Rolling Dynamics of Nanoscale Elastic Shells Driven by Active Particles

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ABSTRACT: Self-propelled elastic shells capable of transducing energy to rolling motion could have potential applications as drug delivery vehicles. To understand the dynamics of the nanoscale size elastic shells, we performed molecular dynamics simulations of shells filled with a mixture of active and passive beads placed in contact with an elastic substrate. The shell skin is made of cross-linked polymer chains. The energy transduction from active beads to elastic shell results in stationary, steady rolling, and accelerating states depending on the strength of the shell–substrate adhesion and the magnitude of a force applied to the active beads. In the stationary state, the torque produced by a friction (rolling resistance) force in the contact area balances that due to the external force generated by the active beads, and the shell sticks to the substrate. In the steady rolling state, a rolling friction force balances the driving force, and the shell maintains a constant rolling velocity. The scaling relationship between the magnitude of the driving force and the shell velocity reflects a viscoelastic nature of the shell skin deformation dynamics. In the accelerating state, the energy supplied by active beads exceeds the energy dissipation due to viscoelastic shell deformation in the contact area. Furthermore, the contact area of the shell with a substrate decreases with increasing shell instantaneous velocity.

INTRODUCTION

Energy transduction into mechanical work is a foundation for the operation of the molecular motors and machines.1–7 For example, a flagellar molecular motor controls bacteria motility through whiplike rotation of the tail. Listeria uses polymerization of the host cell actin into cross-linked filaments for its propulsion. Cells convert polymerization of the actin cytoskeleton at the edge of the cell membrane into directional motion. A variety of the mechanisms utilized in the biological systems for biochemical energy conversion into mechanical work serve as an inspiration for the design of an artificial machine capable of directional transport of a "cargo".8–11

For molecular machines moving along surfaces the energy is transformed into a rolling or sliding motion which starts with overcoming the rolling or sliding resistance. For nanoscale size objects the resistance is caused by an adhesion to a substrate.2,12 In the case of rolling, the origin of the rolling resistance is a hysteresis loss in mechanical work required for creation of a new contact area at the front edge and elimination of a contact area at the back edge of an advancing roller.1,2,14,15 Similar processes are involved in sliding. While understanding the mechanics of the rolling and sliding contacts is the classical topic of the contact mechanics,16–19 going back to the seminal paper by Reynolds,20 the application of the contact mechanics frameworks to elucidate driving forces controlling the dynamics of the self-propelled molecular machines is lagging behind.

To address these shortcomings, we used coarse-grained molecular dynamics simulations to model the rolling motion of an elastic shell filled with “cargo” and active particles. An external force exerted on an active particle is transferred to the shell causing its rolling motion. This takes place when the torque generated by active particles overcomes the rolling resistance between a shell and substrate. Varying the adhesion strength between elastic shell and substrate and the magnitude of the applied forces, we identify different states of shell motion. According to our simulations an elastic shell could be in the stationary state, move with a constant velocity when the net applied force is balanced by the rolling friction force, or accelerate. In each of these states we have established scaling relations between the contact area of the elastic shell and substrate, shell velocity, and strength of the shell–substrate interactions.

RESULTS AND DISCUSSION

To establish universal features of shell motion generated by applying a constant force to active particles (beads) filling a shell interior, we have performed coarse-grained molecular dynamics simulations of a shell on a solid substrate (see Figure 1). A spherical shell with initial thickness \( d_s \) and radius \( R_s \) (Figure 1a) was made by cross-linking bead—spring chains of Lennard-Jones (LJ) beads with diameter \( \sigma \). The cavity of the elastic shell was filled with active/passive LJ-beads with 1:9 and 2:8 ratio (see Figure 1b). The elastic shell was placed on the solid substrate made of the LJ-beads with diameter \( \sigma \) arranged into a face-centered cubic (FCC) lattice with a lattice constant

Received: September 7, 2018
Published: November 1, 2018
equal to \( \sigma \) (see Figure 1c). A constant force \( f \) with the magnitude varying between 0.05\( k_B T/\sigma \) and 0.95\( k_B T/\sigma \) (\( k_B \) is the Boltzmann constant, and \( T \) is the absolute temperature) was applied to active beads along the positive \( x \) direction. The temperature of the system during the shell motion was maintained constant by utilizing the substrate as a heat sink. The interaction parameters used in simulations and simulation details are discussed in the Methods section.

Equilibrium Shell Contact. We begin our discussion by analyzing an equilibrium contact between an elastic shell filled with particles and a substrate. To make a connection between the strength of the interaction potential and the macroscopic contact properties of the elastic shell, we have calculated the work of adhesion \( W \) between an elastic shell and a substrate. The work of adhesion was obtained from a set of potential of interaction parameters for substrate–shell pairs being equal to 0.8\( k_B T \), 1.0\( k_B T \), 1.2\( k_B T \), 1.5\( k_B T \), and 2.0\( k_B T \), respectively. Insets show typical shell deformation and definition of the shell deformation parameters.

\[
U_{eq}^{\text{shell}} \propto E d_o \Delta h^2
\]  

(1)

The main difference between eq 1 and the classical expression for the elastic energy of a particle\(^{17,23,24}\) is the approximation used for deformation volume \( a^3d_o \). Note that this approximation is valid as long as the contact radius is larger than the shell thickness, \( a > d_o \). If the opposite inequality holds, one has to use \( a^3 \) for deformation volume, and the expression for the elastic energy reduces to the classical expression for particle deformation.\(^{17,23,24}\) In the limit of larger deformations, for which a contact radius is on the order of the shell radius \( a \approx R_s \) the compression deformation of a portion of the shell into a planar disk provides the dominant contribution to its elastic energy. In this case the elastic energy of shell deformation is calculated to be\(^{25}\)

\[
U_{eq}^{\text{shell}} \propto E d_o \Delta h^3/R_s
\]  

(2)

The equilibrium shell deformation, \( \Delta h \), is obtained by balancing the elastic energy with the change of the free energy for creation of a contact between shell surface and substrate. This free energy change is proportional to\(^{16,17,23}\)

\[
\Delta F_{adh} \propto -W a^2 \approx -W R_s \Delta h
\]  

(3)

In rewriting eq 3 in terms of the shell deformation \( \Delta h \) we take into account that a contact radius can be approximated as \( a \approx \sqrt{2R_s \Delta h} \). Minimizing the elastic energy given by eqs 1 or 2 and surface free energy change upon contact eq 3 with respect to the shell deformation \( \Delta h \), we obtain for equilibrium contact radius \( a \propto R_s^{1/2} \) and \( a \propto W^{1/4} \), respectively. This is in a good agreement with the simulation results shown in Figure 2.

Dynamics of the Elastic Shells. Having established equilibrium contact properties, we now discuss how the dynamics of the elastic shells on the adhesive substrates depends on the magnitude of the applied force and the strength of the substrate–shell interactions. All our simulations
of the shell motion were initiated from an equilibrium state of the shell in contact with a substrate (see Figure 3a). In the initial state, all beads are uniformly distributed over the cavity volume. After an external force $f$ is applied to each active bead, the active and passive beads begin to separate with all the active beads concentrating on the right side of the cavity as shown in Figure 3b. This concentration of the active beads in a particular region of the shell cavity produces distributed force acting on the shell skin causing its asymmetric deformation. There is a direct correlation between the magnitude of the net force generated by active beads and shell deformation. There are three states of the elastic shell in response to a constant force being applied to the active beads—stationary state, steady rolling state, and accelerating state as illustrated in Figure 4. Under applied external force the contact area first changes its shape as the shell begins to roll (see Figure 5). In the steady rolling state, the contact area remains constant. This is followed by the contact area decrease as the shell accelerates.

The elastic shell remains at rest if the net torque generated by the applied forces is lower than that required to overcome the adhesive contact. The applied torque corresponding to the net force from all active particles $F$ can be estimated as $M_{\text{ext}} \propto FR_c$. Due to this torque an adhesive contact deforms asymmetrically with the front edge having a slightly higher curvature (see Figure 5) and the center of the mass of the shell moving forward from the center of mass of the contact area (see Figure 6a). Deformation of the contact area produces an asymmetric pressure distribution resulting in a restoring moment which counteracts the external torque. The restoring moment $M_c$ was shown to be proportional to the shell pull-out force $F_p \propto WR$, times $R \theta$, $M_c \propto F_p R \theta^{1/2}$ (where $\theta$ is the shell bending angle defined in Figure 6a). Thus, at equilibrium $M_{\text{ext}} = M_c$, resulting in the bending angle to be $\theta \propto F/F_p$. In Figure 6b we show a dependence of the bending angle on the normalized net force. The three data sets collapse into a universal curve confirming the relationship between the bending angle and the force ratio. However, the data set corresponding to the largest work of adhesion between substrate and elastic shell demonstrates a systematic deviation from universal behavior and only approaches a universal curve in the steady rolling state. This could be explained by the dominance of the flattening mode of the shell deformations for which the pull-out force is different by a numerical coefficient.

In the steady rolling state the bending angle continues to increase with the magnitude of the applied force as shown in Figure 6b; however, the contact area between shell and substrate remains unchanged (see Figure 5). In this state, the shell velocity is a monotonically increasing function of the net applied force $F$ (see Figure 7a). In the steady rolling state, the energy supplied by the active particles per unit time is dissipated in the contact area between substrate and shell. The main mechanism for energy dissipation is the viscoelastic response of the shell skin to deformation. The rolling motion of the shell results in a time dependent deformation of each section of the shell as it enters the contact zone on the right at time $t = 0$ and leaves it on the left at time $t = t_{\text{con}} = 2a/v$. The compression strain produced by shell deformation can be approximated by a sinusoidal strain function with magnitude $\epsilon_m \approx \Delta h/a$ and frequency $\omega \approx \pi/t_{\text{con}}$

$$\epsilon(t) \approx \epsilon_m \sin(\omega t) \approx (\Delta h/a) \sin(\pi vt/2a)$$  \hfill (4)

The average rate of energy density dissipation by shell compression during the contact time $t_{\text{con}}$ is on the order of

$$\dot{U}(\omega) = \frac{1}{\omega} \int_0^{t_{\text{con}}} \sigma(t) d\epsilon(t) \propto \epsilon_m^2 \omega E''(\omega)$$  \hfill (5)

where $\sigma(t)$ is the time dependent stress generated by the strain $\epsilon(t)$, of which the out of phase (dissipative) component is equal to $\epsilon_m E''(\omega) \cos(\omega t)$ ($E''(\omega)$ is the frequency dependent Young’s loss modulus). The total rate of energy dissipation due to contact is obtained by multiplying $\dot{U}$ by the deformation volume $d_a a^2$

$$\dot{U}(v) \propto d_a^2 \epsilon_m^2 \omega E''(\omega) \propto d_a \Delta h^2 v E''(v)/a$$  \hfill (6)

In the steady rolling state we have

$$(F - F_v) v \approx \dot{U}(v) \approx d_a \Delta h^2 v E''(v)/a$$  \hfill (7)

where $F_v$ is a crossover force to the steady rolling state. This equation defines shell velocity as a function of the applied force. For the range of the rolling speeds $v$ such that the storage modulus is frequency independent, $E''(v) \approx E$, the shell deformation $\Delta h$ can be estimated by minimizing eqs 1 and 3, which results in $\Delta h \approx WR_e/Ed_b$ and $Ed_b \Delta h^2 \approx Wa^2$. In this case the ratio $v/a$ is a universal function of $(F - F_v)/Wa$ which follows from the expression

1539 DOI: 10.1021/acscentsci.8b00632
ACS Cent. Sci. 2018, 4, 1537–1544
There is a very simple interpretation of the right-hand side (r.h.s.) of eq 8a. It is proportional to the rate of change of the contact free energy \( vW_a \) times an efficiency coefficient \( \tan(\delta) \) (8a).

\[
(F - F_r)v \approx vW_a E''(v)/E \approx vW_a \tan(\delta(v))
\]

\[
(F - F_r)/W_a \approx (v \tau_R/a)^\beta
\]

There is a very simple interpretation of the right-hand side (r.h.s.) of eq 8a. It is proportional to the rate of change of the contact free energy \( vW_a \) times an efficiency coefficient \( \tan(\delta(v)) \). In simplifying eq 8b we take into account that for polymeric networks the loss modulus is a power law function \( E''(\omega) = E(\omega \tau_R)^\beta \) where exponent \( \beta = 1 \) for \( \omega \tau_R \ll 1 \), and \( \beta = 0.5 \) in the frequency range \( \omega \tau_R \gg 1 \) (\( \tau_R \) is the Rouse time of the network strands).\(^{26} \)

Note that, in the case of shell flattening deformation, the analysis leads to a scaling relation similar to eqs 8a and 8b, but one should expect a different numerical coefficient. (For these calculations one has to set \( \varepsilon_m \approx \Delta h/R_s \) and deformation volume to be on the order of \( a^2 d_o \)). In Figure 7b we test the relationship given by eq 8b for systems undergoing steady state rolling. Indeed we have been able to collapse data corresponding to work of adhesion \( W \) varying between 1.64 \( k_B T/\sigma \) and 3.0 \( k_B T/\sigma \). Note that the data set with \( W = 4.2 \) \( k_B T/\sigma \), corresponding to the regime where shell flattening dominates its deformation, is shifted by a constant. This confirms our assessment that the shift is due to a numerical prefactor.

A shell accelerates when the solution of eq 7 disappears. This takes place when the shell deformation becomes velocity dependent. In this state the storage modulus is on the order of the loss modulus and can be approximated as \( E'(\omega) \approx E'(\omega) \approx E(\omega \tau_R)^{0.5,26} \). In this case, to obtain the rate of energy dissipation we have to explicitly take into account dependence.
of the contact radius, $a$, on the shell velocity, $v$. Substituting $E'(v) \approx E(v) (W/Ed)^{2/3}$ into eq 1 and balancing elastic energy of the shell deformation with the surface free energy change upon contact (eq 3) we have

$$a(v) \approx \left(\frac{v}{v_0}\right)^{-1/3} \left(\frac{WR^2}{Ed}\right)^{2/3} \approx a_0 \left(\frac{a_0}{vR}\right)^{1/3}$$

(9)

where we introduced equilibrium contact radius $a_0 \approx R_0 (W/Ed)^{1/2}$. Thus, the contact radius decreases with increasing rolling velocity as $a \propto v^{-1/3}$. The decrease in the contact radius results in the decrease of the rate of the energy dissipation

$$\dot{U}(v) \approx vW_0 (v) E'(v) / E(v) \approx v^{2/3} W_0 (a_0 / vR)^{1/3}$$

(10)

Figure 8 shows the velocity dependent dissipation function $\dot{U}(v)$ given by eqs 6 and 10 and the graphic solution of eq 7. It follows from this figure that a solution disappears when the applied force exceeds a critical value, $F > F_{acc}$. In this range of applied forces, $F > F_{acc}$, the shell moves with acceleration (see Figure 4). Note that this can only take place if there is a range of the shell velocities where the energy dissipation function grows weaker than linear with the shell velocity $v$ (see eq 10).

When the elastic shell accelerates, the contact area monotonically decreases with increasing shell speed (see Figure 9a). In particular, in this state the contact area scales with the instantaneous shell velocity as $S \propto v^{-1}$. Note that this is a stronger dependence of the contact area on the shell velocity than the one estimated from simple energy balance calculations, $S \approx a(v)^2 \propto v^{-2/3}$ (see eq 9). This discrepancy could be due to the fact that during acceleration the shell is thinner in the direction of the axis of rotation, and contact area becomes more asymmetric (see Figure 5). It is interesting to point out that we can collapse all curves into one universal plot by normalizing the contact area $S$ by their values in the steady rolling state, $S_\infty$, and normalizing the instantaneous velocity by its crossover value to the accelerating state, $v_{acc}$. The results of this procedure are shown in Figure 9b.

Figure 10 shows the diagram of different shell states in a plane of the net applied force $F$ and work of adhesion $W$ based on simulation results for 67 studied systems. It follows from this diagram that for each value of the work of adhesion there is a sequence of transitions from stationary state to steady rolling state and finally to accelerating state. The boundaries between these transitions shift to the right with increasing strength of the substrate shell interactions manifested in increase of the work of adhesion, $W$. It is also important to point out that with increasing the work of adhesion the range of forces corresponding to steady rolling states widens.

**CONCLUSIONS**

We have shown that there are three dynamic states (see Figure 10) of the elastic shell in contact with the elastic substrate whose motion is generated by encapsulated active particles. In the stationary state the torque induced by active particles is balanced by the restoring momentum due to asymmetry of the adhesive contact with the substrate. The bending angle, which defines the shell’s center of mass deviation from zero active force state, is a universal function of the ratio of the net applied force $F$ and the pull-out force for adhesive contact as shown in Figure 6b. In the stationary rolling state the steady state velocity is a scaling function of the applied force, $v \propto (F - F_r)^\alpha$.
with exponent $\alpha$ representing different regimes in the loss modulus dependence on the frequency (see eqs 8a and 8b and Figure 7). This also confirms the importance of the viscoelastic properties of the shell materials for friction and interface strength.\textsuperscript{27} It is important to point out that, during steady rolling, a contact area between elastic shell and substrate is independent of the shell velocity (see Figure 9a). The shell accelerates when its velocity reaches a critical value which increases with increasing work of adhesion $W_a$. In this state, the contact area between shell and substrate is inversely proportional to its instantaneous velocity (see Figure 9b). The approach presented here could become a valuable tool for verification of the model assumptions and predictions used to describe the contact mechanics of rolling objects.\textsuperscript{16, 17} It also could be used for modeling and analysis of motion of microcapsules and cells under different external conditions.\textsuperscript{28–33}

At the end we want to point out that to make connection with experimental systems one has to use dimensionless parameters characterizing macroscopic system properties. For example, the net applied force $F$ generating shell motion is convenient to normalize by the characteristic force of adhesion $W_a$. The shell velocity $v$ should be normalized by $a/\tau_R$ describing the crossover between different relaxation regimes in the frequency dependent modulus of the shell. Such data representation reflects the physical processes governing dynamics of rolling shells.

\section{METHODS}

\bf{Molecular Dynamics Simulations.} A spherical shell (Figure 1a) was prepared by cross-linking 5988 bead–spring chains consisting of $N = 32$ Lennard-Jones (LJ) beads (monomers) with diameter $\sigma$. Each chain has in average 20 cross-links connecting it with the surrounding chains. The shell preparation and equilibration procedures have followed the approach developed in refs 21, 22, and 34. The cavity of the elastic shell was filled with 8244 beads (particles) (see Figure 1b) having 1:9 and 2:8 ratio of the active/passive beads. The elastic shell was placed on the solid substrate made of LJ-beads with diameter $\sigma$ arranged into a face-centered cubic (FCC) lattice with a lattice constant being equal to $\sigma$. There are seven layers of beads with each layer consisting of $200 \times 200$ beads as shown in Figure 1c. The system was periodic in the $x$ and $y$ directions.

The interactions between all beads in a system were modeled by the modified truncated–shifted LJ-potential, which accounts for increased size of the beads through setting the interaction range by $\Delta$. In this representation the interaction potential between two beads separated by a distance $r$ is written as follows:

$$U_{\text{LJ}}(r) = \begin{cases} 
4\epsilon_{\text{LJ}} \left[ \left( \frac{\sigma}{r - \Delta} \right)^{12} - \left( \frac{\sigma}{r - \Delta} \right)^6 \right] - \left( \frac{\sigma}{r_{\text{cut}}} \right)^{12} & r \leq r_{\text{cut}} + \Delta \\
+ \left( \frac{\sigma}{r_{\text{cut}}} \right)^6 & r_{\text{cut}} + \Delta < r < r_{\text{cut}} + \Delta \\
0 & r > r_{\text{cut}} + \Delta 
\end{cases}$$

(11)

where $\epsilon_{\text{LJ}}$ is the strength of the LJ-interaction potential, and $r_{\text{cut}}$ is the cutoff distance. The value of the cutoff distances $r_{\text{cut}}$ and the values of the Lennard-Jones interactions parameters, $\epsilon_{\text{LJ}}$, in

![Figure 9](image1.png)

Figure 9. (a) Dependence of the instantaneous contact area $S$ on the shell instantaneous velocity $v$ for different values of substrate–shell interaction parameter $\epsilon_{\text{LJ}}$: 0.8$k_B$T (blue triangles), 1.0$k_B$T (red triangles), 1.2$k_B$T (green triangles), and 1.5$k_B$T (purple triangles). (b) Dependence of the normalized contact area $S/S_0$ on the normalized shell velocity $v/v_{\text{acc}}$. Inset shows dependence of the crossover velocity on work of adhesion. Notations are the same as in panel a.

![Figure 10](image2.png)

Figure 10. Diagram of states of elastic shells.
terms of the thermal energy $k_BT$ between different pairs are summarized in Table 1.

Table 1. Parameters of Interaction Potentials

| interaction\(a\) | $\xi_{ij}[k_BT]$ | $r_{cut}[^\circ]$ | $\Delta[^\circ]$ |
|------------------|-----------------|-----------------|-----------------|
| AB–AB            | 1.0             | 2/16            | 0.5             |
| IB–IB            | 1.0             | 2/16            | 0.5             |
| AB–IB            | 1.0             | 2/16            | 0.5             |
| AB–CS            | 1.0             | 2/16            | 0.25            |
| IB–CS            | 1.0             | 2/16            | 0.25            |
| CS–CS            | 1.0             | 2.5             | 0.0             |
| Sub–CS           | 0.8, 1.0, 1.2, 1.5, 2.0 | 2.5             | 0.0             |
| Sub–Sub          | 1.0             | 2.5             | 0.0             |

\(a\)AB, active beads; IB, inactive (passive) beads; Sub, substrate beads; CS, beads belonging to cross-linked shell.

The connectivity of beads in polymer chains, cross-linking bonds, and bonds connecting beads forming a substrate were modeled by the harmonic springs described by the following potential

$$U_{\text{bond}}(r) = \frac{1}{2} k_{\text{spring}} (r - r_0)^2$$

(12)

where $k_{\text{spring}} = 200k_BT/\sigma^2$ is the spring constant, and $r_0 = 0.72\sigma$ is the equilibrium bond length.

To model shell rolling, we have applied a constant force, $f_i$ to active beads along the positive $x$ direction. The equation of motion for the $i$th bead (including shell beads, active and passive beads, and beads forming top three layers of the substrate) is written as follows:

$$m \frac{d\mathbf{v}_i(t)}{dt} = F_i(t) + f_i n_x$$

(13)

where $m$ is the mass of bead which set to unity for all beads in a system, $\mathbf{v}_i(t)$ is the bead velocity, and $F_i(t)$ represented the net deterministic force acting on the $i$th bead. Note that the constant external force, $f_i$ is only applied to active beads. The force values were varied between 0.05$k_BT/\sigma$ and 0.8$k_BT/\sigma$. To maintain a temperature of the system constant during the shell motion, we utilized substrate as a heat sink. In particular, we applied a Langevin thermostat to the three layers (4th, 5th, and 6th layer from the top) of substrate (see Figure 1c). This was achieved by adding the friction, $-\xi \mathbf{v}_i(t)$, and stochastic force, $\mathbf{F}_i^R(t)$, terms into the equation of motion (eq 13) describing motion of the three middle substrate layers. The stochastic force had a zero-average value $\langle \mathbf{F}_i^R(t) \rangle = 0$ and is $\delta$-function correlated, $\langle \mathbf{F}_i^R(t) \mathbf{F}_j^R(t') \rangle = 6\xi k_BT \delta(t-t')$. The friction coefficient $\xi$ was set to $\xi = m/\tau$, where $\tau$ is the standard LJ-time $\tau = \sigma(m/\varepsilon)^{1/2}$, where $\varepsilon = 1.0k_BT$. The velocity–Verlet algorithm with a time step $\Delta t = 0.001\tau$ was used for integration of the equation of motion (eq 13). Note that, in our simulations, the bottom layer of the substrate was fixed ($v = 0$) to prevent substrate from moving due to interaction coupling with a moving elastic shell.

Each simulation run of the shell motion started from the equilibrium state. This state was obtained by running simulations with zero external force applied to all active particles. The duration of each simulation run is varied to achieve the steady state or equilibrium state and had duration up to $10^4\tau$. All simulations were performed using LAMMPS software.\(^{35}\)

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Y.T. and A.V.D. designed the simulations. Y.T. and H.L. performed the computer simulations and analyzed the data. A.V.D. and H.L. supervised the project. The Article was written by A.V.D., Y.T., and H.L. The project was conceived by A.V.D.

#### Funding

National Science Foundation (DMR-1624569).

#### Notes

The authors declare no competing financial interest.
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