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# Polyelectrolyte-mediated interparticle forces in aqueous suspensions: Molecular structure and surface forces relationship<sup>☆</sup>

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## ABSTRACT

Surface forces in suspensions must be controlled if optimum processing is to be achieved as they govern their processing behaviour. Surface chemistry tools such as pH, ionic strength and adsorbed additives are often used to effect and control these forces. Adsorbed polyelectrolytes give rise to a range of non-DLVO surface forces in colloidal dispersions depending upon their molecular and structural properties. These forces are steric, bridging, patch charge, entropic or “hydrophobic” and others. In this paper these polymer-mediated forces will be discussed in relation to the polymer molecular, structural and physical properties. Scientists, process engineers, metallurgists and related practitioners wishing to optimize the handling/processing of colloidal suspension in the processes employed by their industries may find the experimental and theoretical knowledge, and fundamental principles established in this review paper very beneficial.

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## 1. Introduction

Surface forces govern the processing behaviour of colloidal suspension and emulsion in every process such as slip, gel and tape casting, solid–liquid separation, mixing, polishing, pumping, flocculation and sedimentation (or thickening), flotation, pipeline transportation, coating and spreading, sol–gel, thin-film, polyelectrolyte layering and many others. These forces are subjects of intense fundamental and applied research as the knowledge gained are needed (i) to build theoretical understandings and (ii) to optimize processing and solve processing problems of suspensions and emulsions. These forces are also exploited to control the texture, feel and flow behaviour of commercial colloidal suspension products such as those found in cosmetics, personal health care, food and beverages. The nature and strength of these surface forces are determined by the surface chemistry of the system. The ability to easily regulate suspension behaviour for

optimal processing via surface forces through the appropriate deployment of the various surface chemistry tools is therefore highly desirable. However the knowledge and skills required in deploying these tools in a manner that precisely achieves the desired objectives are presently not well developed. Significant cost saving and greater efficiency in the suspension processing industries are therefore still possible and achievable. Industrial sectors that process suspensions and emulsions are very broad and diverse. They include ceramics, food and agriculture, minerals, chemicals, cosmetic, pharmaceutical, coating and paint, wastewater, electronic and thin film, and many others.

Surface forces may be classified as DLVO and non-DLVO forces. The DLVO forces are van der Waals (vdW) attractive and electrostatic or double layer repulsive (EDL) forces. Most non-DLVO forces arise from adsorbed additives and they are steric, bridging, patch charge and entropic or “hydrophobic” forces. These forces have been studied quite extensively with

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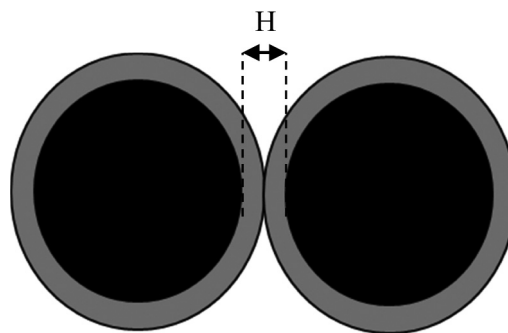
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surface force instruments such as the Surface Force Apparatus (SFA) and Atomic Force Microscopy (AFM) and its many variations, and have been the subject of several reviews in recent years (Borkovec et al., 2012; Claesson et al., 2005; Horn, 1990; Liang et al., 2007; Szilagyi et al., 2014). The force–distance profiles for interaction between surfaces or particles were determined with these force instruments. The study of each force in isolation is often not possible. Van der Waals force is ever present and steric force cannot be avoided in the presence of an adsorbed layer. Electric double layer (EDL) forces could also be present if the force–distance measurements were not conducted at the point of zero net surface charge.

In concentrated suspensions, surface forces cannot be measured or determined directly. For some additives, the predominant surface force operating in a suspension is known from prior knowledge gained from SFA or AFM studies. For other additives, these forces are inferred from their effects on the suspension rheological properties such as viscosity, storage modulus and yield stress. The static yield stress measured directly with the vane technique (Nguyen and Boger, 1983), is an ideal parameter for studying the effect of both DLVO and non-DLVO forces as it measures the strength of the flocculated network structure formed by the action of these forces interacting between particles in the network (Leong et al., 1993a,b, 1995a; Nguyen and Boger, 1983). Indirectly, it measures the strength of the interparticle forces. Electrostatic or double layer repulsive force can be eliminated at the pH of zero zeta potential. At this point, the yield stress is at maximum as only the van der Waals force is in play. In the presence of adsorbed additives, the maximum yield stress is often located at the pH of zero zeta potential. At this pH, non-DLVO forces arising from adsorbed additives are also present. Analyzing the yield stress at zero zeta potential will help to deduce the predominant surface force in operation and quantify the effect of these forces. The predominant non-DLVO force in operation can change depending upon the surface coverage and the pH of the zero zeta potential state for a given additive. For example, patch charge attraction is not important at high surface coverage (Leong et al., 1995a,b). The surface forces that have been deduced and their effects quantified with the vane yield stress technique are steric, patch charge, bridging, depletion and hydrophobic (Leong, 1996, 1999; Leong et al., 1995a,b, 2003; Ong et al., 2009). This study will provide a summary of the author's many works relating surface forces in suspension with the molecular structure, architecture and chemical properties of the polyelectrolytes. The increase or decrease of the maximum yield stress of the suspensions at rest was used as a measure of the action of the various surface forces. Depletion forces involving adsorbed and unadsorbed polyelectrolytes in suspension (Leong, 1996) will not be covered here. This has been covered in a previous review by Johnson et al. (2000). Another reason as to why this review was restricted to author's work only is that the analysis of the surface force in action was conducted at the pH of zero zeta potential or point of zero net charge where the EDL forces are eliminated, thus providing greater clarity of the quantitative knowledge of the action and influence of the various surface forces in concentrated suspensions.

In many processes such as mixing, pumping, clay modelling, ceramic molding and slip casting, the consistency of the suspension is very important. When the consistency of the suspension is either too thin or too thick, the performance of the processes is often affected producing off-specification products. This is particularly true in wet ceramic processing



**Fig. 1 – A cartoon of interaction between two (dark) spherical particles each with an adsorbed layer. The grey denotes the adsorbed layer. The interacting particles are separated by a distance  $H$  as a result of steric interaction between the adsorbed layers.**

and clay modelling (Lange, 1989; Lewis, 2000; Franks and Gan, 2007). Many processing equipment are designed based on the consistency of the material being processed. A mixer designed for fluid-like suspension processing is not suitable for paste processing. Yield stress, both static and dynamic, is a very good indicator of the consistency of the suspension. A high yield stress, typically  $>500$  Pa, denotes a very viscous suspension with a paste-like consistency. The values of the static and dynamic yield stress are usually identical for weakly thixotropic or non-thixotropic suspensions. This is the case with brown coal and red mud suspensions where the vane yield stress is identical to the dynamic yield stress extracted from constitutive equations such as the Bingham and Herschel–Bulkley models, used to fit the flow data (Leong et al., 1987; Nguyen and Boger, 1985). In this paper, vane yield stress results are presented for the analysis of the surface force in operation.

## 2. Surface forces

### 2.1. Repulsive forces

#### 2.1.1. Steric forces

The physical layer formed by adsorbed polyelectrolyte molecules on a particle acts as a steric barrier preventing penetration by another layer on an interacting particle during compressive contact. This pair of interacting particles will experience a very strong repulsive force when the electron clouds of the adsorbed layers overlap during the compression. This Born repulsive force is therefore very short range, on the order of Angstrom scale. These adsorbed layers are effectively keeping the interacting particles further apart as shown in Fig. 1, thereby weakening the strength of the van der Waals (vdW) attraction (Leong et al., 1993a,b). In concentrated and flocculated suspensions, this steric layer will have the effect of reducing the yield stress.

There are two types of steric layers: soft shell and hard wall (Qiu et al., 2007). A soft shell steric layer is formed by polymer molecules with one end adsorbed or grafted on the particle surface and these attached molecules then extended out from the surface forming a brush (Milner, 1991). This layer is compressible and allowed the interpenetration of extended polymer chains from an interacting particle or surface at close contact. Pure soft-shell steric interaction in aqueous suspension is unlikely to be possible. The polymers used must be

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