EVAPORATIVE CRYSTALLIZATION OF ALPHA-LACTOSE MONOHYDRATE

A thesis presented in partial fulfilment of the requirements
for the degree of

Doctor of Philosophy

In

Chemical Engineering

at Massey University, Manawatu
New Zealand.

Shailesh Ghanashyam Agrawal

2012

Dedicated to

Rasikbihari….

Abstract

Evaporative crystallization has been used by Fonterra Cooperative Group (New Zealand) for producing lactose. It represents an important step during lactose manufacturing where control over crystal size can be obtained, a critical parameter governing the yield and end use. The art of operating these crystallizers has been developed by observation and not from scientific principles. This project was undertaken to understand the mechanisms controlling the crystal size in evaporative crystallizers.

A review of the existing literature showed that secondary nucleation is the major source of nuclei in industrial crystallizers. Based on the review, attrition, contact and fluid shear induced nucleation were identified as the probable secondary nucleation mechanisms in the studied system. Experimental investigation on each of the three mechanisms was carried out separately on a laboratory scale.

It was found that the crystal size had the most significant effect on attrition, followed by impeller speed, which together implies that the crystal collision energy intensity is the dominant factor producing new fragments. Contact nucleation was also found to be controlled by crystal-impeller collisions. It was found that at the studied supersaturation there exists a minimum kinetic energy of contact below which secondary nucleation would not occur. This threshold value was used as the basis to assess the contribution of various mechanisms at the industrial scale. Shear nucleation was found to be independent of shear above 5000 s^{-1}.

A mathematical model describing the operation of the industrial crystallizer was formulated. Sensitivity analysis was conducted by simulating the model for a range of operational and kinetic parameter values. It was found that the crystal size is affected most by secondary nucleation. The volume weighted mean size approximately halved with a 5.5 times increase in the secondary nucleation rate.

The model was refined to accommodate size dependent growth rate and growth rate dispersion. The kinetic parameters were fitted to match the measured size distribution from the industrial crystallizer. A range of simulations were conducted for various theoretical and empirical models and compared to that of plant measurements. Based on the results it was proposed that the majority of secondary nucleation is expected to occur in the pump and the boiling zone.

Acknowledgement

It's not easy to move to an alien country to do a PhD. So my first thank to New Zealand and its people for making me feel comfortable and for being my home for close to four years. I will then like to thank my main supervisor, Prof. Tony Paterson for giving me the opportunity to work under him. Thanks Tony for showing constant faith in my abilities and being there whenever I needed advice, academic or personal. This PhD would not be possible without your guidance and encouragement.

Big thanks to my co-supervisors: Dr. Jeremy McLeod, Prof. Jim Jones and Prof. John Bronlund. Thanks Jeremy for proposing this PhD and getting me started into Fonterra during your last few days with them. Thanks for attending my weekly meeting from USA and taking a keen interest in the project. Jim, I learnt a lot about technical writing from you. I would be happy if I could become half as good as you. John, thanks for helping me with my model. That one small session with you on Matlab got me going!

My sincere thanks to my mentors at Fonterra: Tony Styles and Raymond Joe. It is not easy to carry out experimental studies in an industrial environment. Raymond, thanks for making sure that I have all the resources to complete my trials at Kapuni. Tony thanks for arranging the work contract with Fonterra for the last six months of the project when I ran out of funding. My special thanks to Karl Goble, who was the go to person during my stay at Kapuni; whether it be collecting data from the plant or for discussing my mathematical model or simply someone to talk to regarding the project. Karl, you have got the most amazing collection of theses and references on crystallization!! Thank you to Sharon, Michelle and all others at the Kapuni site who made my stay there enjoyable.

Financial security is a critical factor for any PhD student. Therefore, I will like to thank the Foundation for Research Science and Technology, Government of New Zealand for providing me with the scholarship to able me to undertake this project. I would also like to thank Fonterra from the bottom of my heart for sponsoring my conference trips and university fees.

I spent most of my PhD study in Taranaki. So I will like to thank all the people who made my stay enjoyable at Hawera and helped me at some stage or other: The Turuturu cricket team and its members, Chandu Bhana and his family, Daniel Hovell and Jimmy. The cricket matches over the weekend were fun and allowed to take my mind off the PhD for little bit. Chandu, thanks for inviting us to your home and the volleyball games on Mondays. Thank you to Daniel and Jimmy for being great flat mates and providing an environment where I could return to in the evenings and relax. Daniel, I will always remember the round the mountain relay which I ran for your team. It was a great experience! Thanks.

I also need to thank people who helped me during my stay at Massey and in Palmy. First of all I will like to thank the PhD students at SEAT in the sustainable processing cluster: Horng, Ihli, Georg, Konrad, Fitri, for making my PhD experience memorable. My thanks to intern students: Amandine, Amit and Yohannes, for being part of my PhD journey at different stages. Thanks to Ann-Marie, Bruce, Anthony and John for helping me in different ways in my experimental work. Thanks to Gayle and Linda for sorting out all the administrative and financial issues during the length of my PhD.

My sincere thanks to Rajesh for being my first point of contact and helping me settle in New Zealand. Thanks to Mallesh for being a wonderful flatmate during my second term in Palmy. I will also like to thank Nilesh to whom I approached whenever I needed some advice. I will like to express my gratitude towards my friends in the badminton group for all the fun I had while playing with them. Palash, I still hope to beat you some day in singles! Special thanks to Ken Mercer for taking me on the most beautiful journeys and giving me an opportunity to see the real NZ. Ken, I will never forget our three day hiking trip to Nelson Lakes National Park!!

I now like to say thank and pay obeisance to my parents for their constant emotional and spiritual support. I cannot express in words my gratitude towards them for the sacrifices they have made. To my brother Saket, thanks for your belief in me. I also

express gratefulness to all my in-laws, especially parents-in-law for their kind words and love.

My hearty thanks to the most special person who became the part of my life during the latter half of my PhD: my wife Sneha. I need to first say sorry to you that you missed the festivities and occasions which are very special for a newly married Indian girl because I dragged you half away across the world to a completely new country. Thanks for all the love and support. Your smiling face is the biggest energy booster for me!!

Table of Contents

List of Figures

List of Tables

List of Equations