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SOIL ADHERENCE TO SOLID SURFACES: RELATION WITH FOULING AND CLEANING

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Abstract:

This doctoral research was realized within the frame of the SMARTNET Project which aimed at developing coatings to improve the cleanability of stainless steel, targeting open surface applications. Throughout this thesis, the radial-flow cell was selected to study the removal of different soils due to its ability to generate well-controlled wall shear stress distributions on the investigated surfaces. Model surfaces were selected for their different physico-chemical and mechanical properties to study the interactions between the soils and the surfaces in detail.

A thin layer chromatography sprayer giving a narrower and more reproducible droplet sizes distribution was preferred to mimic splashing and produce controlled spatters. The first experimental campaign involving oil droplets showed that the analytical models available to relate the detachment radius with the critical wall shear stress (minimal wall shear stress required for soil detachment) and the soil adhesion strength in the radial flow cell could only be applied for weakly adherent soils for which removal occurs below 3 Pa, due to the complex hydrodynamics near the inlet.

Consequently, the flow inside the radial-flow cell has been characterized using computational fluid dynamics over the whole inlet laminar regime and validated experimentally. Studying the adherence of starch granule aggregates in the radial-flow cell revealed that the conversion of critical radius into critical wall shear stress may be biased when the adhering aggregate height is not negligible with respect to the channel height and when the adherence is such that flow rates above creeping flow conditions are required for soil detachment.

The influence of several environmental factors and substrate properties was then examined to improve the understanding of the mechanisms affecting soiling and cleanability. By influencing droplet spreading and competition between capillary forces at the granule-substrate and granule-granule interfaces, substrate wettability affects the shape and compactness of the adhering aggregates, the efficiency of shear forces upon cleaning, and finally the adherence of soiling particles. Macromolecules originating from the starch granules suspension are adsorbed on the substrate from the liquid phase or carried by the retracting film and accumulated at the granule-substrate interface. They influence granule adherence by acting as an adhesive joint, the properties of which seem to be influenced by the detailed history of drying and exposure to humidity.

On compliant substrates, the aggregate-substrate interactions induce stresses at the granule-substrate interface which may lead to substrate deformation and promote a more intimate contact between the granules and their substrate, thereby appreciably increasing adherence.

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Résumé :

Cette recherche doctorale s’inscrit dans le cadre du projet SMARTNET, visant à produire des revêtements pour améliorer l’aptitude au nettoyage de surfaces ouvertes en acier inoxydable. Dans ce contexte, la cellule à flux radial a été retenue pour étudier le détachement de différentes souillures grâce à sa capacité à générer des gammes de contraintes à la paroi bien contrôlées sur les surfaces étudiées. Des surfaces modèles ont été choisies pour leurs propriétés physico-chimiques et mécaniques afin d’étudier en détail les interactions entre les souillures et leur substrat.

Un asperseur pour chromatographie sur couche mince a été retenu pour produire des éclaboussures contrôlées car il fournissait des distributions de tailles de gouttes plus reproductibles et plus étroites. La première campagne d’expériences réalisées avec des gouttes d’huile a montré que, suite aux conditions hydrodynamiques complexes près de l’orifice d’entrée du fluide dans la cellule, les modèles analytiques disponibles pour lier le rayon de détachement, la contrainte critique à la paroi (contrainte minimale pour provoquer le détachement des souillures) et la force d’adhésion des souillures dans la cellule à flux radial n’étaient applicables que pour les souillures faiblements adhérentes qui se détachent à des seuils de cisaillement inférieurs à 3 Pa.

Par conséquent, l’écoulement à l’intérieur de la cellule à flux radial a été caractérisé au moyen de la mécanique des fluides numérique sur l’ensemble du régime laminaire et validé expérimentalement. L’étude de l’adhérence d’agrégats de granules d’amidon dans la cellule à flux radial a montré que la conversion du rayon critique de détachement en contrainte critique de cisaillement à la paroi peut être biaisée quand la hauteur des agrégats n’est plus négligeable par rapport à la hauteur du canal d’écoulement et quand l’adhérence est telle que des débits supérieurs aux conditions de l’écoulement rampant sont requis pour détacher la souillure.

L’influence de plusieurs facteurs environnementaux et des propriétés des substrats a ensuite été étudiée pour améliorer la compréhension des mécanismes qui influencent l’encrassement et le nettoyage. En influençant l’étalement des gouttes et la compétition entre les forces capillaires agissant aux interfaces granule-substrat et granule-granule, la mouillabilité du substrat va affecter la forme et le compactage des agrégats formés, l’efficacité des forces de cisaillement lors du nettoyage et, enfin, l’adhérence des particules. Des macromolécules originaires de la suspension de granules sont adsorbées sur les substrats depuis la phase liquide ou charriées par le film lors de sa rétraction et accumulées à l’interface granule-subtrat. Ces macromolécules influencent l’adhérence des agrégats de granules en agissant comme un joint adhésif dont les propriétés semblent être influencées par l’histoire détaillée du séchage des gouttes et de l’exposition des agrégats à l’humidité. Sur les substrats souples, les interactions granule-substrat induisent des contraintes à l’interface qui peuvent conduire à une déformation du substrat, augmentant ainsi appréciablement l’adhérence en favorisant un contact plus intime entre les granules et le substrat.

This PhD thesis is the final result of several years of scientific team working and the people who contributed to it should be acknowledged. However, it is also represents $4^{1/2}$ years of my life during which many things happened and for which I also want to express my gratitude.

First I would like to thank Professor Michael Müller (Institut für Pharmazeutische und Medizinische Chemie der Albert-Ludswig-Universität Freiburg) because without him, I don't think I would ever have undertaken this work. He was the first one to really believe in my technical skills and the first one to let me see it. Actually, he opened the door to this work.

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“Muad'Dib tells us in "A Time of Reflection" that his first collisions with Arrakeen necessities were the true beginnings of his education. He learned then how to pole the sand for its weather, learned the language of the wind's needles stinging his skin, learned how the nose can buzz with sand-itch and how to gather his body's precious moisture around him to guard it and preserve it. As his eyes assumed the blue of the Ibad, he learned the Chakobsa way.”

Frank Herbert, Dune, 1965.

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FOREWORD

This thesis is composed of two parts:

- a literature review divided in two sections, the first being a manuscript on hygiene and surfaces and the second, a manuscript on hydrodynamic flow systems commonly used to assess surface fouling and cleaning (Part I).
- the objectives statement and a brief review of the framework (section 3) followed by a detailed presentation consisting in 5 sections (section 4 to 8) in the form of manuscripts followed by a general discussion and its perspectives (section 9) (Part II).

Part I

- Hygiene and Cleanability: a focus on surfaces. Detry, J.G., Sindic, M. and Deroanne, C., 2009. *Critical Reviews in Food Science and Nutrition, accepted.*
- Hydrodynamic systems used to assess surface fouling, soil adherence and cleaning in laboratory installations. Detry, J.G., Deroanne, C. and Sindic, M., 2009. *BASE, accepted.*

Part II

- Objectives and framework
- Cleanability assessment of model solid surfaces with a radial-flow cell. Detry, J.G., Rouxhet, P.G., Boulangé-Petermann, L., Deroanne, C. and Sindic, M., 2007. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 302, 540-548.
- Laminar flow in radial flow cell with small aspect ratios: numerical and experimental study. Detry, J.G., Deroanne, C., Sindic, M. and Jensen, B.B.B., 2009. *Chemical Engineering Science*, 64, 31-42.
- Flow rate dependency of critical wall shear stress in a radial-flow cell. Detry, J.G., Jensen, B.B.B., Sindic, M. and Deroanne, C., 2009. *Journal of Food Engineering*, 92, 86-99.
- Adherence of starch granule aggregates on different materials: experimental factors and physico-chemical mechanisms. Detry, J.G., Sindic, M., Servais, M.J., Adriaensen, Y., Deroanne, C. and Rouxhet, P.G., 2009. *Journal of Colloid and Interface Science, in preparation*.
- Adherence of starch granule aggregates to substrates: comparison between soft and hard hydrophobic substrates. *Project of publication*.
- General discussion and perspectives

PART I (LITERATURE SURVEY) – SECTION 1

HYGIENE AND CLEANABILITY: A FOCUS ON SURFACES

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PART I (LITERATURE SURVEY) – SECTION 2

HYDRODYNAMIC SYSTEMS USED TO ASSESS SURFACE FOULING, SOIL ADHERENCE AND CLEANING IN LABORATORY INSTALLATIONS

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SECTION 2 – HYDRODYNAMIC SYSTEMS USED TO ASSESS SURFACE FOULING, SOIL ADHERENCE AND CLEANING IN LABORATORY INSTALLATIONS

ABSTRACT

Five hydrodynamic systems are presented in this short review: the parallel plate flow cell, the impinging jet, the radial flow cell, the rotating disk and fluid dynamic gauging. These systems are of particular relevance to study surface fouling, surface cleaning or adhesion onto solid surfaces in laboratory environment. The key features of their hydrodynamics are given as well as their practical advantages and drawbacks. Examples of applications fields are also listed.

Keywords:
Cleaning, adhesion strength, fouling, wall shear stress, surfaces

2.1 INTRODUCTION

The ability of a surface to reduce the adhesion of microorganisms (or other contaminants), to inhibit the formation of deposits or to release the adherent deposits and microorganisms is something essential for a wide field of applications like ship hulls, medical implants, dental enamel, pipelines, surgical instruments, buildings, food and pharmaceutical processing… (Changani et al., 1997; Bakker et al., 2003b; Bansal et al., 2006; Liu et al., 2006). Recently, the modification of surfaces or the elaboration of new coatings has been shown to reduce the attachment of bacteria (Zhao et al., 2005a), the formation of scales (Zhao et al., 2005b; Rosmaninho et al., 2006) or the adherence of food deposits (Saikhwan et al., 2006). Furthermore, the modification of surfaces is attracting considerable attention thanks to the advent of affordable tailored coatings and the capability for applying new surface modification technologies to the scale of equipment parts.

Assessing the ability of a surface to reduce adhesion or to easily release contaminants is critical to improve the understanding of adhesion mechanisms, to identify the critical surface features influencing it or to compare different surfaces in well-controlled conditions. Furthermore, attempts to relate surface engineering to a given application do not always allow real time and *in situ* observation in spite of the need to consider environmental variables such as flow (Jensen et al., 2004), heat and mass transfer (Rosmaninho et al., 2007a), passage of an air-liquid interface (Gómez Suárez et al., 2001a), presence of chemicals or surfactants (Joscelyne et al., 1997; Morison et al., 2002; Chateau et al., 2004) etc.

Flow chambers and other similar hydrodynamics devices reviewed in this work proved to be valuable tools to take those environmental variables into consideration while allowing easy observation, easy set-up as well as standardization and comparisons between laboratories. Their simple geometry allows the generation of well-controlled, reproducible flow conditions for which the analytical solutions of the Navier-Stokes equations and the convective diffusion equation are often available (Elimelech, 1994). Most of them are also easy to design and can be used as modules to constitute test rigs together with a pump, a heating device, measuring devices… They can also be adapted to various sizes of samples as long as similarity is preserved, depending on the representativeness of the studied surfaces and on cost or technological limitations.

2.2 THE PARALLEL PLATE FLOW CELL

The test part of the parallel plate flow cell or parallel plate flow chamber is constituted of a bottom plate and an upper plate (one of the two or both being the sample surface) separated by a distance *h* and forming a rectangular flow channel of width *w* (Figure 2.1). It is generally used to generate a laminar shearing Poiseuille flow parallel to the sample surface though its adaptation to generate fully-developed turbulent flow conditions was also reported to expose the samples to the hydrodynamic conditions encountered at the surface of ship hulls (Schultz et al., 2000; Schultz et al., 2003).

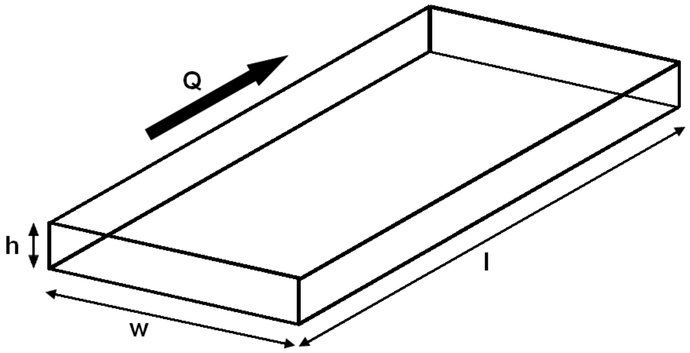

Figure 2.1: Scheme of the flow channel in the parallel plate flow cell. The direction of the flow is represented by the thick arrow and the sample generally constitutes the lower surface of the parallelepiped. Symbols: *w* is the channel width, *h* the channel height, *l* the channel length and *Q* the flow rate.

The flow regime in the test section can be deduced from the Reynolds number, which depends on the properties on the fluid, the flow rate and the dimensions of the flow cell. The Reynolds number in this flow channel is given by (Bakker et al., 2003b):

$$\mathrm{Re} = \frac{\rho \cdot Q}{\eta \cdot (w + h)} \qquad (2.1)$$

where Q is the volumetric flow rate ($m^3.s^{-1}$), h the separating distance between the upper and the lower plate (m), w the width of the flow channel (m) and η the dynamic viscosity ($kg.m^{-1}.s^{-1}$).

Two-dimensional steady and laminar flow can be assumed for Re ≤ 2000 (Bos et al., 1999). However, whatever the design of the system, fully developed laminar unidirectional Poiseuille flow will only be established at a certain distance from the inlet of the rectangular test section. This distance is called the establishment length, L_e (in m). To reduce the establishment length, the inlet should be followed by a gradual expansion (diffuser) before the flow channel and in line with it. The outlet should be preceded by a similar gradual contraction in line with the flow channel (Bakker et al., 2003b). In addition, the dimensions of the flow channel, the flow rate and the nature of the fluid will influence the value of L_e (Lorthois et al., 2001; Mercier-Bonin et al., 2004; Busscher et al., 2006):

$$L_e = constant \times h \times Re \qquad (2.2)$$

The value of the constant varies from 0.013 (Busscher et al., 2006) to 0.273 (Lorthois et al., 2001) depending on the flow cell design. After this length, the flow can be considered as fully developed and the shear rate at the surface of the sample ($\dot{\gamma}$, in s^{-1}) can be considered constant throughout the whole test section. This shear rate is given by (Bakker et al., 2003b):

$$\dot{\gamma} = \frac{3 \cdot Q}{2w(h/2)^2} \qquad (2.3)$$

This expression defines the velocity gradient perpendicular to the wall. For Newtonian fluids like water, multiplying the wall shear rate by the dynamic viscosity of the fluid gives the wall shear stress which is the hydrodynamic force per unit surface area exposed to the flow. The wall shear stress (τ, in $N.m^{-2}$) is parallel to the wall and is expressed by:

$$\tau = \eta\dot{\gamma} \qquad (2.4)$$

The multiplication of the shear stress by the area of the adhering soil, microorganism or particle exposed to the flow would give the hydrodynamic drag force exerted on it (Busscher et al., 2006).

For bacteria, application of successive periods of low and high shear stress was also recently performed for the determination of a “critical wall shear stress” with the parallel plate flow cell. A bacterial suspension is circulated at low wall shear stress for 30 min followed by removal periods of 30 min at increasing wall shear stress. The critical wall shear stress is the wall shear stress at which bacterial attachment and detachment balance each other or, in other words, when the change in number of adhering bacteria was stabilized to zero after application of the higher shear stress. A critical force can then be deduced from this shear stress value (Nejadnik et al., 2008).

In the case of a single spherical particle, the flow around the particle is purely viscous when $Re_p << 1$. In the case of a rigid spherical particle in contact with a wall and exposed to slow linear shear flow, the particle will be exposed to hydrodynamic drag (F_D), torque (M) and lift (F_L) which can be related to the wall shear stress according to (Brooks et al., 1996; Mercier-Bonin et al., 2004):

$$F_D = 6C_D \pi\tau R^2 = 10.26\pi\tau R^2 \qquad (2.5)$$

$$M = 4C_M \pi\tau R^3 = 3.8\pi\tau R^3 \qquad (2.6)$$

$$F_L = 9.257\tau R^2 \operatorname{Re}_p \qquad (2.7)$$

where R is the particle radius, C_D the drag coefficient and C_M the moment coefficient. Lift is generally assumed as negligible in the theory of detachment of particles as long as the flow around the particle is purely viscous. This can be expressed by (Hubbe, 1984):

$$\operatorname{Re}_p = \frac{r_p U \rho}{\eta} << 1 \qquad (2.8)$$

where Re_p is the particle Reynolds number, U is the average flow velocity around the particle (m/s) and r_p is the particle radius (m). Equations (2.5) and (2.6) are valid as long as Equation (2.8) is satisfied and the relation between the hydrodynamic drag force and the wall shear stress can be assumed independent of the flow rate in these conditions (Cardot et al., 2001; Lorthois et al., 2001; Detry et al., 2009b).

However, if Re_p becomes larger than 0.05, inertial effects are present in the flow close to the particle and cannot be further neglected. This will result in a decrease of the drag and moment coefficients which becomes increasingly significant as Re_p increases and in a decreasing proportionality between the wall shear stress and the force exerted on the particle (Hubbe, 1984).

The main advantage of the parallel plate flow cell is the ability to generate a simple flow of constant wall shear stress along a sample surface. Its geometry and the nature of the flow make it easy to place as a “module” into a closed circuit. Furthermore, the use of a transparent material for the upper wall of the channel allows *in situ* observation. One of the main drawbacks of the system is its design which lacks of flexibility. Indeed, once the geometry of the system built, it is impossible to change it afterwards. Care should thus be taken to think about the wall shear stress range that it will be possible to generate, about the required Reynolds number and flow rates (pump) and about the dimensions of the system. As shown by Equations 2.2, 2.3 and 2.4, if high wall shear stresses are required for an application (like the removal of strongly adhering contaminants), the channel height will generally have to be ≤ 200 µm in order to keep with reasonable flow rates and sample sizes (Guillemot et al., 2006). A width-to-height ratio larger than five should be kept in order to exclude side-wall effects (Bos et al., 1999). The reduction of the flow channel height will also have an influence on the size of the contaminants. At low flow rates for instance, an adherent particle should have a diameter ≤ 1/15 h and should be separated from its neighbors by more than five times the particle radius in order to avoid disturbing the flow and satisfy Equation (2.8) (Brooks et al., 1996). Another disadvantage of the parallel plate flow cell is that the flow arrives parallel to the sample surface, meaning that, if the sample is placed in a recess, there should be no misalignment between the sample and the flow cell (Schultz et al., 2000). An alternative would be to replace the whole lower plate of the parallel plate flow cell by the sample (Mercier-Bonin et al., 2004) but this would result in bigger sample sizes, which is not always feasible. To conclude, the parallel plate flow cell seems to be more suitable for weak-adherence systems with small and well-separated adhering soils (oil droplets, particles or microorganisms). Some examples of studies realized with the parallel plate flow cell are given in Table 2.1.

2.3 IMPINGING FLOW SYSTEMS

2.3.1 The impinging jet

The impinging jet is a system widely used to study heat and mass transfer for various applications like annealing of metal and plastic sheets, tempering of glass plates, drying of various materials, cooling of heated components in engines, deicing of aircraft systems… (Tu et al., 1996; Yapici et al., 1999) Examples of applications in relation with the detachment of contaminants from solid surfaces are listed in Table 2.1.

The system consists in a jet flow of liquid exiting through a nozzle of radius r_i, perpendicular to the surface and situated at a distance h from it. A stagnation point is present on the surface where the nozzle axis crosses it. The flow domain of an impinging jet can be divided in three regions for both laminar and turbulent regimes (Deshpande et al., 1982; Yapici et al., 1999): (i) the free jet region where the fluid is not influenced by the surface and where the dominant velocity component is axial; (ii) the impingement region where the fluid impacts the surface and where the dominant velocity component changes from axial to radial; (iii) the wall jet region in which the dominant velocity is radial. As the radial position taken from the center of the inlet nozzle increases, the radial velocity and thus the wall shear stress both decrease (Figure 2.2).

Table 2.1: Some examples of application of the hydrodynamic systems presented in this work.

System	Study	Contaminants	Surfaces	Reference
Parallel plate flow cell	Bacterial adhesion	*Marinobacter hydrocarbonoclasticus*, *Psychrobacter sp.* or *Halomonas pacifica*	Glass	Bakker et al., 2002
			Glass or polyurethane coatings	Bakker et al., 2003a
	Yeast detachment	*Saccharomyces cerevisiae*	Glass	Mercier-Bonin et al., 2004
			Stainless steel	Guillemot et al., 2006
	Mammalian cell detachment	Endothelial cells	Collagen-coated or fibronectin-coated polystyrene	Bouafsoun et al., 2006
	Particle removal	Ammonium fluorescein particles	Glass	Phares et al., 2000a
		Glass beads	Glass	Cardot et al., 2001
		Fibrin-coated latex particles	Fibrin-coated glass	Lorthois et al., 2001
	Effect of an air/liquid interface on detachment	*Streptococcus oralis, Streptococcus sobrinus, Pseudomonas aeruginosa, Actinomyces naeslundii* or *Bacteroides fragilis*	Quartz or hydrophobic coated glass	Gómez Suárez et al., 2001a
		Polystyrene particles	Quartz or hydrophobic coated glass	Gómez Suárez et al., 2001b
	Oil drop removal	Sunflower oil	Several polymeric coatings	Boulangé-Petermann et al., 2003
			Stainless steel of various finishes, hydrophobic or hydrophilic coated stainless steel	Boulangé-Petermann et al., 2006

			Stainless steel or polysiloxane-coated stainless steel	Thoreau et al., 2006
	Antifouling potential of marine coatings	*Ulva linza* zoospores	Micropatterned PDMS	Hoipkemeier-Wilson et al., 2004
		Ulva linza zoospores	Micropatterned PDMS	Carman et al., 2006
Impinging jet	Particle removal	Polystyrene particles	Glass	Smedley et al., 1999
		Polystyrene or ammonium fluorescein particles	Glass	Phares et al., 2000a
	Cell and bacteria detachment	3T3 fibroblasts, L929 fibroblasts or *Staphylococcus aureus*	Titanium, stainless steel or polyethylene terephtalate	Bundy et al., 2001
	Bacteria detachment	*Pseudomonas stutzeri*	Glass or indium tin oxide coated glass	Bayoudh et al., 2005
	Mammalian cell detachment	Endothelial cells	Collagen-coated or fibronectin-coated polystyrene	Bouafsoun et al., 2006
	Erosion of endothelium	/	Canine endothelium	Vaishnav et al., 1983
Radial-flow cell	Bacterial adhesion	*Marinobacter hydrocarbonoclasticus*, *Psychrobacter sp.* or *Halomonas pacifica*	Glass	Bakker et al., 2002
	Bacteria detachment	Coccoid and rod shaped cells	Pyrex glass, plate glass, siliconized glass or stainless steel	Fowler et al., 1980
		Pseudomonas fluorescens	Stainless steel	Fryer et al., 1985
	Receptor-ligand interactions	Receptor coated latex beads	Ligand-coated glass	Cozens-Roberts et al., 1990
	Mammalian cells	3T3 murine fibroblasts	Self assembled monolayers of	Goldstein et al. 1998

	detachment		dodecane thiolate	
	Bacterial spores detachment	*Bacillus cereus spores*	Stainless steel or polypropylene	Klavenes et al., 2002
	Yeast and bacteria detachment	*Saccharomyces cerevisiae* or *Dictyostelium discoideum*	Glass	Décavé et al., 2005
	Particle deposition	Polystyrene latex particles	Mica sheets	Adamczyk et al., 2001
	Surfactant-mediated particle attachment and release	Bare or β-lactoglobulin-coated polystyrene latex particles	Bare or β-lactoglobulin-coated indium tin oxide	Joscelyne et al., 1997
	Oil drop removal	Sunflower oil	Glass, stainless steel, PTFE or polystyrene	Detry et al., 2007
	Starch particles removal	Waxy corn starch granules	Glass, stainless steel, PTFE or polystyrene	Detry et al., 2009b
	Dairy soil cleaning	EHEDG soil	Stainless steel	Jensen et al., 2004
Fluid dynamic gauging	Thickness of soft deposits	Whey protein concentrate Butter	Stainless steel	Tuladhar et al., 2000
	Deposit strength	Tomato paste	Stainless steel	Chew et al., 2004
	Film development in duct flow	Ice film	Stainless steel	Tuladhar et al., 2003
	Polymer swelling and cleaning	Polstyrene co-polymers	Stainless steel	Chew et al., 2005 Chew et al., 2006
	Deposit adhesion strength	Tomato paste	Hydrophilic or hydrophobic coated stainless steel	Saikhwan et al., 2006

Rotating disk	Cleaning	Skimmed milk or whey protein concentrate deposits	Stainless steel	Morison et al., 2002
		Mixture of oils with particles	Polyurethane	Chateau et al., 2004
	Fouling	Calcium phosphate	Modified stainless steel surfaces	Rosmaninho et al., 2006 Rosmaninho et al., 2007b
			Titanium nitride coatings	Rosmaninho et al., 2007a
		Calcium phosphate and proteins	Modified stainless steel surfaces	Rosmaninho et al., 2008

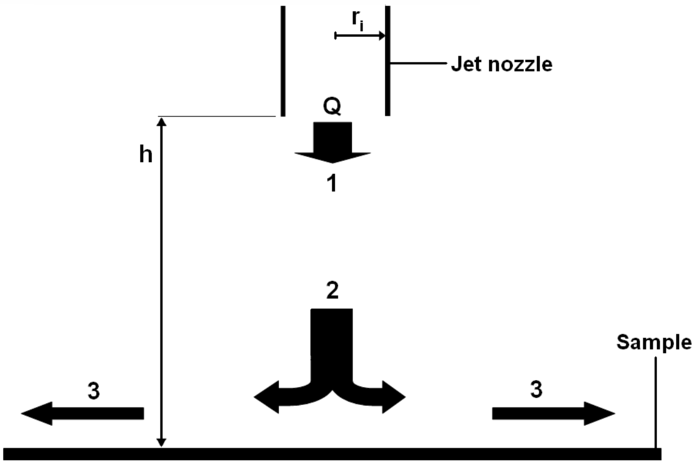

Figure 2.2: Representation of the impinging jet. The main flow path and velocity directions are represented by the thick arrows. (1) Free jet region, (2) impingement region, (3) wall jet region. Symbols: r_i is the radius of the jet nozzle, h is the jet-to-sample separation distance and Q is the flow rate.

A radial gradient of decreasing wall shear stress is thus generated at the impinged surface. The magnitude of the wall shear stress distribution depends on the nozzle-surface distance *h*, the nozzle diameter *r* and the Reynolds number of the fluid in the nozzle. Numerical solutions of the wall shear stress distribution are available for laminar flow (Deshpande et al., 1982; Deshpande et al., 1983) and turbulent flow (Tu et al., 1996; Yapici et al., 1999; Phares et al., 2000b). In the case of liquid jets, both the studied surface and the nozzle must be immersed. This is obviously not the case when the impinging fluid is air.

Soiled surfaces can thus be placed perpendicularly from the nozzle at a distance *h*. The impinging fluid will exert a hydrodynamic force on the adherent soils which will be submitted to a continuous range of shear forces in one experiment. If the hydrodynamic force exerted by the flow exceeds the adhesion force, detachment of the soils will occur near the inlet up to radial positions where the hydrodynamic drag force will be too weak to induce detachment. Then, the nozzle is removed and the radial position up to which removal occurs is measured. This radial position can be converted in wall shear stress with the numerically computed wall shear stress distributions. The wall shear stress associated with removal can then be related to the adhesion force of the soil (Phares et al., 2000b; Phares et al., 2000a).

The main advantage of the impinging jet is that it allows the adherent soils to be submitted to a continuous range of shear forces in a single experiment with respect to applying a sequence of shear rates using the parallel plate flow chamber. It is also very flexible as the nozzle can normally be adapted to a wide range of sample sizes, as long as this size largely exceeds the nozzle diameter (Bitziou et al., 2006). However, *in situ* observations are impossible if the substrate is not transparent and if a reverse observation setup (allowing observation from under the jet) cannot be mounted. Another inconvenient of the system may be the need of numerical computations to find the wall shear stress which may still be unaffordable for small laboratories. Lastly, the system cannot be easily adapted as a module in a test rig. This inconvenience can however be overcome by confining the jet flow.

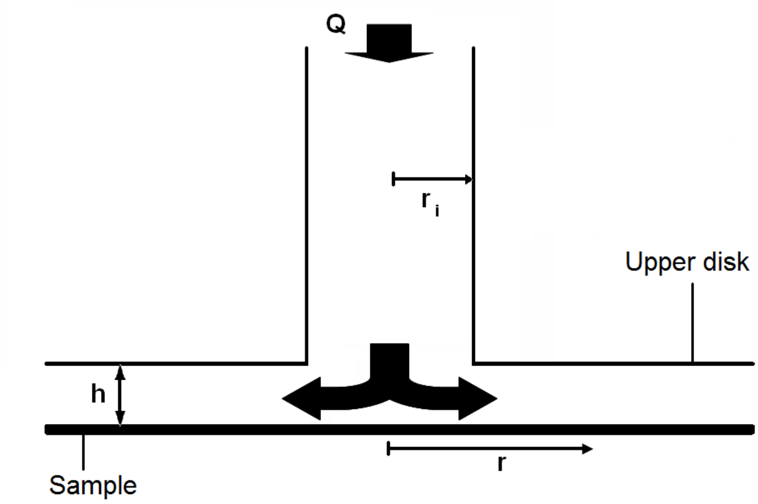

Figure 2.3: Schematic of the radial-flow cell. The flow (thick arrows) impinges on the sample and exits radially outwards, confined between the sample and the upper disk. Symbols: r_i is the inlet radius, h is the disk spacing, r is the radial position and Q is the flow rate.

2.3.2 The radial-flow cell

The radial-flow cell is also known as confined impinging jet, radial-flow chamber or stagnation-point flow chamber. Its use to study biofilm removal was first reported by Fowler and McKay (1980). The device consists of two parallel disks with a narrow spacing in-between (typically, the ratio between the disc spacing h and the inlet radius r_i is << 1). A fluid is pumped through the center of one disk, impinges on the surface of interest (a stagnation point is present on the surface at the intersection with the inlet axis) and flows radially outward between the disks (Figure 2.3). As the flow duct cross-sectional area increases with the radius, the linear fluid velocity and hence the shear stress near the surface decrease radially across the disk. As in the case of free impinging jets, the soils adhering to the surface are submitted to a continuous range of shear forces in one experiment. When the soils (typically cells or particles) are submitted to the shear flow and if the mechanical action of the fluid is sufficient, detachment will occur near the inlet (higher shear stress) up to radial positions where only 50 % of the soils are detached. If the size distribution of the soils is symmetrical, this radial position is called the critical detachment radius and can be associated to a critical wall shear stress, which is related to the mean adhesion force of the soils (Cozens-Roberts et al., 1990; Lorthois et al., 2001).

The confinement of the flow allows an easier fluid recovery and eases the close recirculation of the shearing fluid by comparison to conventional jet impingement (Fowler et al., 1980; Jensen et al., 2004). However, *in situ* observations are again not possible if the sample is not transparent. The system will also be less flexible with respect to the sample size and disc spacing adjustments when included in a closed circuit. Similarly to the parallel plate flow cell, the radial-flow cell seems restricted to small, weakly adherent and well-separated adhering soils (oil droplets, particles or microorganisms) to ensure well characterized hydrodynamics (Brooks et al., 1996).

Analytical solutions are available to compute the wall shear stress at the sample surface for creeping flow and fully turbulent flow. The flow regime in the radial-flow cell is characterized by the local Reynolds number across the disk and the Reynolds number in the inlet pipe. Both are respectively given by:

$$\mathrm{Re}_r = \frac{\rho.Q}{\pi.\eta.r} \qquad (2.9)$$

$$\mathrm{Re}_{inlet} = \frac{2.\rho.Q}{\pi.\eta.r_i} \qquad (2.10)$$

where r is the radial position and r_i the inlet radius, both expressed in meter.

When the disc spacing is narrow and the Reynolds number low enough, the flow between the discs is laminar. This is generally considered for $\mathrm{Re}_{\mathrm{inlet}} < 2000$ (Moller, 1963), even if time periodic or transient unsteady flow structures are reported for inlet Reynolds number varying between 460 and 4000 at aspect ratios $e \geq 2$ ($e = h / r_i$) (Nakabayashi et al., 2002). These instabilities result from inertial effects. They appear above a certain Reynolds number and can lead to the appearance of local 3D flow structures and to the rotation of the global flow pattern around the inlet axis. They tend to disappear when the aspect ratio is reduced (Hsieh et al., 2006). For $\mathrm{Re}_{\mathrm{inlet}} > 4000$, the flow can be considered turbulent in the regions between the discs where the inlet geometry influences the flow (Nakabayashi et al., 2002). At flow rates high enough to produce chaotic flow or even turbulence at low radius, the deceleration of the flow with an increasing distance from the inlet gives a decreasing local Reynolds number and a possible transition to laminar flow (Kreith, 1965).

Forty years ago, Moller developed analytical solutions to predict the wall shear stress between two parallel disks for both laminar and turbulent ideal diverging flow at any radial position (Moller, 1963):

$$\tau_{laminar} = \frac{3.\eta.Q}{\pi.r.h^2} \qquad (2.11)$$

$$\tau_{turbulent} = 0.0288\rho U^2 \operatorname{Re}_r^{-0,2} \qquad (2.12)$$

where U is the mean velocity (m.s^{-1}).

Fryer et al. (1985) showed that the equation for ideal radial laminar diverging flow (Equation 2.11) gives a good approximation of wall shear stress only if the inertial forces are small with respect to the viscous forces or, in other words, for radial positions where the effect of the inlet geometry is no longer present. This corresponds to radial positions satisfying (Fryer et al., 1985; Detry et al., 2009a):

$$\frac{\rho.Q}{24.\pi.r.\eta}.\frac{h}{r} \leq 0.145 \qquad (2.13)$$

For the other radial positions, Equation 2.11 is not accurate as a result of the complex hydrodynamics induced by the inlet geometry (Detry et al., 2007) and a number of recirculation zones can be present near the inlet (Goldstein et al., 1997; Chatterjee, 2000; Hsieh et al., 2005; Hsieh et al., 2006). Then, a numerical solution is needed to compute the critical wall shear stress from the critical detachment radius (Goldstein et al., 1997; Goldstein et al., 1998; Jensen et al., 2004; Detry et al., 2009a). Examples of applications of the radial-flow cell are given in Table 2.1.

An interesting alternative to the diverging flow is the use of converging flow, the fluid being sucked into the inlet (Goldstein et al., 1998). In this case, the recirculation zones are present in the inlet pipe and not in the channel where the measurements are performed. Inertial corrections are still needed but the range of shear stresses that can accurately be estimated by Equation 2.12 for laminar converging flow at an aspect ratio (h/r_i) of 0.2 was reported to be more than twice that estimated with diverging flow (Goldstein et al., 1998).

2.4 FLUID DYNAMIC GAUGING

Basically, fluid dynamic gauging can be compared to the application of converging flow to the free impinging jet system. The technique consists in inducing a flow through a nozzle of diameter d_t close and normal to the surface of a deposit (Figure 2.4). The fluid is sucked from the quasi-stagnant surroundings through a siphon tube. A micrometer controls the vertical position of the nozzle and the distance h between the nozzle and the deposit. If that distance is small (h/d_t ~ 0.25), the flow rate passing through the nozzle will be very sensitive to the spacing between the deposit and the nozzle and the measure of the mass flow rate will allow the computation of the nozzle-deposit distance and hence of the deposit thickness (Tuladhar et al., 2000; Chew et al., 2004a).

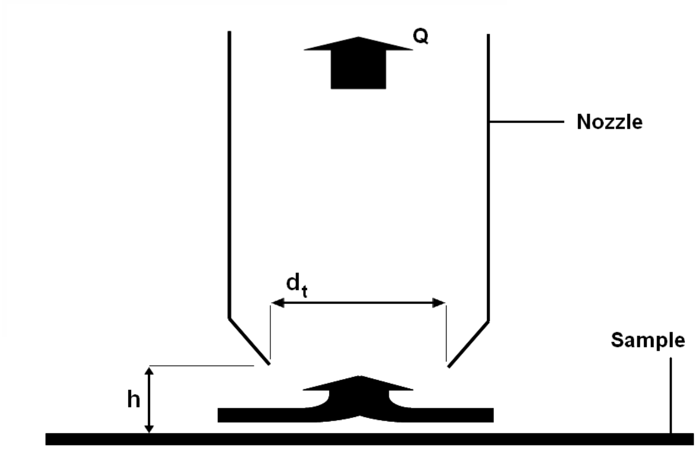

Figure 2.4: The fluid dynamic gauging. Symbols: d_t is the nozzle diameter, Q is the flow rate and h is the distance between the sample or the deposit and the nozzle.

Fluid dynamic gauging thus enables the on-line measurement of the thickness of soft deposit layers (e.g.: whey proteins, tomato paste) adhering to immersed solid surfaces. The build-up of a deposit under defined bulk conditions or the swelling of a deposit during the cleaning process can be measured with this technique. When the shear stress distribution exerted by the flow on the deposit is known, the shearing yield strength of the deposit and its deformation characteristics can be measured as well (Chew et al., 2004b). Several applications of fluid dynamic gauging are presented in Table 2.1.

This ability to measure on-line the thickness of continuous deposits as well as their mechanical properties is the main advantage of fluid dynamic gauging. Indeed, the technique is very flexible in terms of sample sizes and it can be applied to deposits of any thicknesses. Furthermore, the technique can also be mounted in closed test rigs (Tuladhar et al., 2003; Gu et al., 2007). *In situ* observation is not easy with this technique but it is compensated by the ability of performing on-line monitoring. When the adherence of tomato paste on modified surfaces was studied, significantly different behaviors were observed between the surfaces depending on the deposit-substrate interactions. The adhesive strength of tomato paste deposits was better characterized by the hydrodynamic suction stress normal to the surface than by the wall shear stress (Saikhwan et al., 2006). Though this suction stress is particularly interesting to characterize the deposit properties, it is generally not encountered in real equipment in opposition to wall shear stress, which makes it more difficult to transfer the data obtained with fluid dynamic gauging to real equipment if the soil adhesion strength has to be estimated.

2.5 THE ROTATING DISK

The rotating disk or spinning disk is a disk that rotates in a fluid at controlled speed. The sample surface is placed on the disk, can be heated or not and placed in a fouling or in a cleaning solution. In this system, the fluid acquires a rotational motion when it approaches the disk surface. This rotational motion forces it to exit radially (Figure 2.5). For steady laminar regime, the thicknesses of the hydrodynamic and diffusion boundary layers are constant over the whole surface investigated for a given rotational speed (Elimelech, 1994). Again, the flow regime at the disk surface is given by the Reynolds number (Levich, 1962; Schlichting et al., 2000):

$$\mathrm{Re} = \frac{r_d^2 \cdot \omega \cdot \rho}{\eta} \qquad (2.14)$$

where r_d is the disk radius (m) and ω the rotational speed of the disk (rad.s^{-1}). The regime at the surface of the disk will remain laminar as long as $\mathrm{Re} < 3 \times 10^5$.

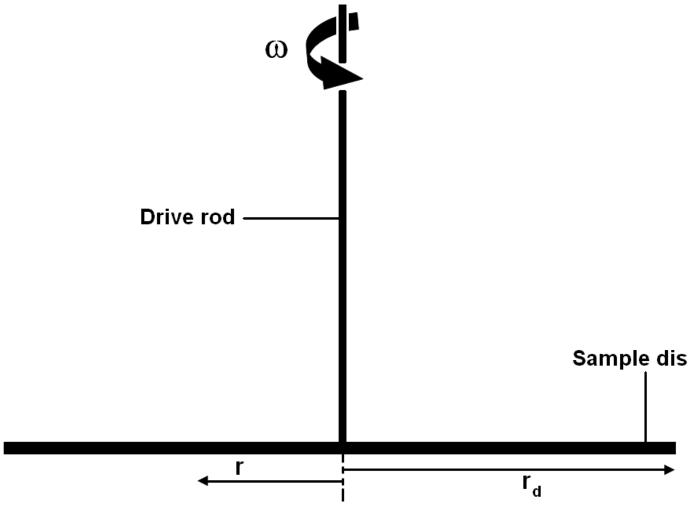

Figure 2.5: Schematic of a rotating disk apparatus. Symbols: ω is the rotation speed, r is the radial position, and r_d is the diameter of the sample. The rod inducing the rotation movement is placed in the center of the sample disk, on the opposite side of the studied surface.

The steadiness of the hydrodynamic and diffusion boundary layers in the laminar regime allows the determination of a well-defined analytical solution to compute the mass transfer to the sample surface (Levich, 1962; Morison et al., 2002):

$$J = \left[0.62\frac{D}{2r_d}(\mathrm{Re})^{1/2}(Sc)^{1/3}\right]\cdot(C_b - C_i) = 0.62D^{2/3}\left(\frac{\eta}{\rho}\right)^{-1/6}\omega^{1/2}(C_b - C_i) \qquad (2.15)$$

where J is the mass transfer flux through the boundary layer (kg.m^{-2}.s^{-1}), D is the bulk diffusion coefficient, Re the Reynolds number, $Sc = \eta/(\rho D)$ the Schmidt number, η the dynamic viscosity of the solution (kg.m^{-1}.s^{-1}), ρ the density of the solution (kg.m^{-3}), C_b the bulk concentration (kg.m^{-3}) and C_i the interfacial concentration (kg.m^{-3}). Methodologies and explanation for the determination of the interfacial concentration can be found in Hunek and Cussler (2002), Morison and Thorpe (2002) and Rosmaninho et al. (2007a).

Under these conditions, the wall shear stress at the sample surface varies linearly with the radial position according to (García et al., 1997):

$$\tau = 0.8r(\rho\eta\omega^3)^{1/2} \qquad (2.16)$$

where τ is the wall shear stress (N.m^{-2}) and r is the radial position (m). In this case, the wall shear stress increases linearly with the radial position conversely to impinging jet flows.

The major advantage of the rotating disk is the ability to produce a linear range of shear stress at the surface of a sample in a single experiment with uniform chemical conditions over the whole sample surface (García et al., 1997). This characteristic makes of the rotating disk a very interesting tool to study fouling and cleaning although it is not frequently considered for such applications. The system is also flexible with respect to the size of the samples and the height of the deposits but is however difficult to include as module in a test rig. Examples of applications of the rotating disk to surface fouling and cleaning are given in Table 2.1.

2.6 APPLICATION TO REAL EQUIPMENT

All above mentioned laboratory systems can be used to generate very useful information required to study cleaning and fouling in controlled conditions. Such information may be the mechanisms of action of a chemical in the breakdown of a soil, the surface parameters influencing the cleaning process, the adhesion strength of a soil to various substrates, the effect of soil ageing on its adherence or the determination of the adequate surface modification that will mitigate fouling, reduce soil adhesion or facilitate soil removal.

However, the experimental data are generally obtained for planar sample surfaces and, except maybe for wall panels, they cannot be easily applied “as generated” (raw data) to equipment of complex geometry in order to predict how the equipment will be cleaned. This was well illustrated by Jensen and Friis (2004; 2005) who tried to relate the critical wall shear stress obtained in a radial-flow cell assay with CFD-computed (computed using computational fluid dynamics) wall shear stresses to predict the cleanability of a mix-proof valve as a function of wall shear stress only. The study revealed that complex phenomena such as fluid exchange at the vicinity of the surface were influencing soil removal to a non negligible extent and that the critical wall shear stress given by the radial flow cell assay was certainly very useful but not totally satisfactory to predict the cleanability of equipment parts though both cleaning procedures were performed under turbulent flow regime. Indeed, other phenomena associated with flow like wall shear stress oscillations (Lelièvre et al., 2002), pulsating flow (Gillham et al., 2000) and fluid exchange (Jensen, 2003) have been suggested to play a non negligible role in cleaning.

Furthermore, several problems arise when CFD codes are applied to model cleaning processes. Adequate meshes and turbulence models must be selected because the equipment geometry can be very complex and cleaning is normally performed in the turbulent regime. In this regime, modeling the fluid flow to predict the cleanability of equipment parts requires precise information on the conditions at the walls which differ significantly from the conditions in the bulk (Schlitling, 2000) and wall functions are used to bridge the turbulent flow with the thin viscous layer near the surface. The mesh near the wall must thus be conceived carefully and significant errors may occur depending on the choice of the turbulent model and of the wall function (ERCOFTAC, 2000; Jensen, 2003). Therefore, the numerical results should always be validated with experimental data, which acquisition can be time consuming, difficult to implement or subject to imprecision depending on the technique used (Lelièvre et al., 2002; Jensen et al., 2005; Kipp et al., 2008).

For these reasons and because modeling can still only give trends on how an equipment will be cleaned (Jensen et al., 2007), the cleanability of closed-equipment conceived to be used on food-processing lines has to be assessed on industrial pilot rigs (Bénézech et al., 2002) with standardized procedures such as the ones developed by the European Hygienic Equipment Design Group (EHEDG, www.ehedg.org) to assess the in-place cleanability of food processing equipment. Several methods already exist and can be used as a basis to compare the cleanability of existing equipment or to assess the hygienic design of new equipment (Hofmann et al., 2006; EHEDG, 2007). The coupling of CFD to the results of the EHEDG test can definitely be used to better understand why certain areas are more difficult to clean and how future equipment should be designed in order to avoid the presence of difficult-to-clean areas (Jensen et al., 2007).

2.7 CONCLUSIONS

Five different hydrodynamic systems have been reviewed. Each presents advantages and disadvantages. It is impossible to recommend the use of only one of them as their suitability will depend on the application under consideration. For instance, the parallel plate flow cell is well suited for weakly adherent cells, oil drops or particles, *in situ* observation and for the application of a defined wall shear stress; the radial-flow cell or the impinging jet are more suited for the application of a range of shear forces and for the study of adhesion and adherence in more dynamic conditions (like to study the effect of surfactants on the reduction of the adhesion force of soils); fluid dynamic gauging is particularly adapted to study the formation of continuous deposits, their mechanical properties and their swelling under the action of cleaning chemicals; the rotating disk allows the generation of well-controlled mass transfer conditions to study fouling and cleaning.

However, as useful as these devices are to better understand the soil-substrate-bulk interactions, their use remains limited to the first phases of research and development. Other tools such as mechanical testing, *in situ* experiments or modeling are still subsequently needed to complete the information gained with these simple hydrodynamic devices, in order to reach the commercial application of new surfaces or materials.

2.8 ACKNOWLEDGEMENTS

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PART II – SECTION 3

OBJECTIVES AND SCOPE

SECTION 3 – OBJECTIVES AND SCOPE

3.1 OBJECTIVES

As introduced by the literature review, this doctoral research is in line with the concerns of adhering soils and soil removal which draw the ever growing attention of engineers and scientists from many industrial sectors. The work presented here was realized within the frame of the SMARTNET Project which aimed at developing coatings to improve the cleanability of stainless steel, targeting open surface applications.

For the development of such surfaces, the use of a relevant (and sometimes very specific) methodology to the targeted application coupled with a good and representative soiling procedure is essential and provides reliable comparisons with pre-existent reference surfaces.

The first objective of this work was thus the development of a methodology to assess the ability of various solid surfaces to be cleaned (cleanability) after a standardized soiling procedure. At the same time, the effects of experimental conditions and of the surface physico-chemical properties on soil adherence were investigated to better understand the dynamic and the mechanisms of soil/substrate interactions. This joint strategy provided relevant scientific knowledge and cleanability assessment techniques, essential for the development of easy-to-clean surfaces.

The main lines of the present work were therefore:

- to transfer the knowledge developed on the parallel plate flow cell at the Research Center of Isbergues to the radial-flow cell,
- to characterize the flow inside the radial-flow cell,
- to better understand
    - the relation between critical wall shear stress and soil adherence in the radial-flow cell,
    - the mechanisms related to soil spreading and organization on a surface after spraying,
    - the mechanisms of soil adherence, soil-substrate interactions and how the properties of a substrate condition its cleanability.

3.2 SCOPE

Throughout this thesis, the radial-flow cell was selected to study the adherence and removal mechanisms of different soils due to its ability to generate well-controlled wall shear stress distributions on the investigated surfaces. Four model surfaces were initially selected for their different physico-chemical properties which were expected to influence the ease of soil removal. Glass was chosen for its hydrophilic character, stainless steel for its ubiquity in food industries, polystyrene as hydrophobic polymer and polytetrafluoroethylene for its very hydrophobic properties. In order to better understand the cleaning mechanisms associated to different surfaces in contact with different soils, the interactions between the soils and the surfaces as well as the relevant flow parameters affecting those interactions were studied in detail in the next 5 sections.

Section 4 (*"Cleanability assessment of model solid surfaces with a radial-flow cell"*) presents trials performed with a known soil-cleaning fluid combination (refined sunflower oil – Mr. Proper®) initially used at the Research Center of Isbergues with the parallel plate flow cell. A soiling procedure was developed to produce controlled spatters and mimic the spatters which occur during food product handling. The use of different substrates revealed different behaviors and cleaning kinetics.

Issues related to the high adherence of the soil highlighted the poor characterization of the flow in the radial-flow cell and the absence of relevant wall shear stress distributions available in the zone where removal occurred. Consequently, in Section 5 (*"Laminar flow in radial-flow cell with small aspect ratios: numerical and experimental study"*), the flow inside the radial-flow cell has been characterized using computational fluid dynamics over the whole laminar regime. The computations were followed by an experimental validation of the numerical results.

After this study of radial-flow hydrodynamics, the concept of critical wall shear stress (minimal wall shear stress required for soil detachment) was investigated with new surface contaminants in Section 6 *("Flow rate dependency of critical wall shear stress in a radial-flow cell"*). These contaminants consisted in a size distribution of starch granule aggregates, constituted of a unimodal size distribution of polyhedral particles. At this point of the research, a robust methodology enabling the study of soil adherence and soil removal from solid surfaces with the radial-flow cell had been developed.

The limitations of the systems being known, a thorough investigation on the effects of experimental factors and physico-chemical processes on the formation and on the adherence of the particle aggregates can be found in Section 7 (*"Adherence of starch granule aggregates on different materials: experimental factors and physico-chemical mechanisms"*). Experimental factors such as water contact time, spatters drying time and surrounding relative humidity were investigated. The evolution of soil adherence with the experimental conditions (macroscopic observations) coupled to microscopic observations of the aggregates present on the model substrates allowed a better understanding of the physico-chemical mechanisms responsible for the aggregate formation and their adherence. Dynamic contact angle measurements and X-ray photoelectron spectroscopy suggested the presence of water-soluble macromolecules at the granule-substrate and granule-granule interfaces. Their role in soil adherence has been studied.

Lastly, in Section 8 (“*Adherence of starch granules aggregates to substrates: comparison between soft and hard hydrophobic substrates”*), the use of very hydrophobic substrates with different mechanical properties (rigidity) showed that the hydrophobic nature of a surface was not a sufficient condition to ensure the facilitated removal of aggregated particles coming from dried droplets of an aqueous suspension. Section 8 shows that additional surface parameters than surface hydrophobicity alone must be considered in the conception of easy-to-clean surfaces.

PART II – SECTION 4

CLEANABILITY ASSESSMENT OF MODEL SOLID SURFACES WITH A RADIAL-FLOW CELL

From:

Detry, J.G., Rouxhet, P.G., Boulangé-Petermann, L., Deroanne, C. and Sindic, M., 2007.

Colloids and Surfaces A: Physicochemical and Engineering Aspects, 302, 540-548.

PART II – SECTION 5

LAMINAR FLOW IN RADIAL-FLOW CELL WITH SMALL ASPECT RATIOS: NUMERICAL AND EXPERIMENTAL STUDY

From:

Detry, J.G., Deroanne, C., Sindic, M. and Jensen, B.B.B. 2009.

Chemical Engineering Science, 64, 31-42.

PART II – SECTION 6

FLOW RATE DEPENDENCY OF CRITICAL WALL SHEAR STRESS IN A RADIAL-FLOW CELL

From:

Detry, J.G., Jensen, B.B.B., Sindic, M. and Deroanne, C., 2009.

Journal of Food Engineering, 92, 86-99.

PART II – SECTION 7

ADHERENCE OF STARCH GRANULE AGGREGATES ON DIFFERENT MATERIALS: EXPERIMENTAL FACTORS AND PHYSICO-CHEMICAL MECHANISMS

From:

Detry, J.G., Sindic, M., Servais, M.J., Adriaensen, Y., Deroanne, C., and Rouxhet, P.G., 2009.

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PART II – SECTION 8

ADHERENCE OF STARCH GRANULE AGGREGATES TO SUBSTRATES: COMPARISON BETWEEN SOFT AND HARD HYDROPHOBIC SUBSTRATES

Project of publication.

PART II – SECTION 9

GENERAL DISCUSSION

SECTION 9 – GENERAL DISCUSSION

This thesis mainly evaluates the radial-flow cell as a tool to study and compare the cleanability of surfaces. The importance of the scientific conclusions drawn from this work is discussed below under fundamental and practical considerations, together with the limitations of this system and the reasons for which it has been chosen.

9.1 METHODS

9.1.1 Soiling

Developing a soiling procedure is always the primary step before considering any assessment of surface cleanability. Making this step as controlled and as reproducible as possible is often very challenging. Moreover, the soiling conditions must also be as close and as relevant as possible of the ones in which the investigated surface will be used.

We started this work (Section 4, "*Cleanability assessment of model solid surfaces with a radial-flow cell"*) by assessing a well-documented soiling method reported as adequate for the parallel plate flow cell, which consisted in the gentle scrubbing near the substrate of a tooth brush first soaked in refined sunflower oil. The method had initially been developed to mimic the random splashing occuring during food product handling (Boulangé-Petermann et al., 2003; Boulangé-Petermann et al., 2004a; Boulangé-Petermann et al., 2004b; Boulangé-Petermann et al., 2006).

The reproducibility of the method has been assessed and compared with the aspersion of the surface with the same oil using a thin layer chromatography (TLC) sprayer. Both methods considered here offer a reasonable compromise between mimicking splashing encountered "on the field" and providing a repeatable laboratory procedure.

As presented in Section 4, the TLC sprayer gave a narrower and more reproducible oil droplet size distribution. It was preferred since its use was easier, more reproducible and gave a higher drop density which facilitated the measurement of the detachment radius in the cleaning experiments. The experiments presented in Section 4 also showed the importance of well-controlled soiling method since the interesting kinetic effects observed after soiling the substrates with the TLC sprayer were masked when the toothbrush was used.

Refined sunflower oil was the soil used in the first part of the experimental work. It had been selected for comparison purposes with preexisting data. However, the scope of applications targeted by this soil seemed too narrow for SMARTNET and its removal was mainly related to the polarity of the surface and to oil spreading.

Therefore, we looked for a soil that could embrace the complexity of the situations faced by open-surfaces when exposed to a real environment while applicable in a well-controlled laboratory environment. We considered working with an aqueous suspension of particles and opted for starch granules. Waxy corn starch granules are polyhedral, mainly composed of amylopectin and have a unimodal distribution of sizes ranging over several dozens of micrometers. Depending on the substrate, they formed aggregates of different morphologies and size distributions. As shown in Sections 6 to 8, this soil is very complex compared to the systems generally found in the literature which are almost exclusively single microorganisms or particles of the same size, eventually interacting with well defined macromolecules.

Besides aerial contamination, the contact with aqueous contaminating fluids and their evaporation is a common source of contamination for open-surfaces. Furthermore, starch particles may be seen as a model for dusts, process powders or even microorganisms, since their surface is a more or less (de)hydrated gel layer mainly composed of polysaccharides. The presence of soluble polysaccharides and proteins in the granule suspension and their role in adhesion mechanisms brings the system closer to the real phenomena occurring, for instance, during the microbial contamination of open surfaces from spatters.

The intention of this thesis was not to study extensively all the situations corresponding to real environments but rather to select a model in order to understand how alterations of the environmental conditions and of the physico-chemical properties of the substrates influenced the formation of the aggregates and their adherence.

9.1.2 Cleaning tests

9.1.2.1 The traditional rectangular flow cell (parallel plate flow cell)

The parallel plate flow cell is presented in detail in Section 2.2. Its two main advantages are its ability to generate a simple flow of constant wall shear stress along a sample surface and its geometry that facilitates its modular insertion into a closed circuit. The use of a transparent material for the upper wall of the flow channel allows *in situ* observation.

The system was discarded because of two main limitations jeopardizing SMARTNET. The first was the unknowing of the wall shear stress value required for soil removal. The second was the geometry of the system required to provide all the flexibility needed in terms of wall shear stress range. Indeed, the size of the samples to be produced within SMARTNET was limited at 50 × 50 mm in the initial phase of the project, knowing that A4 format samples would come later and that their cleanability would also have to be assessed. However, the minimal sample dimensions required were 120 × 20 mm in order to ensure a uniform flow at the entrance of the observation zone for wall shear stresses varying between 0 and 100 Pa. These dimensions are valid for a flow channel height of 200 µm, which was too narrow for individual granules; any increase of the flow channel height resulting in increased sample sizes to keep equivalent shear stresses (Lorthois et al., 2001).

9.1.2.2 The radial-flow cell

Advantages and limitations of literature data

The radial-flow cell was conceived according to the design of Décavé (2002). The system was very appealing compared to its parallel plate counterpart for four reasons:

1. the same sample can be submitted to a range of wall shear stress in a single experiment;
2. sample exposure to an air/liquid interface can be avoided during the experiment and flow cell assembly or disassembly is fast and easy;
3. the size of the samples is not limited to the sole initial dimensions of the flow cell;
4. rapidity of system assembly and dismantling (appealing to study responsive surfaces).

However, on basis of the theory developed in the literature (Moller, 1963) and of the first experiments performed with the radial-flow cell (Section 4), it appeared that the latter provides an evaluation of the wall shear stress required for cleaning in a range below 3 Pa on base of existing analytical models, seriously limiting its use to weakly adhering soils (Jensen and Friis, 2005; Guillemot et al., 2006). Owing to the complex hydrodynamics near the inlet, a single observation only allows a range of wall shear stress to be covered within a factor of 2. This range can be extended on the same sample by changing the disk spacing and the flow rate, provided a direct observation is possible.

Comprehension of the flow within the radial-flow cell

Since the complex flow induced by the inlet geometry of the radial-flow cell prevented a quantitative estimation of the force required to remove the adhering soils, modern computing tools were used to study the characteristics of the flow (flow pattern, velocities and wall shear stresses) inside the radial-flow cell for flow rates at which the inlet flow regime was laminar and for several values of aspect ratio mentioned in the literature.

The numerical study of the flow made by computational fluid dynamics (CFD) is presented in Section 5 (“*Laminar flow in radial-flow cell with small aspect ratios: numerical and experimental* *study”*). The CFD simulations were performed in 2D but the tracer experiments performed in the real radial-flow cell suggest that the discrepancies existing between the numerical wall shear stress values and the real ones should be small at the sample surface, since the radial and axial components of the velocity vectors prevailed on the azimuthal velocity component where soil removal occurred. The use of other experimental techniques such as particle image velocimetry was considered to validate the velocity field inside the flow cell but the experimental results were inaccurate in the zone where removal occurred due to the specific nature of the flow at low flow rates (*vena contracta*) or to the presence of 3D flow structures at higher flow rates.

Complications arising from the nature of the flow within the radial-flow cell

The presence of the azimuthal component of the velocity vectors inside the real radial flow cell did not significantly alter the 2D radial profile of the flow but it is nevertheless delicate to say that this additional velocity component will not have a significant effect on the CFD-computed value of wall shear stress associated to the real resulting hydrodynamic drag force required for soil removal. This aspect was investigated in Section 6 *(“Flow rate dependency of critical wall shear stress in a radial-flow cell”*) where a non-dimensional parameter, the aggregate Reynolds number, was used to characterize the structure of the flow around starch aggregates adhering to samples placed in the radial-flow cell. This Reynolds number value is also useful to provide qualitative information on the drag coefficient relating the wall shear stress to the drag force exerted on the aggregates in the radial-flow cell.

Several conclusions were drawn from the complex nature of the flow (Sections 5 and 6):

1. For an aspect ratio of 1 and for Reynolds number values above 1500, the presence of inertial 3D flow structures in the radial-flow cell will likely be responsible for a decrease of the drag coefficient (Hubbe, 1984; Cardot *et al.*, 2001; Lorthois *et al.*, 2001).

2. The axial extension of the primary recirculation zone of the radial-flow cell (presented in Section 5) influences the shape of the axial flow velocity profile perpendicular to the sample surface. As explained in Section 6, if aggregate removal occurs in this zone and if these aggregates are too high, their top will rise above the linear part of the velocity profile. The drag coefficient will consequently be reduced and a higher wall shear stress will be required to exert the same hydrodynamic drag force on the aggregate (O'Neill, 1968; Hubbe, 1984; Brooks and Tozeren, 1996). In addition to their height, the distance

separating adjacent aggregates placed in a shear flow should be superior to 5 times their diameter to avoid any shielding effects of the shear field by the neighboring aggregates (Brooks and Tozeren, 1996).

However, this last consideration may not be as important for the radial-flow cell as for the parallel plate flow cell since the wall shear stress is related to the radius of the zone where the aggregates are removed in the radial-flow cell. Once the system stabilized (after approximately 3 min at a defined flow rate), the aggregates still present at the outer rim of the cleaned zone should thus be directly exposed to the shear flow with reduced shielding effects.

Use of the radial-flow cell: Suitable hydrodynamic conditions

Concerning weakly adhering soils, analytical equations are available in the literature and their validity domain has been refined in Section 5 as:

$$Re_{modified} = \left| \frac{\rho \cdot Q}{24 \cdot \pi \cdot r \cdot \mu} . \frac{h}{r} \right| \leq 0.145$$

The radial position *r* fulfilling this condition will thus typically refer to soils for which a wall shear stress below 3 Pa is sufficient for detachment. For soils adhering more strongly to their substrate, the maps giving the axial and radial extension of the recirculation zones inside the radial-flow cell (Section 5) should be used to design an adequate flow cell and to ensure the best hydrodynamic conditions possible around the adhering soils. However, a compromise will most probably have to be found between the available equipments, the maximum sample size allowed and the flow rate range deliverable by the pump. Ideally, the height of the soil should be small enough to remain in the linear part of the axial velocity profile directly adjacent to the wall (which was defined as ≤ 1/15 of the available flow channel for the parallel plate flow cell) and to ensure a Reynolds number << 1 around the soil (O'Neill, 1968; Brooks and Tozeren, 1996). This should be the case for detachment radii measured at inlet Reynolds numbers below 200 for substrates soiled with the starch granule aggregates and placed in the radial-flow cell with an aspect ratio of 1.

Empirical use of the radial-flow cell

Owing to the limitations imposed by both the hydrodynamic regime and soil thickness, the results of the last part of this work are expressed in terms of critical radius without any attempt to relate the detachment radius to the corresponding wall shear stress or to adhesion force values; keeping in mind that for a defined flow rate, the higher the critical radius, the lower the adherence.

It is furthermore delicate to estimate and anticipate the magnitude of soil adherence, especially for substrates with very different physico-chemical properties or after exposing the samples to various environmental conditions. Therefore working with a sequence of flow rates ensures a good contrast between different samples or experimental conditions as presented in this work. The method developed to study the adherence of starch granules aggregates is detailed in the Appendix 1 of this thesis.

9.2 PROCESSES

The sequence of events following the moment of contact between a drop of starch granules suspension and the substrate is described in Sections 6 to 8. The factors influencing starch granule adherence, the mechanisms leading to the formation of starch aggregates and the mechanisms responsible for their adherence are identified. The sequence of events, the mechanisms at play and the different factors influencing adherence are summarized below.

9.2.1 Physico-chemical mechanisms responsible for the adherence of starch granules aggregates to substrates

9.2.1.1 Droplet spreading (Sections 6 and 7)

Immediately after impact, the liquid drops spread (or not) on the substrate and take an equilibrium shape depending on the wetting behavior of the liquid on the considered substrate. The morphology of the starch aggregates results from both drop spreading, which decreases as the substrate contact angle increases, and capillarity effects which develop upon drying.

9.2.1.2 Capillary forces (Sections 6 and 7)

After spreading, the sprayed suspension droplets progressively evaporate, which leads to drop volume reduction, to partial immersion of the granules and to the appearance of lateral capillary forces which tend to gather the granules in the center of the drop. Further drop volume reduction leads to formation of water meniscuses, and thereby to creation of granules-granule and granule-substrate capillary forces. These processes are strongly dependent on substrate and granule wettabilities. Droplet spreading and the competition between capillary forces at the granule-substrate and granule-granule interfaces affect the shape and compactness of the adhering aggregates, and finally the adherence of soiling particles. The influence of the rate of drying is also important and is explained by the duration left to capillary forces for acting, slower drying rates resulting in higher adherence.

9.2.1.3 Macromolecules (Section 7)

Macromolecules present at the interface may hydrate and play a role in granule-substrate adherence. Their origin may be due twofold: a gel-like layer present at the surface of starch granules or macromolecules dissolved in the aqueous phase which adsorb on the substrate and/or are carried to the granule-granule and granule-substrate interfaces as water evaporates.

The possibility that the granule surface has gel-like properties is suggested by the high water uptake of starch in a humid atmosphere and by literature (Saibene and Seetharaman, 2006). The presence of macromolecules (mainly polysaccharides with a small proportion of proteins) in the aqueous phase of starch suspensions was demonstrated by its low surface tension and by XPS data (Section 7).

Therefore, it can be concluded that drying does not leave two solids in contact but maintains hydratable macromolecules at the interface which act as an adhesive joint. Its organization seems to be influenced by the detailed history of drying and/or exposure to humidity. These macromolecules are believed to act similarly to binders used in powder granulation (Meurk *et al.*, 2001; Saito *et al.*, 2002): under their dehydrated state they form a rigid joint which breaks easily when exposed to mechanical forces while they are solvated in humid atmosphere and transform the joint in a flexible and sticky adhesive, increasing significantly the adherence of the starch granules.

9.2.1.4 Substrate and soil particle deformation (Sections 7 and 8)

Starch granule adherence increases with time and humidity. A slow drying process, ensured by a high relative humidity (RH), allows the starch granules to reach more intimate contact with the substrate thanks to a longer action of capillary forces (Tang and Busnaina, 2000) and longer time for the reorganization of the macromolecules at the interface. In contrast, a low RH provokes a fast dehydration which freezes quickly the granule packing and reduces the starch-substrate contact (Section 7).

The increasing adherence with RH can also be explained by the fact that capillary forces are known to be responsible for time dependent adherence phenomena as a consequence of substrate or particle deformation (Tang and Busnaina, 2000), the extent of which depends on total interaction forces and on the plastic properties of the interacting bodies (Sharma et al., 1992; Das et al., 1994; Krishnan et al., 1994).

The increase of the water content of starch with RH (Section 7) leads to a higher plasticity which permits deformation of the starch granules and more intimate contact with the substrate. Deformation of the substrate also leads to more intimate granule-substrate contact and partial engulfment of the granules may even be observed when the substrate is sufficiently soft (Section 8).

9.2.2 Factors influencing adherence of starch granules aggregates to substrates

9.2.2.1 Influence of the substrate (Sections 6, 7 and 8)

Substrate wettability affects the spreading of droplets of the aqueous suspension of starch granules sprayed on the surface. Spreading is reduced when the contact angle increases. Due to the competition between capillary forces at the granule-substrate and granule-granule interfaces, the compactness of the aggregates is also influenced by substrate wettability. On hydrophilic substrates, the water drops spread and the granules become faster partially immersed. If the substrate is more hydrophilic than the granules, granule-granule capillary forces are lower than granule-substrate capillary forces, which lead to the formation of flatter, larger and looser aggregates. In contrast, granule-granule capillary forces are stronger than the granule-substrate forces on hydrophobic substrates; leading to the formation of higher and more compact aggregates (Sections 6 and 7).

In shear flow, the higher aggregates formed on hydrophobic substrates offer a larger section to the flow than those adhering to hydrophilic substrates, explaining their easier removal. The latter is also facilitated by the lower granule-substrate capillary forces. Different removal modes were also observed depending on the intensity of such forces: aggregates tended to detach as individual bodies from hydrophobic substrates while only small clusters of granules were detached from the aggregates adhering to hydrophilic surfaces (Sections 6 and 7).

Substrate wettability also influences dewetting upon drying, affecting the possible accumulation of macromolecules between the granules and the substrate and the final amount of macromolecules present at the interface. Whether macromolecules from the supernatant are adsorbed or carried by the retracting liquid film, it is clear that the interface between the starch granules and the substrate contains macromolecules and that both the amount of macromolecules present at the interface and their affinity to remain on the substrate will influence granule adherence. Indeed, preconditioning the substrates with a supernatant of the starch suspension increased the amount of macromolecules on the surface and decreased granule adherence due to the formation of a weak adlayer. Additionally to their quantity, the tendency of macromolecules to remain fixed on the substrate also influences adherence since the lowest adherence was observed for very hydrophobic substrates on which the amount of macromolecules remaining after preconditioning was appreciably lower due to their poor affinity for the substrate (Section 7).

Deformability is another substrate property influencing adherence and can be assessed by measuring the Young's modulus of the material. A substrate softer than the granules adhering on it may deform and bring the granule in more intimate contact with the surface, thereby significantly increasing starch adherence and eventually resulting in partial granule engulfment, independently of the very hydrophobic character of a surface or of its low affinity for the macromolecules present in the starch suspension supernatant (Section 8).

9.2.2.2 Influence of drying duration and humidity (Sections 7 and 8)

The influence of drying duration and humidity on adherence is twofold. Both influence the contact between the adhering granules and the substrate and both influence the mechanical properties of the adhesive joint present at the granule-substrate interface.

Drying duration influences the completeness of drying and the intimacy of the granule-substrate contact. While substrate hydrophobicity mainly influences granule adherence during the first minutes of drying (30 minutes), drying performed on longer intervals (one week) enables all the mechanisms involved in granule adherence to take place: capillary forces, macromolecules and substrate or granule deformation.

Humidity influences mainly the rate of drying and starch plasticity. High humidity slows down the rate of drying, leaving more time to capillary forces for acting; and increases granule plasticity, thereby facilitating eventual granule deformation. High humidity also increases the flexibility and the stickiness of the macromolecules constituting the adhesive joint at the granule-substrate interface. Therefore higher adherence will be observed for starch granule aggregates previously exposed to high humidity for long drying periods (typically one week).

In contrast, a low humidity provokes a fast dehydration leading to reduced granule-substrate contact and to a more brittle macromolecules joint at the granule-substrate interface. Exposing the starch granules in contact with the substrate to a long stay in dry atmosphere will thus generally result in lower adherence.

Adherence is dependent on the detailed history of drying and exposure to humidity, possibly through the reorganization of the macromolecules at the interface. Nevertheless, the last step of drying has a prominent effect.

On very hydrophobic substrates for which the affinity of the macromolecules present in the supernatant is low, adherence is not influenced nor by humidity nor by the history of drying; provided these substrates are sufficiently rigid.

9.3 PERSPECTIVES

9.3.1 Methods

Intensive efforts are deployed to develop the ideal surface that would remain clean under any circumstances and several trails are nowadays reported: modification of the surface roughness, photocatalytic coatings or stimuli responsive coatings. However, these technologies are limited to very specific applications in specific conditions, showing the importance of understanding the physico-chemical mechanisms responsible for the adherence of soils when developing easy-to-clean surfaces.

Indeed, attempts to relate surface engineering to a given application do not always allow real time and *in situ* observation in spite of the need to consider environmental variables such as flow, heat and mass transfer, passage of an air-liquid interface or presence of chemicals and surfactants. Flow chambers and other similar hydrodynamics devices reviewed in Section 2 proved to be valuable tools to take those environmental variables into consideration while allowing standardization and comparisons between laboratories.

9.3.1.1 Soiling methods

The spraying of surfaces with a suspension of starch granules allowed the identification of the mechanisms influencing granule adherence by investigating a range of model substrates in various environmental conditions. The soiling procedure is rapid and convenient and gave robust tendencies when studying adherence despite its apparent random character. The results of the present work suggest that this method is relevant to mimic and study soil adherence resulting from splashing as it occurs in real conditions and may be used for various liquid soils. In contrast with dipping the sample in a soiling solutions, dewetting problems can be limited with spraying but a uniform coverage of the surface by the soil is difficult to achieve.

The soiling method developed in this work is suited for applications targeting open-surfaces but is not relevant to other applications where fouling occurs for instance through scaling or through colonization of the substrate by living organisms. In the last case, soiling should be performed by letting the organisms settle on the immersed substrate in a flow cell before assessing their adherence to the substrate. Scaling can be assessed using the rotating disk to generate constant mass transfer condition on the samples. However, more complex deposits like dairy fouling (Changani et al., 1997; Bansal and Chen, 2006), biofilms (Janknecht and Melo, 2003) or even scale deposits (Brahim et al., 2003; Augustin et al., 2006) should be formed by placing the samples in a test rig where exposure to real processing conditions can be simulated.

9.3.1.2 Cleaning tests

The abundant literature available on the parallel plate flow cell show its efficiency for exposing a sample to a simple shear flow of controlled wall shear stress. The hydrodynamic conditions inside the flow channel are well characterized and facilitate the fine interpretation of the results. However, complying with the criteria required for the proper use of the system may limit its application field because the soiling particles or cells must be well-separated and because the combination of a particle/cell/drop height, with its adherence strength and the maximum affordable sample surface are key factors for the conception of the flow cell.

The radial-flow cell presented in this work exhibits more flexibility but the fine interpretation of the results becomes much more delicate in the case of strongly adhering droplets/particles/cells, as presented in this work. However, the system can yield robust and valuable information on the mechanisms of soil-substrate adherence as well as substrate ranking despite its very complex hydrodynamics. The shear stress range resulting from the system geometry is a very interesting feature of the radial-flow cell that permits the study of dynamic interactions on the same sample, as presented at the beginning of this work. The possibility of realizing *in situ* observation of dynamic systems such as responsive surfaces would increase the attractiveness of the radial-flow cell but it remains difficult to implement for non-transparent samples. Repeated cleaning-soiling cycles are also impossible to perform with the system since the central zone is the only part of the sample to be cleaned. Furthermore, the radial-flow cell is also limited to relatively isolated soil entities and is not suited for thick continuous deposit layers.

For such deposits, the best way to assess surface performances upon cleaning remains their fitting in a test rig where cleaning-in-place (CIP) conditions are reproduced since these deposits are generally formed in pipes and closed equipments which are normally cleaned by CIP. The degradation and the removal of such deposits by chemicals or detergents can be investigated using the rotating disk (Morison and Thorpe, 2002; Chateau et al., 2004). The effect of these chemicals on the deposit structure (swelling or deposit break-down) can be measured by fluid dynamic gauging. This way of assessing cleaning is maybe less useful to equipments and materials manufacturers but is relevant to cleaning products manufacturers since the nature of the deposit and thus the ease at which it can be removed by chemical agents may be influenced by the substrate.

Finally, the use of the parallel plate flow cell, of the radial-flow cell, of the rotating disk or of the fluid dynamic gauging is relatively heavy to implement and relies on acute methodologies such as the one presented in the Appendix 1 of this thesis. Though allowing a deep understanding of the processes involved in soil adherence and cleaning, the development of such cleaning tests is too long and too costly for most of private companies. Therefore, the development of more empirical tests giving similar tendencies and results but easier to adapt to a daily routine should be validated on base of methods such as the one presented here for the radial-flow cell.

9.3.2 Process

The Sections 7 and 8 of this work showed that several parameters must be considered when developing easy-to-clean surface for open-surface applications: the wetting of the substrate by the soiling liquids, the ambient humidity level, substrate deformability, short and long-time soil adherence and the possible influence of biological molecules, other minor constituents and impurities that may concentrate at the soil-substrate interface. In the case of particulate aqueous suspensions of starch granules, the best surface to reduce soil adherence would be a hydrophobic substrate (minimized droplet spreading and low capillary forces) where the affinity of the biological molecules present in the suspension for the substrate would be minimal to provide low granule adherence, independently of humidity. The surface should also be sufficiently rigid to avoid deformation when in contact with starch granules.

Sections 7 and 8 also stress the need of assessing the cleanability of substrates not only on a short term basis (30 min), for which case the results are mainly influenced by substrate hydrophobicity but also on a longer term horizon (1 week), where a stable state is reached and where the influence of eventually present macromolecules, of environmental conditions or of other substrate properties on soil adherence can be assessed.

The mechanisms influencing granule adherence have been identified by investigating a range of model substrates sprayed with a suspension of starch granules and exposed to various environmental conditions. However, performing experiments with responsive surfaces such as the ones targeted by SMARTNET would have been particularly interesting to see how the macromolecules present in the system would affect granule adherence upon exposure to dry or humid conditions. Whether proteins or polysaccharides are the main responsibles for the increased granule adherence attributed to macromolecules is also not clear. Therefore, studying the adherence of starch granules or other model particles in aqueous suspension containing a model protein or polysaccharide and sprayed on the surface may also be of interest to acquire deeper knowledge on the processes described in Section 9.2.

It should be noted that the mechanisms and factors influencing adherence which were identified here are valid for a few layers of granules forming delimited aggregates. If a paste mainly composed of particles was spread on the substrates, the influence of the capillary forces and mechanical stresses appearing in the bulk of the soil upon drying could not be further ignored and may possibly influence the adherence of the deposit. However, the first layers in contact with the substrate would be expected to exhibit a similar behaviour to the starch granules of this work.

Finally, it should be stressed that the processes responsible for granule adherence described here may be considered for the fixation of a single microorganisms on a substrate upon the drying of spatters but they cannot be applied to biofilms since the latter are constituted of a gel matrix of macromolecules containing dispersed microorganisms instead of particles adhering through the adhesive action of macromolecules present in low quantities. However, the matrix of a dried biofilm is expected to behave like the adhesive joint of macromolecules described in this work upon exposure to ambient humidity.

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