Nanofluidics: Systems and Applications

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Abstract—Nanofluidics presents growing and exciting opportunities for conducting fundamental studies for processes and systems that govern molecular-scale operations in science and engineering. In addition, nanofluidics provides a rapidly growing platform for developing new systems and technologies for an ever-growing list of applications. This review presents a summary of the transport phenomena in nanofluidics with a focus on several systems and applications important to problems of public health and welfare. Special emphasis is afforded to the role of the electric double layer and the molecular-scale interactions that occur within confined nanoscale systems.

Index Terms—Microfluidics, nanofluidics, nanomedicine, surfaces, water purification.

NOMENCLATURE

Symbol | Description
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\( J_i \) | Molar flux of \( i \)th species.
\( D_i \) | Diffusion coefficient of \( i \)th species.
\( c_i \) | Molar concentration of \( i \)th species.
\( \nu \) | Velocity of bulk flow.
\( \mu_i \) | Mobility of \( i \)th species.
\( z_i \) | Valence of \( i \)th species.
\( F \) | Faraday’s constant.
\( \Phi \) | Electrical potential.
\( R \) | Universal gas constant.

\( T \) | Absolute temperature.
\( \phi \) | Applied electric potential.
\( \psi \) | Local electric potential.
\( J_A \) | Current density.
\( M \) | Total number of ionic species.
\( K \) | Inverse Debye length.
\( \lambda_D \) | Debye length.
\( a \) | Diameter of nanochannel/nanopore.
\( r_i \) | Radius of \( i \)th ionic species.
\( \eta \) | Viscosity.
\( l \) | Length of nanochannel/nanopore.
\( d \) | Hydraulic diameter.
\( \Delta P \) | Pressure drop.
\( f \) | Body force.
\( \rho \) | Density.
\( E \) | Electric field.
\( I_0 \) | Bessel function of the first kind.
\( \zeta \) | Zeta potential.
\( \dot{Q} \) | Volumetric flow rate.

I. INTRODUCTION

The Digital Age has seen tremendous advances in science and technology that have propelled human imagination to new heights. The vision of developing massively parallel microscale systems for automating operations in the chemical and biological sciences now seems within reach, and the research community wants to move to the next level—building systems to probe, analyze, and use individual molecules to address issues in public health and welfare. Toward these goals, nanofluidics will play an important role as chemistry and biology occur at molecular length scales. The defining feature of nanofluidics is that transport processes occur within systems with critical dimensions on the order of characteristic physical scaling lengths, typically 1–100 nm. In some nanofluidic applications, the key length scales are determined by the shielding properties of aqueous solutions. For example, the Debye length, \( \lambda_D = 1/K \) is ~800 nm for deionized (DI) water (limited by the autoprotolysis of water), and is less than 1 nm for concentrated salt solutions. Therefore, in many systems and applications the effects of shielding and the electric double layer (EDL)
govern scale transport and reactions in nanofluidic structures. Using nanoscale systems and functional nanoscale components in microscale devices provide exciting opportunities for building systems that can probe or manipulate individual molecules. Fig. 1 shows the length scales important for transport at the micro and nanoscales. The length scales are presented from three perspectives: those based on representative devices and applications, the objects that are important for these devices and applications, and lithographic and fabrication techniques that can be used to fabricate the devices.

The advantages of microfluidics, such as limited reagent consumption, high throughput, and development of disposable low-cost devices, are well documented [1], [3]–[6]. In addition, transport physics at the microscale are also well-understood [1], [7]–[10]. However, the story for nanofluidics is quite different. The walls, due to the large surface-area-to-volume ratios (as high as 10⁹ m⁻¹), dominate behavior giving rise to interesting phenomena such as ion perme selectivity, ion enrichment (or depletion), fast injection of small volumes of reagents, rapid mixing, and pH, charge and concentration gradients stable for long periods of time. New experimental tools such as scanning probe microscopy (SPM), confocal microscopy, and single-molecule fluorescence along with the advances in numerical calculations and atomic scale simulation methods provide an increased clarity of understanding of the principles on which new applications can be based [8], [10]–[14].

The purpose of this review is to summarize the transport phenomena in nanofluidics, with special emphasis on the distinctive role of the EDL and the molecular-scale interactions that occur within confined nanoscale systems. The range of devices, how they are fabricated, and the applications differ in many ways from their microscale constructs. Theoretical investigations of nanofluidics, including numerical simulations, have focused on nanoscale transport and several excellent references exist [8], [10], [12], [13], [15]–[21]. Experimental studies, which constitute the majority of this review, may require special fabrication approaches, with a recent review highlighting methods of nanofabrication [22].

II. THEORETICAL BACKGROUND

In contrast to the recent explosion of interest in nanofluidics, the underlying concepts that govern nanofluidics have been known for some time and are discussed next. The theory assumes that the continuum descriptions of fluid transport are valid. Once the continuum assumption breaks down, advanced numerical techniques are generally needed for accurate descriptions of transport phenomena. A discussion of noncontinuum models and some key advances can be found in several excellent references [13], [23]–[30].

Most transport phenomena at the nanoscale are driven either by electrokinetic flow or surface mediated transport. Pressure driven flows, more common for macroscale systems, are rarely employed for nanoscale systems due to the large driving pressures required, as can be seen from the 1/d⁴ dependence of pressure drop, |ΔP|, on channel diameter in the Hagen–Poiseuille equation [31]

\[ |\Delta P| = \frac{128\eta L}{\pi d^4} \dot{Q} \]

where η is the viscosity of the fluid, \( \dot{Q} \) the volumetric flow rate, and L the length of the channel. For instance, with water the pressure drop across a 100 μm long channel 1 nm in diameter for only an attoliter (10⁻²¹ m³) per second incompressible laminar flow would need a pressure drop from (1) greater than 3 GPa, which is impractical for any device. Electrokinetic flows can sustain higher flow rates through nanometer channels without excessive pressures. Thus, electrokinetic flows are a preferred method for driving fluid and ions through nanochannels.

A. Electrokinetic Flow

A charged surface in contact with a liquid forms an EDL. The concept of the EDL has been developed over the last century [16], [17], [32], [33], and several excellent reviews have been presented [18], [19], [34], [35]. A schematic diagram illustrating the classic EDL structure is shown in Fig. 2, showing the positions to which different characteristic surface charge related potentials are referenced within the EDL. Ionic transport...
is classically described by the Nernst–Planck equation and the velocity profiles are often obtained by using the Navier–Stokes equations, which are the equations of motion. The potential distribution in the nanostucture is critical because the dominant transport mechanisms are electrokinetically driven. The ionic and potential distributions are obtained from the Poisson equation. Frequently, a Boltzmann distribution of charged species and potential distributions are obtained from the Poisson equation. For purely electrokinetic flow, the diffusion and electroosmotic terms can be often neglected in comparison to the electro-fluidic interaction in the nanostructure is critical because the dominant transport mechanisms are electrokinetically driven. The mixing is complete within a few hundred microns after injection into the separation channel. The mixing is complete within a few hundred microns after injection into the separation channel. The fluorescence images are collected as a function of time.

Consider a single, cylindrical nanocapillary with the applied potential \( \phi \) and the local potential \( \psi \). Summing fluxes over all the species and accounting for the valence of each species provides the total current density \( J_A \) within a fluidic channel

\[
J_A = F \sum_{i=1}^{i=M} \zeta_i J_i
\]  

which combined with (2) yields

\[
J_A = F \sum_{i=1}^{i=M} \left\{ -D_i \frac{\partial c_i}{\partial x} + c_i \zeta_i \psi - \mu_i \zeta_i^2 c_i \frac{\partial \Phi}{\partial x} \right\}.
\]  

Now, let us look at some important limiting cases as these often arise in the microfluidic and nanofluidic systems being reviewed in this paper. In the case of a concentration gradient being the only driving force, an electrical current arises such that

\[
J_A = F \sum_{i=1}^{i=M} \left\{ -D_i \frac{\partial c_i}{\partial x} \right\}.
\]

This equation can also be obtained from Fick’s laws of diffusion. For purely electrokinetic flow, the diffusion and electroosmotic terms can be often neglected in comparison to the electrophoretic term, yielding

\[
J_A = F \sum_{i=1}^{i=M} \left\{ -\mu_i \zeta_i^2 c_i \frac{\partial \Phi}{\partial x} \right\}.
\]  

Consider a single, cylindrical nanocapillary with the applied potential across it being the only driver for transport of ionic species. In such a case, the velocity of the species in the axial direction is given by

\[
\frac{\partial \vec{v}}{\partial t} + \vec{v} \cdot \nabla \vec{v} = -\frac{1}{\rho} \Delta P + \eta \Delta^2 \vec{v} + \vec{f}.
\]
for which the solution is

\[ v(r) = \frac{\varepsilon}{4\pi\eta} \exp\left\{1 - \frac{I_0(\kappa r)}{I_0(\kappa a)}\right\} \]  

(10)

where \( a \) is the radius of the channel, \( \varepsilon \) is the electrical permittivity, and \( I_0 \) is the hyperbolic Bessel function of the first kind.

Equation (10) is valid for small zeta potentials, \( \zeta \), i.e., typically \( \leq 25 \text{ mV} \). The \( \zeta \) potential is a phenomenological measure of the electric potential at a “slip” plane near a surface, beyond the fixed Stern layer, where the first layer of molecules from the surface are thought to have significant motion. As (10) implies, the transport velocity is proportional to the \( \zeta \) potential and the permittivity of the solution, varies inversely with the viscosity, and is exponentially affected by the shielding length. One caveat for transport in nanochannels is that the concept of the slip or shear plane (Fig. 2) becomes ill-defined when the EDLs from opposed walls within nanopores begin to overlap, and/or structural layering of molecules and charges are a significant portion of the molecules within the channel. In these circumstances of nanoscale transport with interacting or overlapping EDLs, it is more useful to consider the surface charge or the surface potential [38]–[41].

### B. Advanced Numerical Techniques

The theoretical background presented in the previous section assumes the validity of the continuum assumptions, i.e., that the ions are treated as hard spheres and point charges and water is considered as a continuum in the P-B equation. In order to evaluate transport characteristics, the Navier–Stokes and Nernst–Planck equations are used that consider water as a continuous medium with a constant viscosity. Any interactions between the surface and the analytes are considered through the boundary conditions. These assumptions neglect hydrogen bonding between water molecules. Advanced numerical techniques such as molecular dynamic simulations present an attractive method to answer some questions about the atomistic nature of the interactions in confined nanoscale transport. One of the major downsides to advanced numerical techniques is the high computational cost [42]. Therefore, most advanced simulations are limited in scope. Typically, for confined nanoscale transport if the size of the species of interest is about ten times smaller than the characteristic system dimension the continuum approach has been found to be approximately valid. For example, for simple hydrated ions such as \( \text{Na}^+:\text{H}_2\text{O} \) in a nanochannel with critical dimensions on the order of 10 nm or greater, the continuum assumptions hold for dilute solutions. However, contrasting criteria for the breakdown of the continuum methods have also been presented [13], [37], [43]–[46].

Employing advanced numerical techniques to nanofluidics has demonstrated several interesting phenomena spurring greater interest in developing better systems. In addition, the understanding of atomistic behavior governing the transport characteristics has also been enhanced. Effects of finite ion size and hydration on ion distribution and velocity [24], [42], [47], charge inversion and flow reversal [23], density fluctuations [48], role of fluid density on particle transport [49], role of surface charge on transport of ions and water [50], layering and orientation of fluids and ions with changes in fluid density near surfaces due to surface energy interactions [51]–[54], and effect of surface roughness [55] in nanochannels have been investigated. An overall conclusion from these studies is that water and ions organize within a nanochannel that is reminiscent of classical P-B and N-P solutions for potential, but with significant departures in the details that can give rise to unexpected results in transport of species through nanochannels, as will be highlighted in the following discussions.

### III. Applications

#### A. Transport Phenomena

Several interesting phenomena based on the fundamental differences in physics at the nanoscale have been observed. For example, the channel diameter, \( a \), and the inverse Debye length, \( K \), may be combined into the dimensionless parameter \( Ka \). This parameter may be tuned to control the dominant form of nanofluidic transport between electroosmosis (\( Ka \leq 1 \)) and electrophoresis (\( Ka \gg 1 \)) [56]. This tuning can be achieved by changing the EDL thickness relative to the pore diameter either by altering the electrolyte concentration of the solution or by changing the surface charge density by changing the solution pH [56], [57], and functionalizing or applying an electrical bias to the channel walls (as will be discussed later). An implicit assumption in altering the EDL thickness by changing the bulk concentration is that the concentration within the nanopores also changes in concert with the bulk concentration. However, determining the exact concentration within the nanopores can require detailed numerical calculations [13], [35], [58] to account for factors such as ion-ion interactions, changes in hydration level of aqueous species in confined nanoscale spaces, and the molecular nature of water. The role of the EDL in reducing electroosmotic flow (EOF) has also been investigated. It has been shown that once the EDL is greater than approximately 10% of the critical channel dimension, the effect of EDL on EOF can no longer be neglected, and at EDL thickness greater than \( \sim 20 \% \) of the channel dimension EOF is reduced [59]. In a related effect, ion permselectivity occurs due to the excess charge density present on the inner walls of the nanochannels. For example, Au-plated nanocapillary array membranes (NCAMs) reject ions of the same charge (co-ions) and transport ions of the opposite sign (counter-ions). This charge-selective transport is only possible when the EDLs within the nanopores overlap or interact. This permselectivity effect of the surface potential has been demonstrated by biasing the membrane, so that by switching polarity of the applied bias leads to either preferential cation or anion transport [60]. The effect of changing length scales on transport and separation of polyelectrolytes such as DNA has also been investigated [61]. One notable point about separations is that often the need for selectivity and high permeability are in conflict. Few studies have discussed competing roles of selectivity versus permeability across nanofluidic structures.

One of the key factors that affect transport at the nanoscale is the surface charge [39]–[41], [62]. Surface charge can be controlled by either functionalizing the inner surfaces of the nanofluidic systems through physical or chemical modifications or by applying potentials to metal-coated interior pore
walls. Several different surface modification schemes have been developed to affect several parameters for microscale transport [63]–[68]; however, the use of surface coatings to affect transport at the nanoscale is relatively new. Using surface coatings, size-based separations have been demonstrated by controlling the plating time of Au within an NCAM template to tune the internal nanopore diameter. In addition, by using thiol monolayers, chemical transport selectivity can be achieved [69]. Thiol-based surface modifications of Au-NCAMs have been used to generate chemical transport selectivity for different experimental conditions, including selective transport of hydrophilic or hydrophobic species [70], pH-dependent transport [71], separation of proteins by exploiting changes in the isoelectric point of a functionalized surface [72], or direct molecular recognition mediated by thiolated-DNA used to detect base-pair mismatches [73]. Most surface modification schemes to control the surface properties including surface charge utilize either Au-thiol self-assembled monolayers or Si-based monolayers for glass-like systems. Modification of confined surfaces at the nanoscale with synthetic polymers remains a challenge to researchers, although recent advances in derivatization strategies for biological macromolecules have seen both enzymes [74] and antibodies [75] derivatized within nanopores.

The transport phenomena through nanostructures can also be influenced by applying an electrical potential across or to the nanostructures. An early demonstration of controlling ionic permeability through a nanoporous membrane was shown over two decades ago by using an embedded electrode [76]. Using this approach, ion movement through membranes containing fixed ionic sites was shown to depend on the nature and number of charged sites, and ionic resistance of the membrane was electrochemically controlled via the oxidation state on the redox sites within the polymer [76]. Embedded electrode systems are also being developed to probe potential distributions within single nanopores [66].

Rapid progress in microfluidics has led to an increasing demand for developing systems that can handle complex fluidic manipulations in confined nanoscale systems. Integrating nanofluidic components allows for not only further reduction of volumes but also permits manipulation of fluidic samples based on size and charge of analyte species using valves, molecular gates, and fluidic architectures similar to VLSI circuits [9], [66], [77]–[81]. Developing multilayer 3-D integrated microfluidic systems [77] constitutes one viable method to enhance complexity in fluid handling operations, because disparate fluidic manipulations such as preparation, concentration, tagging, separation, or affinity recognition may be accomplished in different planes. Optimal use of these capabilities requires a thorough understanding of nanofluidic transport at its most basic level. To this end, single nanopores, microfabricated in PMMA films can be used as individual interconnects or as platforms to study fundamental nanofluidics [82]. However, several open questions such as the role of surface charge, surface potentials, specific interactions between the walls and the translocating species, mechanisms of transport across nanofluidic structures, and visualization of confined nanoscale flows remain a challenge to the research community.

Three-dimensional systems using nanofluidic interconnects, which provide fluidic communication among microfluidic channels in vertically separated layers, have been of interest for the purpose of fluid handling in mass-transported limited samples [83], [84]. In these micro-nano hybrid systems size-based selectivity for various dextrans has been demonstrated [85], [86]. Size-based selectivity can be understood based on the Stokes–Einstein relation. For a chemical species in bulk solution the diffusion coefficient is governed by

$$D_i = kT/6\pi\eta r_i.$$  \hspace{1cm} (11)

As the size of a nanochannel, nanopore, or nanotube decreases, with increasing molecule size, one expects to observe the physical effects of hindered diffusion. For a molecule of radius $r_i$ diffusing within a nanoscale channel of comparable radius $r_p$, the center of the molecule cannot approach the wall within a distance $r_i$. The extent to which the diffusion coefficient $D_i$ of a molecule in the confined nanoscale space is reduced in contrast to its value in the bulk solution $D_{\text{sol}}$ is given by the Renkin equation

$$D_i/D_{\text{sol}} = 1 - 2.104\lambda + 2.003\lambda^3 - 0.95\lambda^5$$ \hspace{1cm} (12)

where $\lambda = r_i/r_p$. A similar analysis has been presented by Bayley and Martin in their review article on resistive-pulse sensing [87].

The transport characteristics of multilevel 3-D hybrid micro-nano structures are related to the electrical characteristics of the fluidic network. This fluidic network was initially modeled as a simple resistive network in order to explain the polarity reversal in the direction of electrokinetic transport of 200 nm pore diameter NCAMs compared with 15 nm pore diameter NCAMs [80], [85]. In a later study, the capacitance of the NCAM was included [88] in the models. However, further development of equivalent circuit models may be necessary for nanofluidic systems, since it has been shown recently that simple RC circuits, under certain conditions, are not sufficient to explain the transport characteristics at the nanoscale [66]. These 3-D microfluidic structures with nanofluidic interconnects exhibit a variety of interesting and useful electrical characteristics stemming from their nanoscale architecture. For example, they have been shown to maintain large concentration gradients for extended periods of time [89], [90], possess near-diode like behavior with control over rapid injection, mixing, and to preconcentrate attomolar concentration solutions [81], [91], [92] and other mass-limited samples [93], [94].

B. Chemical Analysis

Nanofluidics can add functionality for sample manipulations in analytical chemistry, such as sample injections, separation, purifications, and preconcentration for quantitative and qualitative identification. For example, NCAMs functioning as controllable molecular gates can mediate digital transfer [95] of fluid voxels from one microfluidic channel to another. In addition, NCAMs presenting different pore sizes [96] can transfer analytes with disparate mass characteristics at different rates to achieve intelligent fluidic control. This capability has consequently found use in a miniaturized lead sensor that uses.
DNAzyme [97] as a probe and for preparative post-separation processing for mass limited samples [93]. Control of fluidic transport in nanochannels is crucial to the development of integrated components nano-fluidic-micro-fluidic chemo-electronic devices. In this regard, Karnik et al. reported a metal oxide solution-based system analogous to a metal oxide semiconductor field effect transistor [39] that achieves flow switching via active control of the surface potential in the interior of the nanochannel, thereby demonstrating the ability to control transport of charged species in a nanostructure that could potentially lead to fluiditronics.

Another area in which nanofluidics can have a profound impact on chemical analysis is sample preparation. For example, an analyte may need to be isolated from a complex matrix and made available for analysis in a suitable chemical environment. Iannaceone et al. [98] demonstrated that NCAMs can be used to enhance the intensity of an insulin peak introduced into a mass spectrometer from a microfluidic channel using electrospray ionization; this occurs by reducing the salt adducts inherent in the sample preparation process. Additionally, various receptors can be incorporated into NCAMs. This strategy could be beneficial for sample preconcentration and purification of biomolecules in microfluidic devices. Both antibodies [75] and DNA probes [99] have been successfully incorporated in NCAMs for applications in protein purification and preconcentration. Preconcentration factors of up to 300 have been achieved for single polarity species [100] in NCAMs and up to $10^6$ for proteins and peptides [101] in simple nanofluidic structures.

Beyond these demonstrated applications of micro-nanofluidic architectures have enormous potential for novel chemical separations by reducing required sample amount and improving resolution. This issue has been addressed both theoretically [102], [103] and experimentally, and is mainly driven by interest in DNA separation. Periodically constricted nanochannels [104] have shown successful DNA separations and close packed beads with nanometer spacing [105] have been utilized to accomplish both DNA and protein separations.

C. Nanomedicine

One of the reasons the scientific community is excited about advances in nanofluidics and nanotechnology centers on the possibility of manipulating processes, especially in biological systems, at the molecular level. This possibility has led to rapid development of new materials, methods, and systems for applications in biomedicine [106]–[110]. A recent review explains the concept of nanomedicine and some of the early advances in applying the principles of nanotechnology to medicine [111]. Nanomaterials, such as silica or gold nanoparticles, quantum dots, functionalyzed micro and nanocantilevers, and artificial nanoropes are being increasingly used for detection of DNA [2], [112]–[115], RNA [116]–[118], and proteins [119]–[122]. In some cases, the asymmetry within artificial nanostructures is being exploited to mimic the behavior of natural systems, such as ion-channels [123]–[125]. Multiple fabrication techniques, for example, e-beam lithography, focused ion-beam milling, and nanoimprint lithography have been used to construct nanostructures for trapping and transporting DNA and focusing of proteins using dielectrophoresis [126]. Using well-defined nanochannels from 30–500 nm DNA and proteins have been separated using steric effects that exploit the principle of entropic minimization in the transport of biomacromolecules within confined geometries [123]. Nanostructures and single nanoropes have been used to reduce the entropic contribution to the total free energy of DNA, thereby modulating entry into a separation channel according to the oligonucleotide length [127], [128].

Silicon has been extensively used as a material for building systems for applications in biochemical systems at the nanoscale due to its versatility in microfabrication [129]. However, chemical modification can impart novel properties of the solid-liquid interface that can be usefully exploited. Many materials and methods are currently being researched for drug-delivery applications [130] including dendrimers, such as poly(amide amine) or PAMAM [131]. Dendrimers have recently been attached to confined surfaces within microfluidic devices and show a possible method for attachment of PAMAM dendrimers to nanofluidic devices [68]. Development of systems relying on bacteria as molecular motors for pumping small volumes of liquids has also been investigated [132].

D. Water Purification

Recent reports have highlighted the growing challenge of providing clean, safe water for a variety of applications including potable water, energy generation, and agricultural use [133]–[135]. These problems are central to the general public health and welfare and, therefore, have led to a resurgence of interest in researching new materials, systems, and methods for water purification and detection of various chemical and biological species in water supplies [90], [136]–[139]. Membranes, consisting of nanoropes or nanocapillaries ranging from 1 nm to 1 $\mu$m, have historically played an important role in applications related to water purification [10], [140]–[142]. They have been studied extensively for their role in aqueous systems including studies on the importance of chemical composition, surface charge and morphology [62], [143]–[145], fouling [146], [147], and desalination and filtration [90], [135], [139], [148], [149]. Structures based on the purposeful manipulation of nanofluidics present excellent opportunities for growth in the area of detection and sensing for water purification as illustrated by the use of an integrated microsystem for injection analysis of ammonia in surface waters for environmental applications [150]. Detection of heavy metal ions has found a new pathway in the development of DNA-based sensors for lead and uranium ions [151]–[154]. Nanoparticles have been used for removal of trace organic contaminants such as trichloroethylene and chloroform [155] and mesoporous materials with pores in the sub-10 nm range have been used for removal of polyelectrolyte species such as humic acids from water [156].

IV. Summary

Many applications and scientific advances in nanotechnology have created an intense debate both in the scientific and socio-political forums about the implications and future directions of the multitude of new technologies. Nanofluidics is a rapidly evolving discipline with wide and varied applications. A few of these applications with direct relevance to problems of public health and welfare such as nanomedicine
and water purification have been discussed in this paper. The role of the EDL in determining transport in confined nanoscale transport has been conclusively demonstrated. The field of nanofluidics is highly interdisciplinary taking inspiration from nature and biology with applications in chemistry, materials science, physics, environmental, and engineering systems. It is hoped that this paper will assist researchers from diverse fields by providing a collection of varied references.

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