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Towards the design of an intensified coagulator

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A B S T R A C T

This study compares the hydrodynamics in three millimeter-scale continuous reactor geometries that can be easily used in laboratories and industries – a straight tube, a coiled tube and a Dean-Hex reactor – via numerical simulations and analyses the data in a way that is specifically relevant to coagulation processes, thereby offering insights for engineers to develop new coagulation reactors. A numerical approach based on Lagrangian particle tracking is presented to better understand the impact of the geometry and flow on properties that influence coagulation. The results show that the Dean-Hex meandering geometry provides narrower residence time and shear rate distributions, as well as higher mean average shear rates and Camp number distribution than the other geometries. This is attributed to the generation of transverse flows and radial mixing in the Dean-Hex reactor and suggests that a faster and more homogenous coagulation can be expected.

1. Introduction

Coagulation is a key process in several industries. When considering wastewater treatment the coagulation/flocculation process is used to remove the undesired organic matter from water, and is essential to the success of the wastewater cleaning process [1]. In the polymer industries, latex coagulation typically follows emulsion polymerization, and aims at controlling both particle size distribution (PSD) and the shape of the aggregates. As the quality of the resulting polymer products directly depends on these properties, coagulation must be controlled well.

Two main collision mechanisms can lead to coagulation. When the colloidal particles are small enough (i.e. with diameters less than several hundred nanometers) they will undergo Brownian motion. Temperature thus has a significant impact on coagulation; the term perikinetic coagulation is then used. Above particle diameters of several hundreds of nanometers, the hydrodynamics have an impact on coagulation. When particles/aggregates are large enough so that coagulation is driven only by shear rates, the terms orthokinetic coagulation or shear coagulation are used.

In most industries, latex coagulation/flocculation is performed in agitated vessels [2]. As a consequence, coagulation experiments in these devices can easily be found in the literature [3–9]. However, coagulation in stirred tanks often results in a broad PSD, heterogeneous shapes and other issues, such as fouling, due to poor control of local hydrodynamics and uneven mixing.

In order to overcome these problems, new processes have to be studied and developed. One of the most interesting alternatives studied so far is the use of tubular reactors, whether in laminar [10–14] or turbulent [1,15] flow conditions. When the characteristic length of the reactor is in the millimeter range, coagulation typically occurs in laminar flow conditions. Despite a broad distribution of shear rates due to the parabolic flow profile, it is possible to obtain almost spherical in shape aggregates due to the promotion of ballistic collisions between aggregates presenting size disparities [16]. Another interesting alternative, so far not considered for latex coagulation, is the use of millimeter-scale intensified reactors, which allow transverse mixing throughout the reactor, and thus reduce axial dispersion generated by the laminar velocity profile [17]. Among these reactors, those with meandering channels, sometimes referred to as "Dean-Hex" or zigzag reactors, are of particular interest here due to the relative simplicity of their conception. By promoting the generation of the so-called Dean vortices, these geometries enhance mixing during within the flow [18], and thus offer a way to overcome broad residence time distributions (RTD) encountered in straight tubes in laminar flow.

The aim of this paper is thus to investigate numerically the hydrodynamics inside three millimeter-scale geometries that can be easily used in laboratories and industries – a straight tube, a coiled tube and a Dean-Hex reactor – and extract relevant data with regards to coagulation phenomena, offering insights for engineers to develop new

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Nomenclature

D_{ax}	Axial dispersion coefficient ($m^2\,s^{-1}$)
De	Dean number (−)
d_h	Hydraulic diameter (m)
$E(t)$	Residence time distribution (s^{-1})
$\boldsymbol{F}_{drag}$	Drag force (N)
G	Average shear rate inside the reactor (s^{-1})
$G_{av,part}$	Average shear rate experienced per particle (s^{-1})
$G_{max,part}$	Maximum shear rate experienced per particle (s^{-1})
H	Depth of the square channels (m)
L	Length of the reactor (m)
m_p	Mass of particle tracer (kg)
N	Concentration of particles (m^{-3})
N_{Camp}	Camp number (−)
N_{SL}	Number of sub-layers in the inflation layer (−)
N_w	Total number of weighted particles (−)
Pe	Péclet number (−)
Q	Flow rate ($m^3\,s^{-1}$)
r	Radial coordinate (m)
Re	Reynolds number (−)
R_{tube}	Radius of the tube (m)
R_{curv}	Radius of curvature (m)
S	Cross-sectional area (m^2)
t	Residence time (s)
T	Fluid temperature (K)
t_m	Mean residence time of the particles (s)
$\boldsymbol{u}$	Velocity vector ($m\,s^{-1}$)
u_{inlet}	Inlet velocity ($m\,s^{-1}$)
u_{av}	Average velocity ($m\,s^{-1}$)
u_{nd}	Non-dimensional velocity (−)
V	Volume of the reactors (m^3)
$\mathbf{X}$	Particle location vector (m)
x	Cartesian coordinate (m)
y	Cartesian coordinate (m)

Greek letters

α	Hydrodynamic collision efficiency (−)
δ_t	Characteristic size of the cell volume in the tetrahedral mesh (m)
μ	Fluid dynamic viscosity (Pa s)
φ	Volume fraction of the particles (−)
ρ	Fluid density ($kg\,m^{-3}$)
σ^2	Variance of the distribution (s^2)
τ	Fluid residence time (s)

coagulation processes.

2. Theory

2.1. Shear coagulation theory

In the early twentieth century, Smoluchowski [19] theorized shear coagulation in analogy with chemical kinetics. By assuming that:

- the flow is laminar,
- all collisions are efficient (no repulsions between particles due to electrostatic or hydrodynamic effects),
- the particles follow streamlines,
- the particles can be considered as spheres,
- all the particles have equal radii, r,

he derived an equation describing the evolution of the total number of particles per unit volume (N) with the average shear rate G. The decrease of N is given by Eq. (1):

$$-\frac{dN}{dt} = \frac{16}{3}GN^2r^3 \tag{1}$$

Later, several authors took into account the hydrodynamic interactions occurring when particles are close by correcting the coagulation kinetic equation with a purely hydrodynamic efficiency, α. Among the most commonly used efficiencies, the theoretical expressions proposed by Higashitani et al. [20], Han and Lawler [21] or the semi-empirical formula recently introduced by Selomulya et al. [22] can be mentioned. By inserting the efficiency α into the balance presented earlier, one obtains Eq. (2):

$$-\frac{dN}{dt} = \alpha\frac{16}{3}GN^2r^3 \tag{2}$$

The volume fraction of the particles $\left(\varphi = \frac{4}{3}\pi r^3 N\right)$can be introduced in Eq. (2) to obtain Eq. (3):

$$-\frac{dN}{dt} = \alpha\frac{4}{\pi}G\varphi N \tag{3}$$

By integrating Eq. (3), an expression giving the decrease of N with time is obtained [11] (Eq. (4)). It assumes that the hydrodynamic efficiency, the volume fraction of particles and the average shear rate are constant along the coagulator.

$$\frac{N}{N_0} = \exp\left(\frac{-4\alpha\varphi Gt}{\pi}\right) \tag{4}$$

Interestingly, the coagulation rate given by Eq. (4) is directly related to the product Gt, sometimes called the Camp number. Despite the limitations of this expression, which is rigorously valid only during the early stages of coagulation when the primary particles can be considered as spheres of equal radii, it clearly shows that the higher the Camp number, the higher the coagulation rate.

2.2. Hydrodynamic dimensionless quantities

When the geometry in which the fluid circulates displays curvatures, centrifugal forces can have an influence on the flow behavior. Dean [23] was the first to numerically solve the problem in curved pipes. He demonstrated the presence of recirculation loops under laminar flow conditions when centrifugal forces become significant. He thus proposed the use of a dimensionless quantity – the so-called Dean number (De) – to determine whether centrifugal forces can have a significant impact on the flow. The common expression used for calculating De is defined in Eq. (5):

$$De = Re\sqrt{\frac{d_h}{R_{curv}}} \tag{5}$$

where R_{curv} is the radius of curvature at the center of the bend and Re the Reynolds number, defined using the hydrodynamic diameter of the considered pipe d_h. In laminar flow, an increase in De means a shift of

Table 1
Dean-Hex parameters.

Parameter	Value (mm)
L_1	10.36
L_2	10
L_3	16.33
A_1	2.42
A_2	4.71
A_3	9.42