Modelling Drying Processes

This comprehensive summary of the state-of-the-art and the ideas behind the *reaction engineering approach* (REA) to drying processes is an ideal resource for researchers, academics and industry practitioners.

Starting with the formulation, modelling and applications of the *lumped-REA*, it goes on to detail the use of the REA to describe local evaporation and condensation, and its coupling with equations of conservation of heat and mass transfer, called the *spatial-REA*, to model non-equilibrium multiphase drying. Finally, it summarises other established drying models, discussing their features, limitations and comparisons with the REA.

Application examples featured throughout help fine-tune the models and implement them for process design, and the evaluation of existing drying processes and product quality during drying. Further uses of the principles of REA are demonstrated, including computational fluid dynamics-based modelling, and further expanded to model other simultaneous heat and mass transfer processes.

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> 'The *Reaction Engineering Approach* (REA), which captures basic drying physics, is a simple yet effective mathematical model for practical applications of diverse drying processes. The intrinsic "fingerprint" of the drying phenomena can, in principle, be obtained through just one accurate drying experiment. The REA is easy to use with the guidance of featured application examples given in this book. This book is highly recommended for both academics and industry practitioners involved in any aspect of thermal drying.'

> > Zhanyong Li, Tianjin University of Science and Technology, China

'An interesting book on a novel approach to mathematical modelling of an important process. *Modelling Drying Processes: A Reaction Engineering Approach* is the first attempt to summarize the REA to modelling in a single comprehensive reference source.'

Sakamon Devahastin, King Mongkut's University of Technology Thonburi, Thailand

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Modelling Drying Processes

A Reaction Engineering Approach

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Preface

Drying is one of the oldest and most effective methods for preserving food and biological materials. Low moisture content in foods prevents the growth of bacteria responsible for their deterioration so foods can have extended shelf-lives. When foods became abundant, trade became possible. Today, dried products are the main materials trading round the world but this is not limited to food products. Construction materials, textiles, electronic parts and appliances, biomass-based fuels, pharmaceutics and many other materials important to our daily lives and the business world are all included. Essentially over 80% of the products on Earth require drying as one of the steps in their production. Product quality and process parameters are interactive. Industrial drying is energy hungry; a process involving simultaneous heat, mass transfer and momentum transfer. Product quality is determined through compositional and structural rearrangements, as well as chemical reactions in some circumstances. For existing drying facilities, optimisation is often needed to achieve new goals such as energy reduction, quality improvements and development of new materials. There are also opportunities in designing dryer modifications or even brand new dryers that are superior in performance over conventional devices. Modelling of drying processes is very useful for these purposes.

A number of drying models have been proposed, which are conveniently classified into empirical and mechanistic models. The empirical models give advantages of being simple in their mathematical formulation. However, these models most often cannot explain the physics of drying and their application is limited since they are valid only for a particular set of drying conditions. On the other hand, the mechanistic models are derived based on fundamental phenomena that occur during drying. These phenomena are crucial in material science (and materials processing) though material scientists themselves may not have yet come to appreciate the process engineering aspects which impact on the product microstructure. Some of these models can capture the physics well. These models are, however, often mathematically complex and sometimes contain too many parameters, which need to be determined experimentally (prior to model predictions).

For some decades now, a comprehensive set of macroscopic equations has been developed and used to address heat and mass transfer and mechanical aspects related to drying. The application of macroscopic descriptions of drying (temperature, moisture and sometimes pressure) has been perfected over the past two decades, and relevance has been confirmed in many drying configurations. Some of these involve irreversible thermodynamics formulations, which are lengthy and have many model coefficients. These have

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become the 'classical' approach. However, this classical approach has serious limitations. The concept of multi-scale and multi-physics addresses some of these limitations, e.g. coupled meso-scale and equipment scale problems. When a local thermodynamic equilibrium is not attained, however, the time scales usually overlap. This is a real multiscale configuration and challenging in terms of the great demand in computational power and handling of mathematics. Several scales can be considered simultaneously, ranging from simple exchanges between macroscopic phases to comprehensive formulations in which time evolution of microscopic values and microscopic gradients is considered over a representative elementary volume, according to a recent review by Patrick Perre (for a review of modern computational and experimental tools relevant to the field of drying, see *Drying Technology*, 29, 1529–1541, 2011).

While exploring the detailed physics involved in drying using these multi-scale and multi-physics approaches, it is, from an engineering viewpoint, also important to develop new ideas establishing simpler models. In general, today industrial drying applications require mathematical models that are simple and easy to use. For practical purposes, an effective drying model should be simple, accurate, and able to capture the major physics of drying and its application should be robust. This model should also favour short computation time and it should be easy to establish parameters needed (experimentally) to help quicker decision-making in an industry environment (and with the lowest cost).

The reaction engineering approach (REA), which is a 'middle path' approach, perhaps between the empirical and the mechanistic models, was first thought about by the first author of this book, Chen, in 1996. Through much of the research on its possible applications, it has been revealed that the REA is indeed simple, accurate and robust enough to model many cases of drying, i.e., drying in a constant or variable environment. The REA has also been implemented in industry for prediction of spray dryer performance and shows good agreement with plant data for different scales in the dairy industry. It has also been extended to various other challenging systems of drying, such as polymer drying, intermittent drying, thermal-thick materials, infrared heating and microwave heating. The model is significantly easier to implement and requires less experimentation effort to establish the parameters needed, compared with the more fundamental models. The REA was first taken as a lumped model which does not need us to resolve the spatial distribution of water content, etc.; the lumped-REA (or L-REA), but in recent times, we have also extended the approach to describe spatially distributed systems; spatial-REA (or S-REA).

The REA approach has been initiated and exercised over the past 12 years and there is a significant amount of successful applications already illustrated. As mentioned earlier, it is a middle path between the rigorous theory that requires high-level mathematics and the empirical models that do not represent much physics. We can see, through our own practices and from other colleagues in the same area who have used the REA concept, it is a really straightforward approach to modelling some rather complex drying processes; hence, it is simple and cost-effective to establish accurate REA models to use in industry.

This book is the most fundamental and comprehensive description of the REA approach to drying modelling – the basic idea, rationale, mathematical description and implementation procedures – for various systems. This approach has been extended,

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and experimented with, by several quality Ph.D. graduates, in particular, the second author, Aditya Putranto. Regarding the other more established theories, this book not only provides essential details so the readers can refer to them but also illustrates, by comparison, the physics involved in REA concepts. The disadvantages and advantages between theories are also briefly introduced. The book should benefit both academics in drying research and practicing engineers in industry. Undergraduate students in process engineering may also find it useful for quickly setting up a drying model for design purposes. The main emphasis of this book is how to apply the REA to reality. The book will also elaborate on potential applications of similar thinking to more complex reactive systems that couple with drying processes, hopefully to foster their future development.

Here, the modern ideas of microstructure development and product qualities created by drying processes, and in turn their impacts on moisture transfer, will be introduced. This should make the book more relevant in years to come.

Xiao Dong Chen and Aditya Putranto

Historical background

During my Ph.D. study in the Chemical and Process Engineering Department at Canterbury University, Christchurch, New Zealand, (1988–1990), the main task was to establish mechanistically the understanding of moisture influence on coal oxidation and the impact of moisture transfer in a packed coal particle bed on the development of spontaneous combustion. The experimental aspect was challenging both technically and physically. In addition to coal oxidation and its racemic measurement, I became very interested in the mechanisms of water evaporation and moisture transfer (liquid and vapour) in porous material. Dr Jim Stott (Reader of Chemical Engineering) was my main supervisor and Dr John Abrahamson (Senior Lecturer), in the same department, was my cosupervisor. Jim published some of the pioneering literature on the subject of spontaneous combustion of coal (1959) and built (largely by himself) ingenious experimental rigs. Dr Abrahamson was an inspirational and distinguished individual as well who has been credited as one of the first to have made a carbon nanotube (he called it the 'carbon cylinder') (1978), a theory of ball lightening (2000) and a theory of particle collision frequency in a turbulent field (1972). John was Jim's student some years back.

Working with Jim on the subject of spontaneous combustion development in a moist coal bed has taught me that if the coal bed were completely saturated with water vapour under near ambient pressure (the institutional voids of the bed remain saturated with water vapour), the maximum temperature would remain at around 80 °C. This was predicted from a numerical spontaneous combustion model involving mass transfer of moisture within the coal bed when assuming the vapour concentration in the bed is always saturated. Jim discovered this in the late 1960s, and later, in the 1970s, a Ph.D. student of his proved this more comprehensively. This aspect was more or less republished in 1990s by a research group in Europe (who were perhaps unaware of the work by Jim and his ex-students). However, if an equilibrium relationship between moisture content in the coal particles and vapour concentration in the air surrounding the particles can be adopted, a dry spot can be predicted and the maximum temperature will exceed the boiling temperature of water, therefore rising to an elevated temperature due to oxidation heat (Chen, 1992a). Of course, there are also other influences such as porosity, oxidation rate and oxygen transfer, heat transfer and, sometimes, fluid flow due to a pressure gradient. Nevertheless, this equilibrium relationship is what we are now so familiar with, termed the equilibrium isotherm in drying literature. The oxidation rate of coal itself was also found, in my own experiments, to vary with the residual water content (Chen and Stott, 1993) and I had gone to extra lengths to try to understand this

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phenomenon. This formed the foundation of my understanding of the presence of water affecting chemical (and biochemical) reactions. In food drying, it would mean that the removal of water, to some extent, could significantly reduce rate of deterioration, giving a long shelf life to products (oxidative or microbial) (Chen and Mujumdar, 2008).

As I became aware of moisture transfer, I became very aware of the existence of a 'giant' of drying in the same department, Professor Roger Keey, who wrote the first book on drying principles and practice that was published in English. I had spent a lot of time looking for information on how to model moisture transfer, coupled with chemical reactions and heat transfer and momentum transfer. Keey's books over the years have had an impact on my own work related to this area (especially the latest one; Keey, 1992). In particular, I have picked up the essence of the *characteristic drying rate curve* (CDRC) approach. One of my friends in the drying area, Professor Tim Langrish, a Canterbury graduate, has worked extensively on this idea, which has extended Keey's views on the drying of wood and some other different materials, including foods. His postdoctoral period (after his return from Oxford University) with Professor Keev overlapped with the final year of my Ph.D. (1990). Another distinguished individual whose work has affected my own thinking has been Professor Shusheng Pang, another Canterbury graduate supervised by Roger Keey, who has published some key literature in wood drying related to the application of CDRC. CDRC captures the phenomena of drying by recognising the existence of a constant and falling drying rate period(s). The critical or transitional water content between any of the connecting rate schemes are recognised (Keey, 1992). Doctors Sandeep Chu and Peter Kho, who were student colleagues at Canterbury during the period of 1988–1990 and whose works were supervised by Professor Keey, also had an impact on my later research on drying.

Some others who also influenced me positively were Professor Miles Kennedy, Dr John Peet and Dr Maurice Allen at Canterbury. I had read many of their works during the peaceful evenings when I had pretty much the whole department to myself and some of the weekends during my Ph.D. study at the corner room on the top floor of Simon's Block. The surroundings of Canterbury University were beautiful and peaceful and gave me great times (and spaces) to spend thinking about my work and, of course, my loved ones.

I submitted my Ph.D. thesis three years after I started in late December 1987. I started working at the New Zealand Dairy Research Institute (NZDRI) (which is now the Fonterra Research Centre based in Palmerston North of New Zealand), first as an engineer and then as a senior engineer, working on spray drying and milk powder agglomeration. Dr Kevin Pearce (my section manager), who was a distinguished chemist, gave advice that I understood one has to take in order to take protein chemistry seriously when dealing with engineering problems related to dairy products. This period of time was very constructive for my career development. After coal research, I really wanted to move onto biotechnology and, at the time, the food industry was the nearest thing to biotechnology in which I could secure a good position. I was deeply involved in milk powder technology and have become very familiar with powder technology, dissolution properties of the powders, powder agglomeration and instantisation (Chen, 1992b), glass transition and stickiness (Lloyd et al., 1996), etc. I was lucky enough to

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Xiao Dong Chen (*left*) and Dr Jim Stott (*right*) working on the 2-m-long packed coal column investigating spontaneous heating of coal, 1989, University of Canterbury, Christchurch, New Zealand.

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make a significant contribution in the area of agglomeration (hardware improvement and macrostructural analysis) and new product development that was hampered by high stickiness, with large financial returns for the dairy industry.

My employment as an academic at the Department of Chemical and Materials Engineering, The University of Auckland, started in late 1993, which instantly gave me greater freedom to develop new ideas. I had great fun working at Auckland, benefitting from being surrounded by a number of highly positive individuals at the department and the school. One strong influence came from my colleagues who were experts in materials science. Among many other studies, in 1995–1996, I had developed an idea that was initially thought to be able to 'unify' drying kinetics to the equilibrium relationship (Chen and Chen, 1997). The notion of 'unified' came from, at the time, an ambitious young man (me) but later was proven to be, well, 'kinetics is just kinetics and equilibrium is equilibrium', so they don't have to be 100% linked. What had emerged, however, was that if I could find a simple relationship between the surface vapour concentration and the water content of the porous solid material being dried, noting that this surface vapour concentration less the vapour concentration in the gas phase is the driving force for moisture transfer from the porous material to the drying environment, the model could be a good alternative to the CDRC model. The obvious one for surface vapour concentration to relate to is liquid water content. At the time, I already found some issues with the CDRC approach, as uncertainty can be great depending on the drying processes considered. Keey (1992) has rightly pointed out that the CDRC model was excellent for particles or sample sizes smaller than 20 mm for constant drying conditions.

The link between the surface vapour concentration and the remaining water content in the porous material as well as the material surface temperature was eventually constructed using an Arrhenius-type relationship, which essentially suggested the evaporation of pure water and 'extraction' of the water from the inside the porous material was a reaction and the condensation/adsorption was not an activation process. This is in line with a mathematical description of evaporation and a condensation mechanism formulated by Gray and Wake (1990). Professor Brian Gray (Professor of Applied Mathematics at Sydney University at the time) is a distinguished applied mathematician (he is also a distinguished physical chemist) whom I came to know through the link between me and Professor Graeme Wake (another outstanding applied mathematician from New Zealand) and had influenced my approaches to engineering in more than just one aspect. They were not particularly interested in drying, but they were very much interested in the systems of reactions, both exothermic and endothermic. Evaporation is viewed as an endothermic reaction mathematically speaking.

My father, who was a Professor of Aerodynamics at the Chinese Academy of Sciences, visited me in New Zealand in 1996. I discussed some of my initial ideas with him and we prepared a simple paper for *Chemeca* in 1997. I was also fortunate in hosting a visiting researcher from Xian Jiao-Tong University (China) during 1996–1997, Associate Professor Guozhen Xie, who was a refrigeration expert but was daring enough to pick up drying modelling as the main topic in his year of working with me. We didn't do any experiments on drying but used data reported in the literature. However, in all cases, we had to solve an energy balance to obtain (surface) temperature of the material tested for

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the concept. Most of the examples used (Chen and Xie, 1997) were small-sized samples. Once we had the temperature-time profile for the sample of concern during drying, we could establish the activation energy in the Arrhenius equation (mentioned earlier) to demonstrate the concept.

Then, in 2000, at Auckland, I had a masters student by research, Wayne Pirini, who was interested in drying, so we started experimenting on thin-layer drying of various materials measuring both weight loss and temperature as drying proceeded. The first lot of data on activation energies obtained was reported by Chen, Pirini and Mustafa in 1996. However, I was not aware that the *Biot* number defined in heat transfer literature could not account for the conditions when evaporation occurs. This gave me an opportunity to derive a modified *Biot* number later on (the so-called *Chen–Biot* number). Then, at Auckland in the period of 2000–2004, Dr Sean Lin, my Ph.D. student at that time, did a comprehensive study on droplet drying kinetics for dairy products in particular. He had lots of practical experience before coming to me. He designed and built an excellent cost-effective droplet drying test rig and conducted probably the most careful, accurate experiments on dairy droplet drying. This has allowed the comprehensive establishment of the REA model for dairy droplets that is relevant to the spray drying industry (Chen and Lin, 2005).

Following that, two Ph.D. students under my supervision who were from India, Drs Saptarshi Kar and Kamlesh Patel, had made a significant contribution to the development of the REA concept. Saptarshi applied REA to a spatial distributed case for water transport in skin relevant to transdermal drug delivery for the first time. We deliberately ignored the liquid diffusivity to see if it really mattered. It turned out that it really did matter. Kamlesh had helped in extending the *Chen–Biot* number concept and helped to bring in a new concept called the 'composite REA approach', which describes an approach to estimating the activation energies of sugar mixtures based on the components' own activation energies. They were both tremendous students with high aptitudes to pursue basic research. Saptarshi in particular tended towards a more theoretical rigorousness. They started their Ph.Ds at Auckland and finished at Monash University.

In 12 years at Auckland, I moved from (in the English system) Lecturer (1993) to Senior Lecturer (1995) to Associate Professor (1998) to Personal Chair Professor at the age of 36 (2001). It was the most dramatic time in my life, both in career and personal life. I had my first child, Lisa, who was born in May 2000. Sad events had taken her mother away from her in 2001. I must thank the Engineering Dean at the time, Professor Peter Brothers, who, in my darkest days in 2001, promised his institution's support in allowing me to do whatever I needed to do and go wherever I needed to go without worrying about losing my job.

Beyond that, I enjoyed tremendous learning experiences, friendships, and support from my colleagues at the Department of Chemical and Materials Engineering: Professor Geoff Duffy (who was most influential individual in my stay at Auckland), Associate Professor Kevin Free, Professor John J. J. Chen, Professor Wei Gao, Professor Mohammed Farid, Professor Neil Broom, Professor George Fergusson and Dr Necati Ozkan. I was inspired by the genius professors such as Professor John Boys (Electrical Engineering), Professor Peter Hunter (Engineering Science) and Professor Debes

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Aditya Putranto (*left*) and Xiao Dong Chen (*right*), November 2012, International Drying Symposium (IDS 2012) chaired by Xiao Dong Chen, Xiamen, China.

Bhattahtrayya (Mechanical Engineering) for their innovations. I benefited tremendously from collaborating with Associate Professor Sing Kiong Nguang, who is a genius in mathematical problems in system and dynamics engineering. In that period of time at Auckland, I picked up the idea of combining process engineering and material science and became familiar with microscopy and material science techniques. My colleagues have created an incredibly creative and happy environment for me to work in. Of course, there were giants who supported me graciously over those years; Professor John Hood (Vice Chancellor of The University of Auckland and then, later, Vice Chancellor of Oxford University) and Professor Diane McCarthy (Dean of Medicine at Auckland and later President of Royal Society of New Zealand). Without their recognition of my ability and my contribution, my rapid promotion at Auckland would not have been possible.

Coming back to the main technical topic, can the REA model do the things that a CDRC model cannot? For small-size particles and constant drying conditions, CDRC seems to be very comparable with REA. With this question, and many others, I had moved to the Department of Chemical Engineering at Monash University (Melbourne, Australia) to take up the Chair of Biotechnology at Monash University in 2006.

In 2009, I had great fortune in that a high-calibre student from Indonesia, Dr Aditya Putranto, a humble young man, joined my group at Monash to do a Ph.D. with me.

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He demonstrated superior ability in testing and further developing REA for numerous applications, which are presented in this book.

In 2010, I moved to Xiamen University on the southeast coast of China, from which my grandparents graduated in history and English literature in 1930 and 1931, respectively as a National Expert Professor of Chemical Engineering (also known as the 1000-Elite Chair Professor). I have not stopped the excellent collaborations with Aditya and we continue to expend the REA. Of course, my other great Ph.D. students, Nan Fu, Winston Wu and Sam Rogers (in the period of 2008-2011), a postdoc fellow, Dr Yan Jin (in 2009), and Dr Mengwai Woo (2010–2011), have also continued to contribute experimentally, and theoretically, to the establishment of REA and its applications to the real world. Notably, Nan generated significantly new data on the REA approach to dairy droplet drying and linked drying to crystallisation and particle solubility. She has extended the techniques of single droplet drying to a more powerful means in order to understand drying-quality inter-relations. Dr Jin has comprehensively modelled the three-dimensional transient flows in large-scale spray dryers and has incorporated the REA approach. Dr Woo has independently investigated the robustness of the REA approach for modelling droplet drying in the context of computational fluid dynamics of spray dryers.

In no way can I claim it was only me who made REA development possible, but I can claim the original idea and model framework to be mostly mine. I sincerely thank all the previously mentioned individuals and others whom I have not mentioned but who have made contributions to the development of the REA in one way or another. REA is also the result of a belief that engineering theory should be as simple and robust as possible in order to enable a broad range of applications.

Finally, I would like to dedicate the book as follows:

To my lovely family starting from my wife Lily and the children Lisa, Nathan and Benjamin.

To my grandparents, my parents, my sister and brother-in-law for their neverending love and support.

To others whom I have loved and who have loved me selflessly.

Xiao Dong Chen Xiamen City, Southeast Coast of China August 2012

Aditya Putranto would like dedicate this book as follows:

To his parents and sister for their endless love and support. To the Creator and others whom have shared, and will share the love and faithfulness of the Creator.

Aditya Putranto Melbourne Australia August 2012

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