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Modeling of gel layer transport during ultrafiltration of fruit juice with non-Newtonian fluid rheology

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ABSTRACT

The rheology of fruit juice mixtures generally follows non-Newtonian behavior of power law form. The clarification of fruit juices by membrane separation illustrates an example of enhancing the shelf life of a real fruit juice by removing degradable components. However, the presence of high molecular weight proteins, pectins, polysaccharides, fibers, etc., tends to form gel over the membrane during filtration causing fouling and affecting its performance. The proposed model developed from the first principle boundary layer analysis, describes the physical mass transport phenomena and quantifies the various extents of fouling using different membrane materials and operating conditions. The model results are useful in understanding the complex solute–membrane interplay in fouling and can predict the effect of gel layer thickness on the process throughput.

In this work, the model results were validated experimentally in clarification of blood orange juice in batch mode using two polysulphone (PS) membranes and polyacrylonitrile (PAN) membrane in hollow fiber configuration, with different molecular-weight-cut-off (MWCO). The results clearly indicated that PS membranes are more prone to fouling at higher pressures compared to PAN membrane. An increase in the feed flow rate had a significant effect in reducing the growth of gel layer mainly in PS membranes.

1. Introduction

Cross flow membrane filtration processes are widely used in food processing industries (Girard and Fukumoto, 2000), biotechnology (Cheryan, 1998), the pharmaceutical sector (Wang and Chung, 2006), clarification and concentration of fruit juice (Rai et al., 2010; Mondal et al., 2011a; Thomas et al., 1987; Mohammad et al., 2012). In most of the filtration processes, batch mode is often used, since the permeate is the preferred product. For efficient design of large scale systems, prediction and detailed understanding of the mass transfer phenomena with coupled fluid flow is important. The relevant flow configuration and flow regimes are significant in modeling the process performance. The mass transfer coefficient is generally calculated from the Sherwood number correlations using Leveque relation, derived from heat and mass transfer analogies. However, these correlations fail to take into account the effects of non-Newtonian rheology of the fluid and changes due to developing mass transfer boundary layer on the hydrodynamics of the flow regime and consequently on the mass transfer coefficient. The available mass transfer correlations for membrane separation processes along with their shortcomings have been already reviewed in detail (van Den Berg et al., 1989; Gekas and Hallstrom, 1987).

Generally, citrus fruit juice containing pectins are non-Newtonian indicating that they do not exhibit a direct proportionality between shear stress and shear rate. The Ostwald–de Waele model (also known

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as the power-law model) is used to describe this rheological behavior for most fruit juice solutions (Rao et al., 1984),

$$\tau = K\dot{\gamma}^n \tag{1}$$

where τ is the shear stress, $\dot{\gamma}$ is the shear rate, K is the consistency index and n is the flow behavior index. There are several reports of carrot (Vandresen et al., 2009), blueberry (Nindo et al., 2005), mango (Dak et al., 2007), pummel (Chin et al., 2009), pineapple (Dak et al., 2008), pomegranate (Yildiz et al., 2009), sugarcane juice (Filho et al., 2011), passion fruit (Jiraratananon and Chanachai, 1996), mandarin (Falguera et al., 2010), date (Gabsi et al., 2013), guava (Sánchez et al., 2009) and blood orange juice (Mizrahi and Berk, 1972), that confirm the power law model is the most appropriate in all cases. The knowledge of the fluid rheology is important for design and optimization of unit operations.

A significant phenomenon leading to the decline in flux is concentration polarization (Porter, 2005; Sablani et al., 2001). A simple description of concentration polarization is obtained from a stagnant film model, used by Sherwood et al. (1965) to analyze reverse osmosis. Many researchers (Opong and Zydney, 1991; Zydney, 1997; Johnston and Deen, 1999) have used stagnant film model that considers a thin layer of solute adhered to the membrane surface, leading to one-dimensional problem in which the solute concentration depends only on distance from the membrane surface. To overcome this problem, a detailed numerical solution of the governing momentum and solute mass balance equation with pertinent boundary conditions may be used (Kleinstreuer and Paller, 1983; Bouchard et al., 1994; De and Bhattacharya, 1997). However, these studies do not incorporate the effects of fluid rheology and involves inherent complexities and rigorous computational requirements, rendering it unattractive and not useful for fruit juice clarification applications.

Detailed studies related to two-dimensional concentration fields for laminar cross flow ultrafiltration in tubes or parallel-plate channels have been reported in literature (Shen and Probstein, 1977; Gill et al., 1988; Denisov, 1999; Bhattacharjee et al., 1999; Madireddi et al., 1999), spiral-wound membrane modules (Kozinski and Lightfoot, 1971). Field and Aimar (1993) have modified Leveque's correlation for laminar flow in rectangular channel by using a viscosity correction factor. However, the effects of suction were not considered in their study which has been incorporated later by De and Bhattacharya (1997). Sherwood number relationship incorporating the effects of suction (in presence of a membrane) for laminar flow in rectangular, radial, and tubular geometries, have been formulated starting from first principles (De and Bhattacharya, 1997). However, that study includes the osmotic pressure controlled filtration only and the effect of developing mass transfer boundary layer.

It has been shown that due to concentration polarization, the variation of the physical properties with concentration is significant in the performance of the ultrafiltration and subsequent development of the boundary layer, specially a gel layer controlling case (Gill et al., 1988; De and Bhattacharya, 1999; Bowen and Williams, 2001). During filtration of high molecular weight proteins, polymer, paint, clay, etc., a highly viscous solid-like layer is formed over the membrane surface above its solubility limit (commonly known as gel concentration) and it obeys the classical gel filtration theory. The primitive gel layer model is derived from conventional film theory (Blatt et al., 1970), considering a uniform mass transfer boundary layer thickness instead of developing boundary layer which is more fundamentally correct. Moreover, the viscosity of the solution is a strong function of the solute concentration and it varies significantly within the mass transfer boundary layer, as the increase in solute concentration from bulk to gel layer concentration is not considered. Gel layer concentration is quite often three to seven times of bulk concentration. This variation of viscosity as a function of concentration was not included in the film model. Probstein et al. (1978) developed a two-dimensional model, developing mass transfer boundary layer under laminar flow condition in a rectangular channel, overcoming one of the limitations of the constant thickness boundary layer. Clarification of fruit juice by ultrafiltration has been found to be gel controlling in many occasions due to presence of protein, cell debris, cellulose, etc. (Rai et al., 2010; Sarkar et al., 2008; Mondal and Chhaya De, 2012a).

The present theoretical analysis is focused on developing mass transfer analysis of ultrafiltration of fruit juice considering non-Newtonian power law rheology in a hollow fiber module. Solution of the convective-diffusive species transport equation is performed under the framework of the boundary layer analysis. The model includes the developing mass transfer boundary layer over the gel layer, effects of concentration dependence on viscosity and non-Newtonian fluid rheology. The present model quantifies the flux decline as well as the volume reduction factor (VRF) during batch mode of operation from the first principles by solving the overall material balance, overall solute balance and solute balance within the mass transfer boundary layer. A numerical solution of these balance equations leads to the flux decline and VRF profile. Therefore, the present model is a comprehensive one including various fundamental aspects of transport phenomena involved. The extensive analytical treatment makes the model easy to estimate the throughput in either of the operation modes by simple computational techniques. Some of the key and distinguishable features of the present modeling approach are summarized here:

1. The present model is completely predictive and does not require any knowledge of the experimental flux values to model the system, which is unlike the case with other fouling models. The steady state flux value is often provided as an input to other existing models.
2. The values of the physical properties and process constants are completely realistic and are not fitting parameters which are obtained by regression in other black box models.
3. The present model captures the essential underlying physics of the species transport in the concentration boundary layer and gel formation, which is not described well by many semi-empirical fouling models.
4. Most fouling models are based on the Hermia's (1982) and Field et al. (1995) description of the constant pressure filtration, which does not take into account of the increase in feed concentration during batch mode of filtration and the effects of the channel narrowing due to gel formation.
5. The results of the model can be directly interpreted on the effect of the process parameters (transmembrane pressure drop, flow rate, etc.) and thus useful in understanding the interplay of the operating conditions and controlling such systems in practice.
6. Finally, many existing fouling models are not suitable in incorporating the fluid rheological effects on the overall mass transport phenomena.

Gel layer thickness is very difficult to measure experimentally during filtration. There have been very few literature studies on the in situ measurements of the gel layer during unstirred batch membrane filtration (Chen et al., 2004; Guell et al., 2009). In the present study, the experiments are carried out in crossflow mode in hollow fiber configuration. To the best of our knowledge, there is no analytical or instrumentation facility available till date to measure directly the thickness of gel layer inside the hollow fiber during filtration. There have been few literature reports in the past regarding estimation of the gel layer thickness. The classical gel layer theory used to predict the gel thickness was based on a single mass balance at the membrane interface and require the knowledge of the gel layer concentration and permeate flux together (Blatt et al., 1970; Belfort et al., 1994; Porter, 1972). Thus, it is impossible to predict the permeate flux or the gel concentration from this theory, and are often used as input from the experiments to calculate the gel thickness. Also, this is based on the assumption of constant concentration boundary layer developed over the membrane surface. There has been attempt to include the shear-induced-diffusion of the particulate suspension (Davis, 1992) and solving the transient 1D species transport equation (Karode, 2001) as an improvement to the existing models. One of the prevalent theories in this regard is the application of the Happel cell model (1965) based on particle flux conservation (Song and Elimelech, 1995). The gel is considered to be a concentrated particulate suspension of