Edited by
Evangelos Tsotsas and
Arun S. Mujumdar

Modern Drying Technology

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Edited by E. Tsotsas and A. Mujumdar

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Modern Drying Technology

Volume 3: Product Quality and Formulation

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The Editors:

Prof. Evangelos Tsotsas
Otto von Guericke University
Thermal Process Engineering
Universitätsplatz 2
39106 Magdeburg
Germany

Prof. Arun S. Mujumdar
National University of Singapore
Mecnical Engineering/Block EA 07-0
9 Engineering Drive 1
Singapore 117576
Singapore

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Series Preface

The present series is dedicated to drying, i.e. to the process of removing moisture from solids. Drying has been conducted empirically since the dawn of the human race. In traditional scientific terms it is a unit operation in chemical engineering. The reason for the continuing interest in drying and, hence, the motivation for the series concerns the challenges and opportunities. A permanent challenge is connected to the sheer amount and value of products that must be dried – either to attain their functionalities, or because moisture would damage the material during subsequent processing and storage, or simply because customers are not willing to pay for water. This comprises almost every material used in solid form, from foods to pharmaceuticals, from minerals to detergents, from polymers to paper. Raw materials and commodities with a low price per kilogram, but with extremely high production rates, and also highly formulated, rather rare but very expensive specialties have to be dried.

This permanent demand is accompanied by the challenge of sustainable development providing welfare, or at least a decent living standard, to a still-growing humanity. On the other hand, opportunities emerge for drying, as well as for any other aspect of science or living, from either the incremental or disruptive development of available tools. This duality is reflected in the structure of the book series, which is planned for five volumes in total, namely:

Volume 1: Computational tools at different scales
Volume 2: Experimental techniques
Volume 3: Product quality and formulation
Volume 4: Energy savings
Volume 5: Process intensification

As the titles indicate, we start with the opportunities in terms of modern computational and experimental tools in Volumes 1 and 2, respectively. How these opportunities can be used in fulfilling the challenges, in creating better and new products, in reducing the consumption of energy, in significantly improving existing or introducing new processes will be discussed in Volumes 3, 4 and 5. In this sense, the first two volumes of the series will be driven by science; the last three will try to show how engineering science and technology can be translated into progress.

In total, the series is designed to have both common aspects with and essential differences from an extended textbook or a handbook. Textbooks and handbooks usually refer to well-established knowledge, prepared and organized either for learning or for application in practice, respectively. On the contrary, the ambition of the present series is to move at the frontier of "modern drying technology", describing things that have recently emerged, mapping things that are about to emerge, and also anticipating some things that may or should emerge in the near future. Consequently, the series is much closer to research than textbooks or handbooks can be. On the other hand, it was never intended as an anthology of research papers or keynotes – this segment being well covered by periodicals and conference proceedings. Therefore, our continuing effort will be to stay as close as possible to a textbook in terms of understandable presentation and as close as possible to a handbook in terms of applicability.

Another feature in common with an extended textbook or a handbook is the rather complete coverage of the topic by the entire series. Certainly, not every volume or chapter will be equally interesting for every reader, but we do hope that several chapters and volumes will be of value for graduate students, for researchers who are young in age or thinking, and for practitioners from industries that are manufacturing or using drying equipment. We also hope that the readers and owners of the entire series will have a comprehensive access not to all, but to many significant recent advances in drying science and technology. Such readers will quickly realize that modern drying technology is quite interdisciplinary, profiting greatly from other branches of engineering and science. In the opposite direction, not only chemical engineers, but also people from food, mechanical, environmental or medical engineering, material science, applied chemistry or physics, computing and mathematics may find one or the other interesting and useful results or ideas in the series.

The mentioned interdisciplinary approach implies that drying experts are keen to abandon the traditional chemical engineering concept of unit operations for the sake of a less rigid and more creative canon. However, they have difficulties of identification with just one of the two new major trends in chemical engineering, namely process-systems engineering or product engineering. Efficient drying can be completely valueless in a process system that is not efficiently tuned as a whole, while efficient processing is certainly valueless if it does not fulfil the demands of the market (the customer) regarding the properties of the product. There are few topics more appropriate in order to demonstrate the necessity of simultaneous treatment of product and process quality than drying. The series will try to work out chances that emerge from this crossroads position.

One further objective is to motivate readers in putting together modules (chapters from different volumes) relevant to their interests, creating in this manner individual, task-oriented threads trough the series. An example of one such thematic thread set by the editors refers to simultaneous particle formation and drying, with a focus on spray fluidized beds. From the point of view of process-systems engineering, this is process integration – several "unit operations" take place in the same equipment.

On the other hand, it is product engineering, creating structures – in many cases nanostructures – that correlate with the desired application properties. Such properties are distributed over the ensemble (population) of particles, so that it is necessary to discuss mathematical methods (population balances) and numerical tools able to resolve the respective distributions in one chapter of Volume 1. Measuring techniques providing access to properties and states of the particle system will be treated in one chapter of Volume 2. In Volume 3, we will attempt to combine the previously introduced theoretical and experimental tools with the goal of product design. Finally, important issues of energy consumption and process intensification will appear in chapters of Volumes 4 and 5. Our hope is that some thematic combinations we have not even thought about in our choice of contents will arise in a similar way.

As the present series is a series of edited books, it can not be as uniform in either writing style or notation as good textbooks are. In the case of notation, a list of symbols has been developed and will be printed in the beginning of every volume. This list is not rigid but foresees options, at least partially accounting for the habits in different parts of the world. It has been recently adopted as a recommendation by the Working Party on Drying of the European Federation of Chemical Engineering (EFCE). However, the opportunity of placing short lists of additional or deviant symbols at the end of every chapter has been given to all authors. The symbols used are also explained in the text of every chapter, so that we do not expect any serious difficulties in reading and understanding.

The above indicates that the clear priority in the edited series was not in uniformity of style, but in the quality of contents that are very close to current international research from academia and, where possible, also from industry. Not every potentially interesting topic is included in the series, and not every excellent researcher working on drying contributes to it. However, we are very confident about the excellence of all research groups that we were able to gather together, and we are very grateful for the good cooperation with all chapter authors. The quality of the series as a whole is set mainly by them; the success of the series will primarily be theirs. We would also like to express our acknowledgements to the team of Wiley-VCH who have done a great job in supporting the series from the first idea to realization. Furthermore, our thanks go to Mrs Nicolle Degen for her additional work, and to our families for their tolerance and continuing support.

Last but not least, we are grateful to the members of the Working Party on Drying of the EFCE for various reasons. First, the idea about the series came up during the annual technical and business meeting of the working party 2005 in Paris. Secondly, many chapter authors could be recruited among its members. Finally, the Working Party continues to serve as a panel for discussion, checking and readjustment of our conceptions about the series. The list of the members of the working party with their affiliations is included in every volume of the series in the sense of acknowledgement, but also in order to promote networking and to provide access to national working parties, groups and individuals. The present edited books are

complementary to the regular activities of the EFCE Working Party on Drying, as they are also complementary to various other regular activities of the international drying community, including well-known periodicals, handbooks, and the International Drying Symposia.

June 2007

Evangelos Tsotsas
Arun S. Mujumdar

Preface of Volume 3

The first two volumes of this series have treated "Computational tools at different scales" and "Experimental techniques" that can empower "Modern Drying Technology" with the aim of producing superior products with better processes. Now, it is time to turn from the means to the goal, treating "Product quality and formulation" in Volume 3. This emphasis on the product is deliberate, because even the most efficient process is not of real value, if not able to fulfill – if not push – the requirements of the market. The topic is presented in seven chapters:

Chapter 1: Quality changes in food materials as influenced by drying processes
Chapter 2: Impact of drying on the mechanical properties and crack formation in rice
Chapter 3: Characterization and control of physical quality factors during freeze-drying of pharmaceuticals in vials
Chapter 4: In-line product quality control of pharmaceuticals in freeze-drying processes
Chapter 5: Understanding and preventing structural changes during drying of gels
Chapter 6: Morphology and properties of spray-dried particles
Chapter 7: Particle formulation in spray fluidized beds

Chapter 1 refers to a big, utterly important group of products to be dried, namely foods. It summarizes food properties, introduces the glass transition temperature as a humidity dependent landmark between the glassy and the rubbery state of amorphous materials, and discusses biochemical, physical and mechanical transformations that can take place during drying. Furthermore, it connects drying with quality changes during storage and with properties relevant to the final use of the processed food.

One good example of what can happen after drying is the fissuring and breakage of rice kernels due to stresses and strains that developed during the process. Therefore, this example is used in Chapter 2 in order to show how the previously discussed general principles can be cast into specific and precise characterization methods and models for the preservation of the quality of a valuable but perishable good.

In Chapter 3 the focus is shifted to pharmaceuticals, specifically to active ingredients with a high molecular weight, such as therapeutic proteins or enzymes. Such compounds are usually produced biotechnologically, so that they often have to be transformed from an aqueous solution to a solid form. This is commonly carried out by freezing and then freeze-drying, in order to protect the complex molecular structure from deterioration. The chapter discusses thoroughly, what kinds of damage can occur during the process, and how they can be avoided. And, it shows impressively, how intimate the interrelation of freeze-drying to the preceding process of freezing is. This interconnection results from the fact that the solid skeleton of freeze-dried cakes is created during freezing by the size and spatial placement of the ice crystals. Such causality offers rich opportunities of beneficial manipulation by changes in the freezing protocol, controlled nucleation or annealing, which are worked out in detail.

Though the degradation of pharmaceuticals during freeze-drying is not permissible too conservative an operation also should be avoided, because it is very expensive. The key for resolving this dilemma between product quality and process efficiency is monitoring and control. Consequently, methods that can be used for monitoring and control during freeze-drying of pharmaceuticals are presented in Chapter 4. This is done in a very comprehensive and precise way, distinguishing among methods that refer to single vials, groups of vials, and the entire dryer for the primary or the secondary period of drying. Close reference to process analytical technology (PAT) is given throughout.

Gels are a class of materials with high porosity, very small primary particle size, and a plethora of possible applications. However, such applications require that the gels can be dried without destroying the structures which are generic for their properties. This is not an easy task, because very small primary particles imply very high capillary forces during drying, so that the material can crack and break. Chapter 5 points out that convective drying may still be successful if applied in an educated way, and compares with numerous alternatives, such as freeze-drying and supercritical drying. Apart from the detailed discussion of processing options, the preparation and the characterization of gel materials are elucidated.

Though the preservation of existing structures is a big goal, structures and the conjugated properties can even be created by drying. This is always the case when the removal of water or some other solute is accompanied by the formation of the solid phase, as in spray drying, which is treated comprehensively in Chapter 6. This chapter refers to solutions of components with a low or high molecular weight, as well as to suspensions of small or large particles, and shows how drying conditions and material properties influence the morphology of the resulting products. Methods of formulation by encapsulation of, for example, flavors or enzymes, are presented in detail, including stability and quality of the obtained products.

The idea of formulation by drying is elaborated further in Chapter 7. Here, drying after spraying on fluidized particles with the aim of producing agglomerates, layered granules, or coatings is discussed. It is worked out on many examples, how the processes and the products can be enhanced by manipulation of material properties, operating conditions, and apparatus design. The physical background is explained

down to the molecular scale in order to derive conditions for adhesion around small particle contacts. Understanding and characterization of properties relevant to the processing or final use of the particles are, again, important issues. Furthermore, modeling tools with different degrees of resolution and sophistication – such as discrete particle modeling, Monte Carlo simulations, and neural networks – which can separately or in combination support process and product development are presented.

Readers looking for thematic threads within the Modern Drying Technology series will easily recognize many, including those between the present:

– Chapter 1 and Chapter 2 of Vol. 2 (drying of foods)
– Chapters 2 and 5 and Chapters 3 and 4 of Vol. 1 (thermo-mechanics)
– Chapter 4 and Chapter 1 of Vol. 2 (monitoring)
– Chapter 5 and Chapter 3 of Vol. 2 (x-ray tomography)
– Chapter 6 and Chapter 5 of Vol. 1 (spray drying)
– Chapter 7 and Chapter 6 of Vol. 1, as well as Chapter 5 of Vol. 2 (fluidized bed formulation)

Readers interested in transport phenomena at different scales will find molecular, pore-scale, particle-scale and particle system or processing equipment considerations, as in every volume of the series, and those aiming at interdisciplinary approaches will see clear links to food engineering, pharmaceutical technology, biotechnology, mechanics, and material science. People looking for their specific product may not be able to find it in the present volume, but they may learn from methods and approaches successfully applied to other products. For a book without encyclopedic ambitions, which aims at the educated use of modern scientific methods in practice, this would be the biggest success.

As to the acknowledgements, for Volume 3 they are identical to those in the series preface. We would like to stress them by reference and not repeat them here.

June 2011

Evangelos Tsotsas
Arun S. Mujumdar

List of Contributors

Editors

Prof. Evangelos Tsotsas
Otto von Guericke University
Magdeburg
Thermal Process Engineering
PSF 4120
39106 Magdeburg
Germany
Email: Evangelos.Tsotsas@ovgu.de

Prof. Arun S. Mujumdar
Dept. of Mechanical Engineering
9 Engineering Drive 1
Singapore 117576
Singapore
Email: mpeasm@nus.edu.sg

Authors

Prof. Julien Andrieu
Université C. Bernard - Lyon1/
E.S.C.P.E.
Laboratoire d'Automatique et de Génie des Procédés (LAGEP)
UMR Q 5007 CNRS UCB
Bâtiment 308G
43 boulevard du 11 Novembre 1918
69622 Villeurbanne Cedex
France
Email: andrieu@lagep.univ-lyon1.fr

Dr. Sergiy Antonyuk
Technical University Hamburg-Harburg
Solids Process Engineering and Particle Technology
21073 Hamburg
Germany
Email: antonyuk@tu-harburg.de

Prof. Antonello A. Barresi
Politecnico di Torino
Dipartimento di Scienza dei Materiali e Ingegneria Chimica
Corso Duca degli Abruzzi 24
10129 Torino
Italy
Email: antonello.barresi@polito.it

Dr. Catherine Bonazzi
INRA/AgroParisTech
UMR1145 Ingénierie Procédés Aliments
1 avenue des Olympiades
91300 Massy
France
Email: catherine.bonazzi@agroparistech.fr

Dr. Francis Courtois
INRA/AgroParisTech
UMR1145 Ingénierie Procédés Aliments
1 avenue des Olympiades
91300 Massy
France
Email: francis.courtois@agroparistech.fr

Prof. Elisabeth Dumoulin
INRA/AgroParisTech
UMR1145 Ingénierie Procédés Aliments
1 avenue des Olympiades
91300 Massy
France
Email: elisabeth.dumoulin@agroparistech.fr

Dr. Davide Fissore
Politecnico di Torino
Dipartimento di Scienza dei Materiali e Ingegneria Chimica
Corso Duca degli Abruzzi 24
10129 Torino
Italy
Email: davide.fissore@polito.it

Prof. Takeshi Furuta
Tottori University
Department of Chemistry and Biotechnology
Graduate School of Engineering
4-101, Koyama-minami
Tottori, 680-8552
Japan
Email: takeshi@bio.tottori-u.ac.jp

Prof. Stefan Heinrich
Technical University Hamburg-Harburg
Solids Process Engineering and Particle Technology
21073 Hamburg
Germany
Email: Stefan.heinrich@tu-harburg.de

Dip.-Ing. Michael Jacob
Glatt Ingenieurtechnik GmbH
Nordslrasse 12
99427 Weimar
Germany
Email: info@glatt-weimar.de

Prof. Wahbi Jomaa
University Bordeaux 1
Laboratory TREFLE
Esplanade des Arts et Métiers
33405 Talence
France
Email: wahbi.jomaa@u-bordeaux1.fr

Prof. Angélique Léonard
Laboratoire de Génie Chimique
Département de Chimie Appliquée
Université de Liège
Bâtiment B6c - Sart-Tilman
4000 Liège
Belgium
Email: a.leonard@ulg.ac.be

Jun.-Prof. Thomas Metzger
Otto von Guericke University Magdeburg
Thermal Process Engineering
PSF 4120
39106 Magdeburg
Germany
Email: thomas.metzger@ovgu.de

Prof. Stefan Palzer
Nestlé Product Technology Centre York
Nestec York Ltd
P.O. Box 204
Haxby Road
York YO 91 1XY
United Kingdom
Email: stefan.palzer@rd.nestle.com

Jun.-Prof. Mirko Peglow
Otto von Guericke University
Magdeburg
Thermal Process Engineering
PSF 4120
39106 Magdeburg
Germany
Email: Mirko.Peglow@ovgu.de

Prof. Hajime Tamon
Department of Chemical Engineering
Kyoto University
Katsura
Kyoto 615-8510
Japan
Email: tamon@cheme.kyoto-u.ac.jp

Prof. Evangelos Tsotsas
Otto von Guericke University
Magdeburg
Thermal Process Engineering
PSF 4120
39106 Magdeburg
Germany
Email: Evangelos.Tsotsas@ovgu.de

Dr. Séverine Vessot
Laboratoire d'Automatique et de Génie
des Precédés (LAGEP)
UMR Q 5007 CNRS UCB Lyon1-CPE
Bât. 308G, 43 Bd. du 11 Novembre 1918
69622 Villeurbanne Cedex
France
Email: vessot@lagep.cpe.fr

Prof. Peter Walzel
Universität Dortmund
Fachbereich Bio- und
Chemieingenieurwesen
Particle Technology
Emil Figge Str. 68
44227 Dortmund
Germany
Email: peter.walzel@
bci.tu-dortmund.de

Recommended Notation

- Alternative symbols are given in brackets
- Vectors are denoted by bold symbols, a single bar, an arrow or an index (e.g., index: *i*)
- Tensors are denoted by bold symbols, a double bar or a double index (e.g., index: *i*, *j*)
- Multiple subscripts should be separated by colon (e.g., $\rho_{p,dry}$: density of dry particle)

A	surface area	m^2
a_w	water activity	—
B	nucleation rate	$kg^{-1}\,m^{-1}\,s^{-1}$
b	breakage function	m^{-3}
$C(K)$	constant or coefficient	various
c	specific heat capacity	$J\,kg^{-1}\,K^{-1}$
D	equipment diameter	m
$D(\delta)$	diffusion coefficient	$m^2\,s^{-1}$
d	diameter or size of solids	m
E	energy	J
F	mass flux function	—
$F(\dot{V})$	volumetric flow rate	$m^3\,s^{-1}$
f	relative (normalized) drying rate	—
f	multidimensional number density	—
G	shear function or modulus	Pa
G	growth rate	$kg\,s^{-1}$
g	acceleration due to gravity	$m\,s^{-2}$
H	height	m
H	enthalpy	J
H	Heaviside step function	—
h	specific enthalpy (dry basis)	$J\,kg^{-1}$
$h(\alpha)$	heat-transfer coefficient	$W\,m^{-2}\,K^{-1}$
$\tilde{h}(h_N)$	molar enthalpy	$J\,mol^{-1}$
Δh_v	specific enthalpy of evaporation	$J\,kg^{-1}$
I	total number of intervals	—

J	numerical flux function	—
J	Jacobian matrix	various
$j(\dot{m}, J)$	mass flux, drying rate	$\mathrm{kg\,m^{-2}\,s^{-1}}$
K	dilatation function or bulk modulus	Pa
$k(\beta)$	mass transfer coefficient	$\mathrm{m\,s^{-1}}$
L	length	m
$M(m)$	mass	kg
$\tilde{M}(M, M_\mathrm{N})$	molecular mass	$\mathrm{kg\,kmol^{-1}}$
$\dot{M}(W)$	mass flow rate	$\mathrm{kg\,s^{-1}}$
$\dot{m}(J, j)$	mass flux, drying rate	$\mathrm{kg\,m^{-2}\,s^{-1}}$
$\dot{m}$	volumetric rate of evaporation	$\mathrm{kg\,m^{-3}\,s^{-1}}$
N	number	—
N	molar amount	mol
$\dot{N}(W_\mathrm{N})$	molar flow rate	$\mathrm{mol\,s^{-1}}$
n	molar density, molar concentration	$\mathrm{mol\,m^{-3}}$
n	number density	$\mathrm{m^{-3}}$
n	outward normal unit vector	
$\dot{n}(J_\mathrm{N})$	molar flux	$\mathrm{mol\,m^{-2}\,s^{-1}}$
P	power	W
P	total pressure	$\mathrm{kg\,m\,s^{-2}}$
p	partial pressure/vapor pressure of component	$\mathrm{kg\,m\,s^{-2}}$
$\dot{Q}(Q)$	heat flow rate	W
$\dot{q}(q)$	heat flux	$\mathrm{W\,m^{-2}}$
R	equipment radius	m
R	individual gas constant	$\mathrm{J\ kg^{-1}\,K^{-1}}$
$\tilde{R}(R_\mathrm{N})$	universal gas constant	$\mathrm{J\,kmol^{-1}\,K^{-1}}$
r	radial coordinate	m
r	pore (throat) radius	m
S	saturation	—
S	selection function	$\mathrm{s^{-1}}$
s	boundary-layer thickness	m
T	temperature	K, °C
t	time	s
u	velocity, usually in z-direction	$\mathrm{m\,s^{-1}}$
u	displacement	m
V	volume, averaging volume	$\mathrm{m^3}$
$\dot{V}(F)$	volumetric flow rate	$\mathrm{m^3\,s^{-1}}$
v	specific volume	$\mathrm{m^3\,kg^{-1}}$
v	general velocity, velocity in x-direction	$\mathrm{m\,s^{-1}}$
W	weight force	N
$W(\dot{M})$	mass flow rate	$\mathrm{kg\,s^{-1}}$
w	velocity, usually in y-direction	$\mathrm{m\,s^{-1}}$
X	solids moisture content (dry basis)	—

x	mass fraction in liquid phase	—
x	particle volume in population balances	m^3
x	general Eulerian coordinate, coordinate (usually lateral)	m
x_0	general Lagrangian coordinate	m
$\tilde{x}(x_N)$	molar fraction in liquid phase	—
Y	gas moisture content (dry basis)	—
y	spatial coordinate (usually lateral)	m
$y(\omega)$	mass fraction in gas phase	—
$\tilde{y}(y_N)$	molar fraction in gas phase	—
z	spatial coordinate (usually axial)	m

Operators

∇	gradient operator
$\nabla.$	divergence operator
Δ	difference operator

Greek letters

$\alpha(h)$	heat-transfer coefficient	$W\,m^{-2}\,K^{-1}$
$\beta(k)$	mass-transfer coefficient	$m\,s^{-1}$
β	aggregation kernel	s^{-1}
δ	Dirac-delta distribution	
$\delta(D)$	diffusion coefficient	$m^2\,s^{-1}$
ε	voidage	—
ε	emissivity	—
ε	small-scale parameter for periodic media	—
ε	strain	—
η	efficiency	—
θ	angle, angular coordinate	rad
κ	thermal diffusivity	$m^2\,s^{-1}$
λ	thermal conductivity	$W\,m^{-1}\,K^{-1}$
μ	dynamic viscosity	$kg\,m^{-1}\,s^{-1}$
μ	moment of the particle-size distribution	various
ν	kinematic viscosity	$m^2\,s^{-1}$
π	circular constant	—
ρ	density, mass concentration	$kg\,m^{-3}$
$\sum$	summation operator	
σ	surface tension	$N\,m^{-1}$
σ	Stefan–Boltzmann constant for radiative heat transfer	$W\,m^{-2}\,K^{-4}$
σ	standard deviation (of pore-size distribution)	m
σ	stress	Pa
τ	dimensionless time	—

Φ	characteristic moisture content	—
φ	relative humidity	—
φ	phase potential	Pa
ω	angular velocity	rad s^{-1}
ω(y)	mass fraction in gas phase	—

Subscripts

a	at ambient conditions
as	at adiabatic saturation conditions
b	bound water
bed	bed
c	cross section
c	capillary
cr	at critical moisture content
D	drag
dry	dry
dp	at dewpoint
eff	effective
eq	equilibrium (moisture content)
f	friction
g	gas (dry)
H	wet (humid) gas
i	inner
i,1,2,. . .	component index, particle index
i,*j*,*k*	coordinate index, $i, j, k = 1$ to 3
in	inlet value
l	liquid (alternative: as a superscript)
m	mean value
max	maximum
mf	at minimum fluidization
min	minimum
N	molar quantity
o	outer
out	outlet value
P	at constant pressure
p	particle
pbe	population balance equation
ph	at the interface
r	radiation
rel	relative velocity
s	solid (compact solid phase), alternative: as a superscript
S	at saturation conditions
surf	surface
V	based on volume

v	vapor, evaporation
w	water
w	wall
wb	at wet-bulb conditions
wet	wet
∞	at large distance from interface

Superscripts, special symbols

v	volumetric strain
*	rheological strain
*	at saturation conditions
$\bar{}$ or $\langle\rangle$	average, phase average
$\bar{}\alpha$ or $\langle\rangle^\alpha$	intrinsic phase average
$\tilde{}$	spatial deviation variable

EFCE Working Party on Drying: Address List

Dr. Odilio Alves-Filho
Norwegian University of Science and Technology
Department of Energy and Process Engineering
Kolbjørn Hejes vei 1B
7491 Trondheim
Norway
odilio.filho@gmail.com

Prof. Julien Andrieu *(delegate)*
UCB Lyon I/ESCPE
LAGEP UMR CNRS 5007
batiment 308 G
43 boulevard du 11 novembre 1918
69622 Villeurbanne cedex
France
andrieu@lagep.univ-lyon1.fr

Dr. Ir. Paul Avontuur
Glaxo Smith Kline
New Frontiers Science Park H89
Harlow CM19 5AW
UK
Paul.Avontuur@gsk.com

Prof. Christopher G. J. Baker
Drying Associates
Harwell International Business Centre
404/13 Harwell
Didcot
Oxfordshire OX11 ORA
UK
ChristopherGJ.Baker@gmail.com

Prof. Antonello Barresi *(delegate)*
Politecnico di Torino
Dip. Scienza dei Materiali e Ingegneria Chimica
Corso Duca degli Abruzzi 24
10129 Torino
Italy
antonello.barresi@polito.it

Dr. Rainer Bellinghausen *(delegate)*
Bayer Technology Services GmbH
BTS-PT-PT-PDSP
Building E 41
51368 Leverkusen
Germany
rainer.bellinghausen@bayertechnology.com

Dr. Carl-Gustav Berg
Abo Akademi
Process Design Laboratory
Biskopsgatan 8
20500 Abo
Finland
cberg@abo.fi

Dr. Catherine Bonazzi *(delegate)*
AgroParisTech – INRA
JRU for Food Process Engineering
1, Avenue des Olympiades
91744 Massy cedex
France
Catherine.Bonazzi@agroparistech.fr

Paul Deckers M.Sc. *(delegate)*
Bodec, Process Optimization and Development
Industrial Area 't Zand
Bedrijfsweg 1
5683 CM Best
The Netherlands
deckers@bodec.nl

Prof. Stephan Ditchev
University of Food Technology
26 Maritza Blvd
4002 Plovdiv
Bulgaria
sditchev@gmail.bg

Dr. German I. Efremov
Pavla Korchagina 22
129278 Moscow
Russia
efremov_german@mail.ru

Prof. Trygve Eikevik
Norwegian University of Science and Technology
Department of Energy and Process Engineering
Kolbjørn Hejes vei 1B
7491 Trondheim
Norway
Trygve.M.Eikevik@ntnu.no

Dr.-Ing. Ioannis Evripidis
Dow Deutschland GmbH & Co. OHG
P.O. Box 1120
21677 Stade
Germany
evripidis@dow.com

Prof. Dr. Istvan Farkas *(delegate)*
Szent Istvan University
Department of Physics and Process Control
Pater K. u. 1.
2103 Godollo
Hungary
Farkas.Istvan@gek.szie.hu

Dr.-Ing. Dietrich Gehrmann
Wilhelm-Hastrich-Str. 12
51381 Leverkusen
Germany
Dietrich.Gehrmann@t-online.de

Prof. Dr.-Ing. Adrian-Gabriel Ghiaus *(delegate)*
Technical University of Civil Engineering
Thermal Engineering Department
Bd. P. Protopopescu 66
021414 Bucharest
Romania
ghiaus@instalatii.utcb.ro

Prof. Dr.-Ing. Gheorghita Jinescu
University "Politehnica" din Bucuresti
Faculty of Industrial Chemistry,
Department of Chemical Engineering
1, Polizu street
Building F Room F210
78126 Bucharest
Romania
g_jinescu@chim.upb.ro

Prof. Dr. Gligor Kanevce
St. Kliment Ohridski University
Faculty of Technical Sciences
ul. Ivo Ribar Lola b.b.
Bitola
FYR of Macedonia
kanevce@osi.net.mk

Prof. Dr. Markku Karlsson *(delegate)*
UPM-Kymmene Corporation
P.O. Box 380
00101 Helsinki
Finland
markku.karlsson@upm-kymmene.com

Ir. Ian C. Kemp *(delegate)*
GMS, GSK
Priory Street
Ware, SG12 0XA
UK
ianckemp@btinternet.com

Prof. Dr. Ir. P.J.A.M. Kerkhof
Eindhoven University of Technology
Department of Chemical Engineering
P.O. Box 513
5600 MB Eindhoven
The Netherlands
p.j.a.m.kerkhof@tue.nl

Prof. Matthias Kind
Universität Karlsruhe (TH)
Institut für Thermische
Verfahrenstechnik
Kaiserstr. 12
76128 Karlsruhe
Germany
matthias.kind@ciw.uni-karlsruhe.de

Prof. Eli Korin
Ben-Gurion University of the Negev
Chemical Engineering Department
Beer-Sheva 84105
Israel
ekorin@bgumail.bgu.ac.il

Emer. Prof. Ram Lavie
Technion – Israel Institute of Technolgy
Department of Chemical Engineering
Technion City
Haifa 32000
Israel
lavie@tx.technion.ac.il

Dr. Ir. Angélique Léonard *(delegate)*
Université de Liège, Département de
Chimie Appliquée
Laboratoire de Génie Chimique
Bâtiment B6c - Sart-Tilman
4000 Liège
Belgium
A.Leonard@ulg.ac.be

Jean-Claude Masson
RHODIA, Recherches et Technologies
85 avenue des Frères Perret
BP 62
69192 Saint-Fons Cedex
France
jean-claude.masson@eu.rhodia.com

Prof. Natalia Menshutina
Mendeleev University of Chemical
Technology of Russia (MUCTR)
High Technology Department
125047 Muisskaya sq.9
Moscow
Russia
chemcom@muctr.edu.ru

Jun.-Prof. Dr. Thomas Metzger
Otto-von-Guericke-University
Thermal Process Engineering
P.O. Box 4120
39016 Magdeburg
Germany
thomas.metzger@vst.uni-magdeburg.
de

Prof. Antonio Mulet Pons *(delegate)*
Universitat Politecnica de Valencia
Departament de Tecnologia d'Aliments
Cami de Vera s/n
46071 Valencia
Spain
amulet@tal.upv.es

Prof. Zdzislaw Pakowski *(delegate)*
Technical University of Lodz
Faculty of Process and Environmental Engineering
ul. Wolczanska 213
93-005 Lodz
Poland
pakowski@wipos.p.lodz.pl

Prof. Patrick Perré *(delegate, chairman of WP)*
AgroParisTech
14 Rue Girardet
54042 Nancy
France
perre@nancy-engref.inra.fr

Dr. Roger Renström
Karlstad University
Department of Environmental and Energy Systems
Universitetsgatan 2
65188 Karlstad
Sweden
roger.renstrom@kau.se

Prof. Michel Roques
Université de Pau et des Pays de l'Adour
ENSGTI, 5 rue Jules- Ferry
64000 Pau
France
michel.roques@univ-pau.fr

Dr. Carmen Rosselló *(delegate)*
University of Iles Baleares
Dep. Quimica
Ctra Valldemossa km 7.5
07122 Palme Mallorca
Spain
Carmen.rossello@uib.es

Emer. Prof. G. D. Saravacos *(delegate)*
Nea Tiryntha
21100 Nauplion
Greece
gsaravac@otenet.gr

Dr.-Ing. Michael Schönherr
BASF, GCT/T - L 540
Research Manager Drying
Process Engineering
67056 Ludwigshafen
Germany
michael.schoenherr@basf.com

Prof. Dr.-Ing. Ernst-Ulrich Schlünder
Lindenweg 10
76275 Ettlingen
Germany
euschluender@t-online.de

Dr. Alberto M. Sereno *(delegate)*
University of Porto
Department of Chemical Engineering
Rua Dr Roberto Frias
4200-465 Porto
Portugal
sereno@fe.up.pt

Dr. Milan Stakic
Vinča Institute for Nuclear Sciences
Center NTI
P.O. box 522
11001 Belgrade
Serbia
stakicm@yahoo.com

Prof. Stig Stenstrom *(delegate)*
Lund University
Institute of Technology
Department of Chemical Engineering
P.O. Box 124
22100 Lund
Sweden
stig.stenstrom@chemeng.lth.se

Prof. Ingvald Strommen *(delegate)*
Norwegian University of Science and Technology
Department of Energy and Process Engineering
Kolbjørn Hejes vei 1b
7491 Trondheim
Norway
ingvald.strommen@ntnu.no

Prof. Czeslaw Strumillo *(delegate)*
Technical University of Lodz
Faculty of Process and Environmental Engineering
Lodz Technical University
ul. Wolczanska 213
93-005 Lodz
Poland
cstrumil@wipos.p.lodz.pl

Prof. Radivoje Topic *(delegate)*
University of Belgrade
Faculty of Mechanical Engineering
27, marta 80
11000 Beograd
Serbia
topic@eunet.yu

Prof. Dr.-Ing. Evangelos Tsotsas *(delegate, former chairman of WP)*
Otto-von-Guericke-University
Thermal Process Engineering
P.O. Box 4120
39016 Magdeburg
Germany
evangelos.tsotsas@vst.uni-magdeburg.de

Dr. Henk C. van Deventer *(delegate)*
TNO Quality of Life
P.O. Box 342
7300 AH Apeldoorn
The Netherlands
henkvandeventer@tno.nl

Michael Wahlberg M.Sc.
Niro
Gladsaxevej 305
2860 Soeborg
Denmark
mw@niro.dk

Prof. Roland Wimmerstedt
Lund University
Institute of Technology
Department of Chemical Engineering
P.O. Box 124
22100 Lund
Sweden
Roland.Wimmerstedt@chemeng.lth.se

Dr. Bertrand Woinet *(delegate)*
SANOFI-CHIMIE, CDP
bât. 8600
31-33 quai armand Barbès
69583 Neuville sur Saône cedex
France
bertrand.woinet@sanofi-aventis.com

Prof. Ireneusz Zbicinski
Lodz Technical University
Faculty of Process and Environmental Engineering
ul. Wolczanska 213
93-005 Lodz
Poland
zbicinsk@mail.p.lodz.pl