	A	Initial random values
Lamellae Morphology	0.36	0.48	0.38	2.3	0.0	0.7	1.5	$\psi_i = -0.3 \pm 0.01$

The simulation results in Figure 2 were obtained by using OP of D2Q9 model Laplacian (equation (16)). In Figure 2 (a), (b) and (c), the snapshots show different stages of evolution of lamellae in a microphase separation of A–B diblock copolymers. In Figure 2 (a), the microphase separation starts with respect to time and lamellae can be seen with some shapes in Figure 2 (b). In Figure 2 (c), the red coloured lamellae with an interfacial yellow colour are basically microphase–separated in either A or B block. The simulation for lamellar forming system becomes stable at 10000th time step as shown in Figure 2 (b), that is, the microphase separation has reached at its equilibrium state. Simulation results in snapshots (a), (b) and (c) of Figure 2 were obtained by using the parameter values given in Table 2. On the other hand, (d) snapshot of Figure 2 shows a binary blend where OP Laplacian scheme was employed and the simulation parameters were used from Table 3. It is also lamellar forming system simulation by setting B=0. A macrophase separation takes place rather than microphase separation in a pore system. In this case, two subdomains can be seen where yellow interfacial parts macrophase-separate red coloured A-rich and blue coloured B-rich subdomains. The well-defined lamellae formations in Figure 2 (b) and (c) are basically isotropic. It must be noted that in this work, the simulation results in Figure 2 are referred as default CDS results. All the 2D simulation results based on other Laplacian schemes will be compared with these default results.

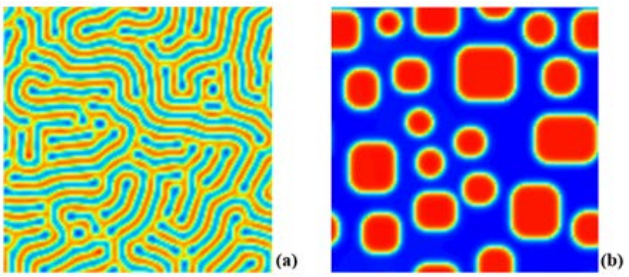

Fig. 3 Laplacian scheme 5P

In Figure 3, the simulation results were obtained by using Laplacian scheme 5P. In snapshots (a) and (b) of Figure 3, the parameters were employed from Table 2 and Table 3 respectively. It can be observed that the lamellae formations in Figure 3 (a) are not very well-defined

compared to Figure 2 (c). In Figure 3 (b), subdomains form rectangular shapes which create a clear difference compared to Figure 2 (d). Therefore, it can be inferred that the Laplacian scheme 5P did not perform well to produce isotropic results and it can be termed as anisotropic Laplacian scheme.

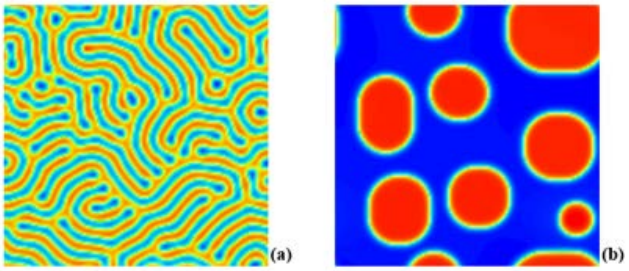

Fig. 4 CDS results based on Laplacian scheme PK

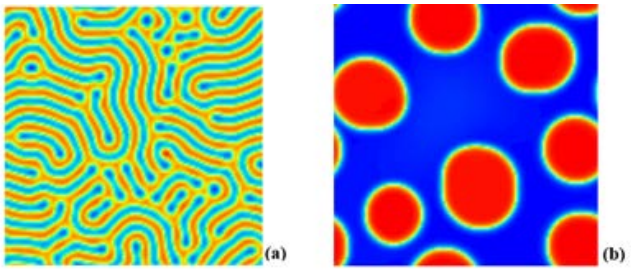

Fig. 5 CDS results based on Laplacian scheme BV1

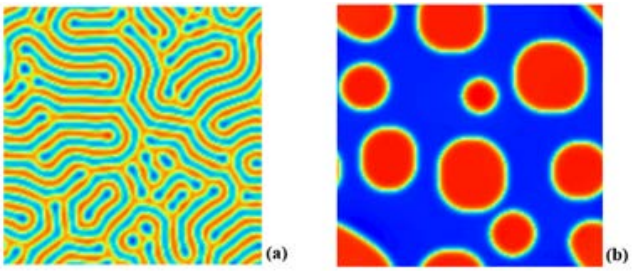

Fig. 6 CDS results based on Laplacian scheme BV2

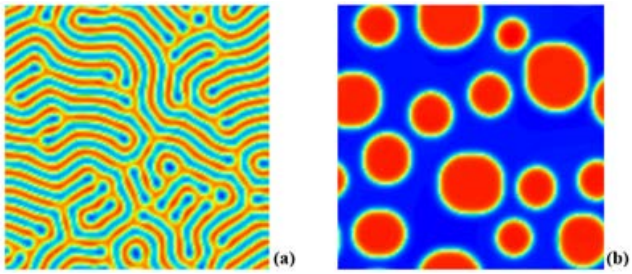

Fig. 7 CDS results based on Laplacian scheme BV3

The CDS results given in Figures 4,5,6 and 7 were obtained using Laplacian schemes PK, BV1, BV2 and BV3 respectively. It can be observed in snapshot (a) of these figures, the lamellae formations are well aligned and well defined and proper lamellar chains are proper in shape. The Laplacian schemes PK, BV1, BV2 and BV3 produce isotropic results of microphase separation. The snapshots in (b) parts of these figures were obtained the similar results as those of default CDS using Laplacian OP, i.e. circular shapes of red subdomains. Thus, these Laplacians performed at an optimum level for the simulations.

5. Conclusion

The stencil, as a computational molecule, is considered an important entity for the evolution of an order parameter. Therefore, this study has been carried out for the investigation of several different two-dimensional stencil operators based on Laplacian schemes. It is analysed that the original averaging (Laplacian OP) operator is not only the discrete representation of Laplacian in CDS and in CDS, the TDGL equations should not only be considered for producing isotropy. From the simulations, the 5P Laplacian scheme was found unstable for simulations and not on isotropic scale as required. On the other hand, PK was found isotropic and performed like default Laplacian scheme OP. The 9-point isotropic BV1, BV2 and BV3 stencils were found isotropic and stable for simulations like default CDS Laplacian scheme. The study of different isotropic discrete Laplacian schemes provides a direction to use more isotropic Laplacian schemes and to avoid the grid related artefacts (anisotropies).

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