Size-dependent dielectrophoretic crossover frequency of spherical particles

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Dielectrophoresis (DEP) has been extensively used in lab-on-a-chip systems for trapping, separating, and manipulating of micro particles suspended in a liquid medium. The most widely used analytic model, the dipole model, provides an accurate prediction on the crossover frequency of submicron particles, but cannot explain the significant drop in crossover frequency of larger particles. Here, we present numerical simulations using the Maxwell stress tensor (MST) and finite element method to study the size effect of the DEP crossover frequency of spherical polystyrene particles suspended in de-ionized water. Our results show that the surface conductance due to the electrical double layer plays a key role, and the size dependency of crossover frequency obtained by the MST method agrees reasonably well with published experimental data. The exponents of the power law are approximately -1.0 and -4.3 for smaller (diameter $< 4.6 \mu\text{m}$) and larger particles (diameter $> 4.6 \mu\text{m}$), respectively. The free surface charge distribution reveals that the charge begins accumulating on the particle equator for particle diameters larger than a critical diameter of $4.6 \mu\text{m}$, a result not captured by the dipolar approximation. This method may be extended to analyze bioparticles with complex shapes and composition, and provides new insights into the interpretation of dielectrophoresis applications using lab-on-a-chip systems. © 2016 AIP Publishing LLC.

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I. INTRODUCTION

Dielectrophoresis (DEP) is a phenomenon in which a neutral polarizable particle, suspended in a medium and subjected to a nonuniform electric field, experiences a net force due to the difference in polarizability between the particle and the medium.¹ The force is directed toward locations with increasing (positive DEP, pDEP) or decreasing (negative DEP, nDEP) field intensity depending on whether the particle is more or less polarizable than the medium at the applied frequency. The direction of the DEP force alternates at a characteristic frequency, known as the crossover frequency. Bioparticles, such as cells, bacteria, and macromolecules, behave as dielectric particles in alternating current (ac) fields, and hence their movement can be controlled by using properly designed electric field and frequency. DEP has been one of the most important and commonly used techniques in lab-on-a-chip systems for manipulation,^{2–5} sorting, separation,^{6–9} and characterization of bioparticles.^{10–16}

Separation of heterogeneous populations of particles into homogeneous subgroups is often achieved by utilizing their distinct crossover frequencies. Hence, the determination of crossover frequency is of great importance. Several experimental methods have been proposed to determine the crossover frequencies of polystyrene (PS) particles by using visual estimation,¹⁷ optical,¹⁸ and curve fitting of the real part of the Clausius-Mossotti factor (CMF).¹⁹ The results show a power law dependence of the crossover frequency on the particle size with an exponent about -0.8 for submicron particles, and an exponent about -4.3 for larger particles. Such

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dramatic change in the exponents of the crossover frequency versus the particle size is still not well understood.

Crossover frequency can also be calculated using DEP theory based on simplified dipole model and more accurate multipolar model. Although the dipolar approximation has been extensively used to calculate the crossover frequency, it is valid only for small spherical particles and for cases where the field does not vary strongly within the particle. Washizu and Jones²⁰ developed general expressions, considering the induced higher-order moments, for the multipolar approximation for a homogeneous sphere in an arbitrary electric field. Schnelle *et al.*²¹ measured the DEP force of a particle in an eight-electrode DEP cage and concluded that the contribution of quadrupole moment forces is significant for particles larger than about a quarter of the electrode gap. Although the multipolar approach has been developed for spherical particles, there are no expressions for nonspherical particles, such as ellipsoidal, biconcave, and cylindrical particles, which are prevalent in biological systems. Maxwell stress tensor (MST) is the most general and rigorous numerical method for calculating the DEP force of particles of various shapes and sizes.^{22–25}

Another important factor that must be considered in the DEP analysis is the particle surface conductance, which consists of the conductance due to the movement of charge in the diffuse double layer and the conductance due to the charge movement in the Stern layer, known as the electrical double layer (EDL). It was demonstrated that the dielectrophoretic behavior of submicron latex spheres is dominated by the composition of the particle surface^{26–28} and influences the DEP behavior greatly.^{17,29}

In this paper, we present numerical simulations using MST and finite element method to calculate the crossover frequencies of PS particles from 200 nm to 6 μm in diameter and to provide a better understanding of the physics that determines the size dependency of the crossover frequency observed experimentally in the literature.^{17–19} Although a large amount of work (over 2000 publications between the year 2000 and 2010)³⁰ has been carried out on the theory, developed technology, and applications of DEP, many of the experimental results were explained using the dipolar approximation even for large cells. This method may be extended to analyze bioparticles with complex shapes and composition and provides new insights into the interpretation of dielectrophoretic crossover frequency for lab-on-a-chip systems.

II. THEORY

A. Dipole model and surface conductance

According to the dipole model, the time-average DEP force F_{DEP} acting on a lossy homogeneous isotropic dielectric spherical particle is given by³¹

$$\langle F_{DEP} \rangle = 2\pi\epsilon_m R^3 \text{Re}[K(\omega)] |\nabla|E_{rms}|^2, \quad (1)$$

where R is the particle radius, ϵ_m is the absolute permittivity of the suspending medium, E_{rms} and ω are the root-mean-square amplitude and angular frequency of the applied ac electric field, respectively, and $\text{Re}[K(\omega)]$ is the real part of the complex CMF, given by

$$K(\omega) = \frac{\tilde{\epsilon}_p - \tilde{\epsilon}_m}{\tilde{\epsilon}_p + 2\tilde{\epsilon}_m}, \quad (2)$$

where $\tilde{\epsilon}_m$ and $\tilde{\epsilon}_p$ are the complex permittivity of the medium and particle, respectively, and the general complex permittivity is given by $\tilde{\epsilon} = \epsilon - \frac{\sigma}{\omega}$, where σ is the conductivity. The crossover frequency can then be determined by evaluating $\text{Re}[K(\omega)] = 0$.

Field-induced mobility of counterions in the double layer gives rise to the surface conductance K_s whose influence on the overall polarizability can exceed that of the bulk conductivity σ_{bulk} . The total conductivity of a particle can be written as the sum of the bulk conductivity σ_{bulk} through the particle and the surface conductance K_s surrounding the particle²⁷

$$\sigma_p = \sigma_{\text{bulk}} + \frac{2K_s}{R}. \quad (3)$$

For PS spheres, the bulk conductivity ($\sigma_{\text{bulk}} = 10^{-14}$ S/m) is negligible, and the total conductivity is dominated by the surface conductance K_s . It is noted that the particle size effect is included in Eq. (3), and small changes in K_s may result in large changes in the dielectrophoretic spectrum and crossover frequency. Eq. (3) was derived assuming that the EDL is much thinner than the particle diameter, which is in general valid; its effect on the smallest particle will be discussed in detail in Sec. IV D. Here, we assume that EDL is formed instantaneously. The transient effect due to EDL relaxation and the exact EDL distribution on the particle is not considered.

While the simple dipole model has been used extensively in the literature to provide qualitative information on whether pDEP or nDEP is expected in certain particle and medium combinations, and to predict the trend as a function of the electric frequency, its result has limited accuracy in cases where the particle size is important or the electric field is null or highly nonuniform.

B. MST

Another approach for calculating the DEP force and crossover frequency is the MST method. The total force on a dielectric particle with a finite volume located in an electric field can be calculated by integrating the stress tensor τ over the particle volume. Consider electric quasi static system in this work, this tensor is defined as²²

$$\tau = \tilde{\epsilon} \left[\mathbf{E}\mathbf{E} - \frac{1}{2}(\mathbf{E} \cdot \mathbf{E})\mathbf{I} \right], \quad (4)$$

where $\mathbf{E}$ is the electric field, $\mathbf{I}$ is the unit tensor, and the product of two vectors without a dot denotes the dyadic product. Hence, the DEP force on the volume of the particle is obtained by

$$\mathbf{F} = \int_V \mathbf{f} dV = \int_V \nabla \cdot \tau dV = \oint_S \tau \cdot \mathbf{n} dS, \quad (5)$$

where V and S represent the volume and the surface enclosing the volume, respectively, and $\mathbf{n}$ is the unit normal vector of S . The specific volume electromagnetic force is given by

$$\mathbf{f} = \nabla \cdot \tau = \epsilon_p \left[(\nabla \cdot \mathbf{E})\mathbf{E} + (\mathbf{E} \cdot \nabla)\mathbf{E} - \frac{1}{2}\nabla E^2 \right]. \quad (6)$$

These integrations can be performed numerically using finite element method for arbitrary particle shapes and sizes.

III. FINITE ELEMENT SIMULATIONS

In this study, we use simple pin-shaped-electrode configuration as shown in Fig. 1(a) and construct two-dimensional (2D) axisymmetric finite element models by using the commercial package COMSOL Multiphysics as shown in Fig. 1(b). The model is $40 \mu\text{m}$ and $20 \mu\text{m}$ long along the z and r axis, respectively. The gap between the electrodes is $10 \mu\text{m}$, and the spherical particle is offset from the center location by $0.5 \mu\text{m}$ toward the top electrode. The applied voltage is $10 \text{ V}_{\text{p-p}}$. The particle material is polystyrene (PS, relative permittivity is 2.55 and bulk electric conductivity is $\sigma_{\text{bulk}} = 10^{-14}$ S/m) and the suspending medium is de-ionized water (DI water, relative permittivity is 78.5 and the electric conductivity is 2×10^{-4} S/m). The particle surface conductance is 2.56 nS according to the measurement by Honegger *et al.*¹⁹ The mesh near the particle/medium interface is very fine in order to accurately simulate the large gradient in electric potential in this region.

FIG. 1. (a) Schematic diagram of the model. PS particle suspended in DI water and subjected to AC electric field induced by copper electrodes. The distance between the two electrodes is $10\ \mu\text{m}$ and the particle is shifted $0.5\ \mu\text{m}$ away from the center of the gap. (b) The finite element mesh. Mesh quality is important near the boundary of particle and medium for accurate calculation since the MST equations are partial differential equations.

For the cylindrical axisymmetric coordinate, Eq. (6) is reduced to

$$f_z = \epsilon_p \left[\left(\frac{\partial E_z}{\partial z} + \frac{\partial E_r}{\partial r} \right) E_z + \left(\frac{\partial E_z}{\partial r} - \frac{\partial E_r}{\partial z} \right) E_r \right]. \quad (7)$$

The boundary conditions are applied at $r=R$, the surface of the particle. First, the electric potential must be continuous across the particle-medium boundary, $\Phi_p(r=R, \theta) = \Phi_m(r=R, \theta)$. Second, the instantaneous charge conservation condition is satisfied, i.e., $J_{rm} - J_{rp} + \frac{\partial \sigma_f}{\partial t} = 0$, at $r=R$, where $J_{rm} = \sigma_m E_{rm}$ and $J_{rp} = \sigma_p E_{rp}$ are the normal components of the ohmic current outside and inside the particle, respectively, and σ_f is the free electric surface charge as defined by

$$\sigma_f = \epsilon_m E_{rm} - \epsilon_p E_{rp}, \quad \text{at } r=R, \quad (8)$$

where E_{rm} and E_{rp} are the normal components of the electric field intensity outside and inside the particle, respectively.

Effects such as deformation, rotation, and motion of the particles as a function of time are not considered.

IV. RESULTS AND DISCUSSION

A. Effect of the particle size, and the critical diameter

Figure 2 shows the comparison of the normalized crossover frequencies, as a function of the particle diameter, calculated by the MST, the dipole model, and the published experimental results.^{17–19} The particles are all made of polystyrene, but the medium conductivity was measured to be $2 \times 10^{-4}\ \text{S/m}$ and $1 \times 10^{-3}\ \text{S/m}$ in Honegger *et al.*¹⁹ and Wei *et al.*,¹⁸ respectively. In order to facilitate the comparison of the trend of the crossover frequency versus particle diameter, we normalize the curves of all cases with respect to the crossover frequency at $D=0.2\ \mu\text{m}$. At $D=0.2\ \mu\text{m}$ the crossover frequency is $\sim 5\ \text{MHz}$, $4\ \text{MHz}$, and $10\ \text{MHz}$ for Honegger *et al.*,¹⁹ Green and Morgan,¹⁷ and this work, respectively. In general, the MST captures the trend of the experimental results reasonably well. Several factors, such as the electrode design, electric properties of the suspending medium, temperature, and the exact particle shape and size, may influence the measurement of crossover frequencies, which may explain the difference in the crossover frequencies observed by different researchers. Among those, medium

FIG. 2. Comparison of the normalized crossover frequency versus particle diameter between dipole model, MST, and published experiment data by Honegger *et al.*,¹⁹ Green and Morgan,¹⁷ and Wei *et al.*¹⁸ Both dipole model and MST have taken the particle surface conductance into account. “s” indicates the slopes, i.e., the exponents of the power law. The medium conductivity is 1×10^{-3} S/m in Green and Wei, and is 2×10^{-4} S/m for other cases.

conductivity is of great importance and may be responsible for the larger discrepancy between Honegger’s and Wei’s experimental results. The effect of medium conductivity will be discussed in more detail in Sec. IV C.

We observe the existence of two distinct slopes for smaller and larger particles. The slopes, i.e., the exponents of the power law, calculated by the MST are approximately -1.0 and -4.3 for submicron and larger particles ($D > 5 \mu\text{m}$), respectively, which agree remarkably well with the experimental results ($s = -0.7$ – -0.8 for submicron particles, and $s = -4.2$ – -4.3 for larger particles). The dipole model predicts a single slope, $s = -1.0$, which is consistent with the MST and experimental results for submicron particles, but fails to predict accurately for larger particles. A slope of $s = -1$ is expected for a small particle as its crossover frequency is inversely proportional to R due to the dominance of the term involved surface conductance K_s/R when evaluating $\text{Re}[K(\omega)] = 0$.

As the particle diameter increases, the particle conductivity decreases, as shown in Eq. (3), and polarization becomes weaker, resulting in the significant drop of the slope of crossover frequency curve. We define the intersection of the two linear segments (Fig. 2) as the critical diameter D_{cr} , which is $4.6 \mu\text{m}$ in this study.

The influence of the surface conductance on the crossover frequency is shown in Fig. 3. Without considering the surface conductance, both dipole model and MST method incorrectly predict that the crossover frequency does not exist for the case of PS particle suspended in DI water.

In order to better understand the physics of the size effect, we simulated the distribution of the z -component of the electric field intensity, E_z , for particle diameters from $0.2 \mu\text{m}$ to $6 \mu\text{m}$. The ratio of the particle diameter to the electrode gap is fixed at 0.6 for all cases, and the applied voltage is scaled proportional to the diameter so that the electric field intensity is comparable and the effect of electrode gap is not included (Fig. 4). The presence of the particle is included in the calculations, and indeed significantly alters the electric field distribution (Fig. 5). The size effect is considered by using the total conductivity described in Eq. (3). The fields are calculated at the crossover frequency for each diameter (11.17, 4.50, 2.18, 1.02, 0.65, 0.43, 0.27, and 0.14 MHz for $D = 0.2, 0.5, 1, 2, 3, 4, 5$, and $6 \mu\text{m}$, respectively). The region of high E_z is more concentrated near the two electrodes for smaller particles, whereas it is more dispersed along the circumference for larger particles. The induced free electric surface charge density σ_f can be calculated by Eq. (8).

Figure 6 shows the comparison of the distribution of σ_f for different diameters. The particles are located closer to the top electrode (Fig. 1(a)) so the charge density is higher near $\theta = 0$ than $\theta = \pi$. For smaller particles, such as $D < 4 \mu\text{m}$, the charges are more concentrated

FIG. 3. Crossover frequency versus surface conductance K_s for 2- μm diameter PS particle. The crossover frequency increases with K_s as the particle's total conductivity increases, and the particle becomes more polarizable. The crossover frequency does not exist for $K_s = 0$.

near the two ends, and there is almost no charge between 1.0 rad and 2.0 rad, which is a surface charge distribution that can be reasonably described using the physical picture of an effective dipole. The resemblance between the charge arrangements of an effective dipole model and the surface charge distributions evaluated for finite size particles also supports the agreement between the dipole model and the MST simulation results when the particle diameter is small as found in Fig. 2. However, for larger particles, such as $D > 5 \mu\text{m}$, the charge is accumulated even in the region near the equator ($\theta = \pi/2$), which is a result not captured or described when using the dipole approximation. The accumulation of charge near $\pi/2$ becomes more apparent for particles larger than $5 \mu\text{m}$, and hence the crossover frequencies of such particles are

FIG. 4. Schematic of the DEP model for studying the distribution of electric field intensity and free surface charge density.

FIG. 5. Distribution of the z -component of the electric field intensity, E_z , as a function of particle diameter from $0.2 \mu\text{m}$ to $6 \mu\text{m}$. The unit is V/m .

expected to deviate from those predicted by the dipole model. Thus we conclude that there exists a critical diameter, $4 \mu\text{m} < D_{cr} < 5 \mu\text{m}$ in this case, at which the particle begins deviating from the dipolar approximation. This observation is consistent with the results in Fig. 2.

Under the current combination of DI water and particle properties, our numerical results show that a larger particle ($D > 6 \mu\text{m}$) only experiences nDEP and its crossover frequency is either very low or does not exist, which is consistent with the experimental $\text{Re}[K(\omega)]$ spectrum in Honegger *et al.*¹⁹ Biological cells with sizes up to $10\text{--}15 \mu\text{m}$ are routinely handled in lab-on-a-chip systems, and their crossover frequencies may exist due to different combinations of medium/particle properties.

AC electro-osmosis (ACEO) plays a significant role for microparticles and nanoparticles in low conductivity media, especially in DI water, and a low applied frequency range ($1\text{--}30 \text{ kHz}$).³² The crossover frequencies at the critical diameter are approximately 400 kHz for the experimental data from prior works as well as our simulation result (Fig. 2). At such high

FIG. 6. Free surface charge density distribution along the particle circumference.

frequencies, the DEP is more dominant than the ACEO. However, the measured crossover frequency of the largest particle ($5\text{ }\mu\text{m}$) in Honegger's experiment is $\sim 20\text{ kHz}$, and at such a lower frequency the ACEO effect may be present. ACEO is not included in our numerical simulations.

B. Effect of the electrode gap

We studied the effect of electrode gap on the crossover frequency of a PS particle ($D = 1\text{ }\mu\text{m}$) using the model in Fig. 7. The crossover frequency decreases by 20% as the gap decreases from $30\text{ }\mu\text{m}$ to $1.67\text{ }\mu\text{m}$ (Fig. 8) probably due to the high electric field gradient at small gaps. However, such a variation in the crossover frequency is small compared to the results presented in Fig. 2, and the fixed electrode gap (Fig. 1(a)) and the fixed particle to gap ratio setups give the same results.

FIG. 7. Schematic of the DEP model for studying the effect of electrode gap.

FIG. 8. Crossover frequency versus the electrode gap for $D = 1 \mu\text{m}$.

C. Effect of the medium conductivity

The results presented in Fig. 2 are based on an idealized condition in which PS particles are suspended in DI water ($\sigma_m = 2 \times 10^{-4} \text{ S/m}$). Though this condition is convenient for validating the model by comparing the numerical and experimental results with minimum uncertainty, it may be limited in representing the typical lab-on-a-chip applications of DEP in which bioparticles such as cells are manipulated in much more conductive media such as physiological buffers with a σ_m in the range of 0.01 S/m – 1 S/m . Medium conductivity is indeed of significant effect on the crossover frequency. Hughes and Morgan characterized the crossover frequency and surface conductance of unlabeled and carboxylated 216-nm-diameter latex spheres as a function of the medium conductivity.²⁶ They found that the relationship between the crossover frequency and medium conductivity cannot be predicted simply by treating the particle as a solid homogeneous dielectric with constant surface conductance as described in Eq. (3). They modified Eq. (3) by adding a term that is a function of the suspending medium conductivity as follows:²⁶

$$\sigma_p = \sigma_{\text{bulk}} + \frac{2K_s}{R} + \frac{2\sigma_m}{\sqrt{\kappa R}} = \sigma_{\text{bulk}} + \frac{2K_s^*}{R}, \quad (9)$$

where κ^{-1} is the Debye length, and $K_s^* = K_s + \sigma_m \sqrt{R/\kappa}$. An empirical fit to their experimental data of unlabeled particles was obtained in Eq. (9) with $K_s = 1.2 \text{ nS}$ for σ_m ranging from $1 \times 10^{-3} \text{ S/m}$ to $1 \times 10^{-1} \text{ S/m}$. Here, we extend our analysis to consider particle suspending in media of higher conductivities by using Eq. (9). Fig. 9 shows the simulated crossover frequency versus the medium conductivity for $D = 0.2 \mu\text{m}$. We observe that the crossover frequency drops by approximately 40% as σ_m increases from $2 \times 10^{-4} \text{ S/m}$ to $5 \times 10^{-3} \text{ S/m}$.

As mentioned previously, the medium conductivity was measured to be $2 \times 10^{-4} \text{ S/m}$ and $1 \times 10^{-3} \text{ S/m}$ in Honegger *et al.*¹⁹ and Wei *et al.*,¹⁸ respectively, and the crossover frequencies measured by Wei *et al.* were indeed lower than those by Honegger *et al.*.

D. Effect of the EDL thickness

Eq. (3), which modifies the particle conductivity by taking into account the surface conductance, is of great importance and provides a more accurate interpretation of the particles surrounding by an EDL. Eq. (3) was derived by assuming the particle as a sphere and the EDL as a concentric conducting shell. The effective conductivity calculated from the core-shell structure is expressed as²⁷

FIG. 9. Simulated crossover frequency versus the medium conductivity σ_m for $D = 0.2 \mu\text{m}$. The crossover frequency drops by $\sim 40\%$ as σ_m increases from $2 \times 10^{-4} \text{ S/m}$ to $5 \times 10^{-3} \text{ S/m}$. The crossover frequency cannot be obtained from our model for cases with σ_m greater than $5 \times 10^{-3} \text{ S/m}$.

$$\sigma_p = \sigma_{EDL} \times \frac{\left(3 + \frac{3d}{R}\right) \frac{\sigma_{\text{bulk}}}{\sigma_{EDL}} + \frac{6d}{R}}{\frac{3d}{R} \frac{\sigma_{\text{bulk}}}{\sigma_{EDL}} + \left(3 + \frac{6d}{R}\right)}, \quad (10)$$

where d and σ_{EDL} are the EDL thickness and conductivity, respectively. Since $\sigma_{\text{bulk}}/\sigma_{EDL}$ approaches zero in our case, the expression may be reduced to

$$\sigma_p = \sigma_{EDL} \times \frac{2d}{R + 2d} = \frac{2K_s}{R + 2d}, \quad (11)$$

where $K_s = d \sigma_{EDL}$. It is noted that Eq. (11) is further reduced to Eq. (3) for $R \gg d$. The EDL thickness in DI water is $\sim 0.1 \mu\text{m}$, which is comparable to the smallest particle size $0.2 \mu\text{m}$ used here. For those small particles, the effective particle conductivity is smaller than our current values, which reduces the crossover frequency. This may contribute to the difference between our simulation result ($\sim 10 \text{ MHz}$) and the prior experimental data ($\sim 5 \text{ MHz}$).

V. CONCLUSION

We numerically studied the DEP crossover frequency of spherical PS particles, suspended in DI water, with diameters ranging from $0.2 \mu\text{m}$ to $6 \mu\text{m}$ using MST and finite element analysis. This method requires no assumptions on the particle size, shape, homogeneity, and electric field uniformity in contrast to the classic dipole model. Our results show that the MST method captures the trend of the published experimental results, whereas the dipole model is only accurate for $D_{cr} < 4.6 \mu\text{m}$ and significantly overestimates the crossover frequency for particles larger than D_{cr} . The distribution of free surface charge density reveals that the charge begins accumulating on the particle equator for particle diameters larger than D_{cr} , which is a result not captured using the dipole model. The slopes calculated by the MST are approximately -1.0 and -4.3 for submicron and larger particles, respectively, which agree remarkably well with the experiments. The surface conductance due to the field-induced mobility of counterions in the EDL must be included in the total particle conductivity; both dipole model and MST incorrectly predict that the crossover frequency does not exist if the surface conductance is not considered. This method can be readily applied to study nonspherical particles such as ellipsoidal, cylindrical, and biconcave shapes. This work considers the effect of EDL by modifying the particle conductivity and assumes that the EDL is formed instantaneously. Hence, the transient effect

due to EDL relaxation and the exact EDL distribution on the particle is not included in the model, which may introduce certain prediction errors especially for larger particles.

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