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ZERO DISCHARGE FLUID DYNAMIC GAUGE (ZFDG) FOR STUDYING THE SWELLING OF SOFT SOLID FOULING LAYERS

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ABSTRACT

This paper demonstrates the performance of a benchtop device to study the swelling or shrinking of soft solid layers immersed in a liquid environment in situ and in real time. This is achieved by measuring the flow and pressure drop of liquid through a nozzle positioned near to, but not touching, the layer. A particular feature is that the liquid volume is isolated from its surroundings, hence the name Zero net discharge Fluid Dynamic Gauging (ZFDG), and making ZFDG suitable for aseptic operation. Calibration tests were performed on flat, clean 316 stainless steel substrates with a nozzle of diameter 1.78 mm and indicated a resolution of \pm 5 µm and uncertainty of \pm 10 µm. Gelatin layers were tested as model soils under various pH conditions. These exhibited a fast initial hydration step followed by swelling: power law, second order and diffusion-relaxation models were fitted to the experimental data and indicated 'sub-Fickian' behaviour. There was good agreement between gravimetric and ZFDG measurements, and ZFDG proved more reliable at higher pH. Ionisation of the protein at high pH promotes notably greater swelling in the gelatin.

INTRODUCTION

In the chemical and biotechnological sectors, fouling is generally defined as the accumulation of unwanted material on solid surfaces with detrimental consequences, *i.e.* fouling layers are unwanted coatings. Fouling can result in high maintenance costs, low process efficiency and additional capital expenditure. It is therefore essential to understand the interactions between deposits and flowing fluids in order to prevent the former attaching, or to promote their detachment (cleaning). This requires both modelling and experiments in monitoring and quantifying the growth and removal of fouling deposits.

During cleaning, it is challenging to identify the extent of cleaning and the mechanism involved. Many fouling deposits are soft solids and are deformed, or weaker, once the layer is removed from its native environment. Biofilms are examples: these often collapse when removed from water. Furthermore, properties of the layer may change over time. The technique of fluid dynamic gauging (FDG) was developed to measure the thickness and estimate the mechanical strength of soft solid layers immersed in liquid *in situ* and in real time, (Tuladhar *et al.*, 2000; Chew *et al.* 2004). It does not require knowledge of physical and chemical properties of the solutions and samples except the presence of a locally stiff surface. It has some advantages over other techniques, such as ultrasound, magnetic resonance imaging in being relatively cheap and amenable to flow conditions.

This paper reports a step change in FDG measurement, called zero net discharge fluid dynamic gauging (ZFDG) wherein the liquid volume remains constant during measurement. In previous FDG measurements, liquid was added or withdrawn, which is undesirable for aseptic operation or when costly or hazardous liquids are used. The ZFDG concept was demonstrated in principle by Yang *et al.* (2014): this paper demonstrates its application in a semi-automated device that could, with further automation, be made to scan over a surface in a similar manner to the FDG device described by Gordon *et al.* (2010).

Fig. 1 illustrates the principles of ZFDG operation. The schematic elevation shows the gauge nozzle geometry and dimensions. The nozzle is located near the surface, at clearance h_0 , and liquid is ejected from or sucked into the nozzle at mass flow rate \dot{m} . The pressure drop across the nozzle, ΔP , is measured and is related to \dot{m} , the nozzle lip width, w_r , nozzle rim thickness, w_e , nozzle throat diameter d_t , and clearance, h_0 :

$$\Delta P = f(\dot{m}, \frac{h_0}{d_t}, \frac{w_r}{d_t}, \frac{w_e}{d_t})$$
(1)

 ΔP is expressed as the discharge coefficient, C_d , which is the ratio of actual and ideal mass flowrates, *i.e.*

$$C_d = \frac{\dot{m}_{actual}}{\dot{m}_{ideal}} = \frac{4\dot{m}}{\pi d_t^2 \sqrt{2\rho\Delta P}}$$
(2)

For a given geometry C_d is a function of h_0/d_t alone: measuring C_d allows the distance h_0 (or distance from a fouling layer surface, h) to be calculated. By alternatively ejecting or withdrawing liquid at a fixed, low flow rate through the nozzle, measurements of ΔP can be made with no total volume change over an extended period of time. This is the ZFDG concept: the individual components were demonstrated by Yang *et al.* (2014), supported by computational fluid dynamics simulations of the flow patterns in ejection and suction.



Fig. 1. Schematic elevation of ZFDG geometry with dimensions: $\theta = 54^{\circ}$, $d_t = 1.78$ mm, $d_i = 4$ mm, $w_e = 0.2$ mm, $w_r = 1.32$ mm. Dotted streamline – ejection; dashed streamline – suction (showing flow recirculation).

Calibration is performed on a flat clean solid substrate, and ΔP is measured by adjusting h_0 at constant \dot{m} . Calibration plots present discharge coefficient, C_d , against dimensionless clearance, h_0/d_t . When a fouling layer is present, the nozzle-substrate distance, h_0 , is known from independent measurements. ΔP is measured at different nozzle locations (z) and nozzle-soil clearance, h, is determined using the calibration plot. The deposit thickness is calculated from

$$\delta = h_0 - h \tag{3}$$

ZFDG APPARATUS

Fig. 2 shows photographs and a schematic of the apparatus. The syringe pump (Hamilton[®] Glass, $d_0 = 32.97$ mm syringe; Harvard Apparatus PHD UltraTM Series pump) controls the flowrate and direction. The measured accuracy in \dot{m} is 1% of the set value. The gauging liquid is contained in a Perspex tank ($280 \times 280 \times 160 \text{ mm}^3$). The nozzle is installed at the end of a long stainless steel tube (L = 310 mm), sealed by two O-rings, and the nozzle – surface clearance is automatically controlled by a stepper motor (Zaber Technologies, T-LSR075B, CE). The zero position is set by using a feeler gauge with known thickness (*e.g.* 0.1 mm) and calibration tests are started from $h_0 = 5.0$ mm.

The nozzle is brought towards the substrate (50 mm diameter 316 stainless steel discs, approximately 1.0 mm thick) at steps of 0.02 mm until it reached h_0 (or h) = 0.1 mm, at which point the pressure drop across the nozzle usually exceeds the pressure transducer operating limit (7.0 kPa Honeywell[®], 24PCE analogue differential pressure transducer). At each nozzle location, ΔP is measured: that acquired while the syringe pump is running is denoted ΔP_{dyn} ; the static measurement, ΔP_{static} , is recorded after the pump stops, in order to correct for any hydrostatic component in the pressure drop. Data collection and

processing is performed using LabVIEW[®] (National InstrumentsTM), which also controls the nozzle location and syringe pump motion. The apparatus is designed to move the nozzle and tank in laterally orthogonal directions, *i.e.* x and y: lateral position was adjusted manually in these tests.

Aqueous gauging solutions were prepared using deionized water (pH 5.6), adjusted to various pH values up to 11.6 by adding 1 M NaOH solution. Tests were performed at 16.5 °C and atmospheric pressure. Mass flow rates of 0.17 – 0.90 g/s were used, giving Reynolds numbers at the nozzle throat, $Re_t = 4m/\pi\mu d_t$, of 105 - 567.



(b)





Fig. 2. ZFDG apparatus (*a*) photograph of test rig; (*b*) photograph of nozzle; (*c*) schematic. Labels: A – syringe pump, B - stepper motor, C - pressure transducer (PT), D - nozzle, and E - data acquisition device (DAQ).

Calibration protocol

The nozzle is moved towards the substrate, alternatively ejecting and withdrawing liquid at each nozzle location. ΔP is recorded at clearances from 5.0 mm to 0.5 mm at steps of 0.1 mm, and from 0.5 mm to 0.1 mm at steps of 0.02 mm, giving more increments in the useful measurement region (0.05 < h/d_t < 0.20). The control software waits for the ΔP reading to reach steady state, which takes about 5 s. Calibration plots obtained with nozzle moving away from the substrate gave identical results.

MATERIALS AND METHODS

Sample preparation

Gelatin layers were prepared on the SS316 plates used in calibration tests. This steel is widely used in food and pharmaceutical manufacturing. The discs were pre-cleaned by washing in deionised water, soaking in acetone and drying in air.

Coating solutions of gelatin were prepared by adding 9 g of powdered beef gelatin (84 wt% protein, 15 wt% water, 1 wt% carbohydrate; Dr. Oetker, from local supermarket) to 100 ml deionised water and dissolved by heating gently (125 °C, 30 min). The soiling liquid had a liquid content of 93 wt%: 2 ml was then pipetted on to the disc surface and surface tension spread the liquid out to give a thin even layer. The sample was then air dried (16.5 °C, ~24 h) before being stored chilled. The procedure gave uniform, dry (~10-20 wt% water content), colourless layers, with thicknesses measured by micrometer in the range 50-80 μ m.

Gravimetric tests were performed in a spill tray containing aqueous solutions. Gelatin coated stainless steel discs were immersed in the solution, and after given soaking times, were removed from the liquid and wiped using a paper towel. The wet mass was measured by a balance and the thickness by a digital micrometer.

RESULTS & DISCUSSION ZFDG Calibration

Fig. 3 shows sets of calibration plots (C_d vs h_0/d_t) obtained using flowrate of 0.33 g/s and deionised water as a gauging liquid. The plots show similar behaviour to that reported for previous FDG systems (e.g. Chew et al., 2004b, Gordon *et al.*, 2010). For a fixed flow rate, ΔP is greatest and C_d smallest when the nozzle is close to the surface. C_d increases with increasing h_0/d_t , and approaches an asymptote, in this case 0.32. The value of the asymptote depends on Re_t . In this pressure mode gauging configuration, lower C_d values are preferred as the ΔP measured for a given flow rate is large. The error bar associated with the uncertainty in the pressure transducer measurement becomes more significant at larger h_0/d_t . At small h_0/d_t values the shear stress imposed by the gauging liquid on the surface is large and there is thus a trade-off between measurement accuracy and measurement reliability (Salley *et al.*, 2010). The range of h_0/d_t values in the inset in Fig. 3 represent the optimal range of gauging conditions: the calibration plot is usefully close to linear in this region.



Fig. 3. Calibration plots, of C_d against dimensionless clearance h_0/d_t , for 0.33 g/s water at 16.5°C. Symbols: square – ejection (E), triangle – suction (S). Solid and dashed lines show polynomial fittings for ejection and suction measurements, respectively.

Fig. 3 shows that suction mode gives a lower C_d at the same h_0/d_t , which is due to recirculation zones generated in the diverging part of the nozzle in suction (Yang *et al.*, 2014).

Accuracy and Resolution

The main uncertainties in measurements arise from the accuracy of zeroing the nozzle–substrate clearance, h, since ΔP is very sensitive to lower values of h. The accuracy of the mass flow measurements is good. Pressure transducer uncertainty is reduced by amplification and filtering to increase the signal to noise ratio.

The accuracy of measurement was checked by comparing ZFDG data with values obtained with a digital micrometer (Mitutoyo[®]). The thickness of a test layer was measured using the micrometer as 900 μ m. ZFDG measurements were performed on the test layer at three flow rates (0.17 – 0.50 g/s) and the agreement with the micrometer measurements compared at each h/d_t value tested. These tests indicated that the best agreement was obtained with h/d_t near 0.10, with an uncertainty of \pm 10 μ m. For a given flow rate, this corresponds to a given ΔP value: in practice, the nozzle is moved towards the surface until the pressure transducer reads this value, and the layer thickness can then be determined from Equation (3).

The dynamic thickness measurements on gelatin were performed with a flowrate of 0.33 g/s with a change in h/d_t from 0.20 to 0.05 with steps of 0.01 every 5 s. Suction mode gave an accuracy of ±10 µm and ejection mode slightly worse, at ± 20 µm. The best achievable resolution was~ ± 5 µm for measurements at 0.09 $< h/d_t < 0.11$. When the syringe

movement is started there is a brief instability causing a disturbance in ΔP which lasted about 1 s. These transients were eliminated from the data before processing for thickness measurements.

SWELLING STUDIES

Swelling Models

Three quantitative models are used for characterising the gelatin layer swelling kinetics.

Power law

Polymer swelling behaviour was reviewed by Ritger and Peppas (1987). The swelling kinetics are often described by a power law relationship, *viz*.

$$m = m_{\infty}k_1 t^n \tag{4}$$

where *m* is the solvent uptake at time *t*, m_{∞} is the solvent uptake at swelling equilibrium, *t* is the time for the layer being immersed, k_1 is the kinetic constant and *n* is diffusion index. Assuming the solvent uptake is equally distributed over the surface and the surface area does not change throughout the swelling process, the increase of mass is proportional to the change in thickness of the soil layer. This gives

$$\delta - \delta_0 = (\delta_\infty - \delta_0) k_1 t^n \tag{5}$$

where δ_0 is the initial dry thickness, δ_{∞} is the equilibrium thickness and δ is the layer thickness at time *t*. By interpreting swelling profiles (plots of δ against *t*), regression can be used to identify the parameters, $(\delta_{\infty}-\delta_0)$, k_1 and the diffusion index, *n*: the latter indicates the type of kinetics in the swelling process.

Second order swelling

This model was proposed by Schott (1992) to fit the swelling dynamics for gelatine and cellulose in aqueous media. The rate of swelling is assumed to be proportional to

- i. swelling capacity (fractional amount of swell), given by $(m_{\infty} m)/m_{\infty}$, at time *t*, and
- ii. the internal specific boundary area S_{int} , representing the enclosing sites of polymer networks that still have not yet interacted with water at time *t* but will hydrate and swell in due course, given by

$$S_{int} = k' \left(\frac{m_{\infty} - m}{m_{\infty}}\right) \tag{6}$$

Polymeric networks are held together by hydrogen bonds and other secondary valance forces between adjacent chains. When solvent penetrates into the polymeric structure, the inter-chain secondary valance bonds are broken and new hydrogen bonds are formed, particularly if the solvent promotes charge formation on the polymer. The polymer network expands to accommodate the influx of solvent through the relaxation produced by osmotic pressure. Therefore, S_{int} is a significant parameter in this swelling model. The overall rate of swelling is written as,

$$\frac{d\mathbf{m}}{dt} = k \left(\frac{m_{\infty} - m}{m_{\infty}}\right) S_{int} = kk' \left(\frac{m_{\infty} - m}{m_{\infty}}\right)^2 = k_2 (m_{\infty} - m)^2 \quad (7)$$

Integrating from the initial condition at t = 0, $m = m_0$, yields

$$m(t) = \frac{m_0 + m_\infty k_2 t(m_\infty - m_0)}{1 + k_2 t(m_\infty - m_0)}$$
(8)

By assuming uniform density and thickness across the layer,

$$\delta = \frac{\delta_0 + \delta_\infty k_2 t(\delta_\infty - \delta_0)}{1 + k_2 t(\delta_\infty - \delta_0)} \tag{9}$$

Diffusion-relaxation

This model was first proposed by Frish *et al.* (1969). It combines diffusion in glassy polymers and 'relaxation-controlled transport' components, where the latter is treated as a convection process and caused by the internal stresses due to penetration of solvent and reforming of hydrogen bonds. It was then developed to describe the release behaviour of dynamically swelling hydrogels (Peppas *et al.*, 1989), and gives a dynamic expression of the form

$$\frac{m}{m_{\infty}} = k_3' t^{1/2} + k_4' t \tag{10}$$

where $k_4't$ indicates relaxation-controlled transport and $k_3't^{1/2}$ represents the diffusion-controlled process. Again, by assuming uniform density and thickness across the layer,

$$\delta = k_3 t^{1/2} + k_4 t \tag{11}$$

This expression was shown to fit experimental data reported by Kwei and Zupko (1969) for uptake of trichloroethylene and methylethylketone in an epoxy polymer by Wang *et al.* (1969).

Swelling of gelatin layers

The swelling of gelatin layers was studied under various pH conditions at constant (laboratory) temperature. The initial dry thickness of the layers was $50-80 \ \mu m$.

Reproducibility

The ZFDG system did not allow the nozzle to scan the surface (Gordon *et al.*, 2014) and thus collect several data sets from each sample, so reproducibility is essential in order to gauge the reliability of individual results. Several samples (at least four) were tested for each condition studied in order to establish the variability between samples. Fig. 4 shows acceptably good intra-sample reproducibility for notionally identical experiments. There is good quantitative agreement, giving standard deviations of 40 μ m and 34 μ m using ejection and suction modes, respectively.

The thickness evolution profiles show the same behaviour. Ejection mode profiles were identical. It takes about 30s to load the sample initially, during which there is a fast hydration step driven by diffusive, chemical and electrostatic interactions (Ofner III and Schott, 1986). The layer thickness is then approximately 100 μ m. Thereafter the rate of swelling decreases as more solvent diffuses into the polymer matrix. The layer thickness approaches an asymptote, at some time beyond within 5000 s. Similar behaviour was reported by Gordon *et al.* (2010) using a scanning FDG system. The key difference in these measurements is that with ZFDG the volume of liquid is retained in the system and could be reduced to less than 1 L

if needed. Moreover, measurements could be made at intermittent intervals, so that the layer is exposed to quiescent liquid between gauging operations.



Fig. 4. Reproducibility testing, for initially dry gelatine layers immersed in pH 9.4 solution (16.5 °C) at t = 0 s. Gauging flowrate 0.33 g/s ($Re_t = 189$), suction mode. Data decimated for clarity: 1 out of 40 data points is plotted. Sample initial wet thickness 1.02 ±0.01 mm: dry thickness (1) 63 ±10 µm – square; (2) 55 ±10 µm – diamond: (3) 52 ±10 µm – triangle; (4) 51 ±10 µm – cross. Black error bars show maximum experimental uncertainty.

Gravimetric Measurements

Scott (1992) monitored the swelling of gels gravimetrically, which is both simple and effective. Fig. 5 compares the results for ZFDG and gravimetric testing on notionally identical gelatin layers using deionised water (pH 5.6 owing to absence of dissolved CO₂) and pH 11.0 solutions. The data are presented as the volume fraction of water (solvent), ε , assuming that the molar volumes of water and polymer are both constant, calculated by equation (12). Thickness measurements are also performed during the gravimetric samples, shown in Fig. 5 (b).

$$\varepsilon = \frac{m_t - m_{dry,gelatine}}{m_t} \tag{12}$$

The gravimetric measurements show that ε increases rapidly initially, from about 10% to 80% within 8 min, followed by slow swelling to an asymptotic value of around 90%. There is a larger initial rate of water/solvent uptake at the higher pH and the asymptote is approached faster, shown in Fig. 5 (a). This is consistent with the thickness data measured with the micrometer and ZFDG. The evolution of thickness measured by ZFDG in suction is shown in Fig. 5 (c). There is a smooth, strongly non-linear trend evident.

The plot of gravimetric data parameters in Figure 5(*b*) provides insight into the gelatin layer behaviour. Initially ε increases with δ , indicating expansion of the polymer matrix. Later ε is almost independent of δ , attributed to relaxation.

There is very good agreement between the two thickness measurement techniques for deionised water, whereas there is a noticeable discrepancy between the two data sets at longer times at pH 11, when ε exceeds 50%, in Fig. 5(c). Consistently smaller values are recorded by the micrometer, which is likely due to (*i*) loss of water during the period from the sample being removed from the liquid and measurement, and (*ii*) the increasing softness of the material at higher voidage: the micrometer measurement is recorded when the stub experiences a reaction force and some compression of the layer is required to record a datum. The force imposed by the gauging flow is small by comparison. The ZFDG data collected in suction and ejection modes at longer times differed by about 5%, indicating some elasticity in the layer.

Fitting the results to the power law model, gave

pH 5.6 $\varepsilon = 0.16^{*}t^{0.22}, \, \delta_{\text{grav}} = 0.06^{*}t^{0.26}, \, \delta_{\text{ZFDG}} = 0.055^{*}t^{0.282}$ pH 11 $\varepsilon = 0.20^{*}t^{0.22}, \, \delta_{\text{grav}} = 0.05^{*}t^{0.32}, \, \delta_{\text{ZFDG}} = 0.017^{*}t^{0.462}$

The power law indices for the two thickness measures differed at high pH. Both sets feature a diffusion index < 0.5, termed 'sub-Fickian' diffusion, indicating that the water penetration rate is slower than the polymer chain relaxation rate (Wang *et al.*, 2008).

ZFDG demonstrates significant advantages over gravimetric testing: (*i*) the samples do not have to be removed from solution; (*ii*) more frequent measurements can be made (and with scanning, more sites tested), (*iii*) softer layers can be studied reliably; and (*iv*) measurements are not influenced by the user's experimental technique. Furthermore, ZFDG tests can be conducted at other temperatures (and pressures) readily, rendering it a powerful tool for studying fouling or cleaning.

Influence of solution pH

Alkaline solutions are generally cheap cleaning agents that can break down protein layers through the action of hydroxyl ions (Lelieveld, 2003). The swelling of gelatin in alkali at lower pH is governed by osmotic pressure differences arising between the protein phase and the external solution (Bowes and Kenten, 1950). As a protein, however, the biopolymer contains ionisable functional groups. When there is electrostatic repulsion among the groups, it leads to chain expansion which can affect the macromolecular chain relaxation (Gierszewska-Drużyńska and Ostrowska-Czubenko, 2011). The swelling mechanism then becomes more relaxation-controlled. Ionisation is expected to have an influence when the pH in the layer approaches the pK_a values for the amine groups in gelatin: for glycine (21 wt% of total), at 9.6; for glutamic acid (10 wt%), at 9.7; for proline (12 wt%), at 10.6; and for hydroxyproline (12 wt%), also at 10.6 (Stevens, 1992).

Fig. 6 shows swelling profiles recorded for solution pH ranging from 5.6 to 11.6. In all cases the rapid initial hydration phase is completed within 200 s and is followed by slower swelling. There is a noticeable effect of the strongest alkali after 8 min (\sim 500 s), with a swelling rate about twice that of other cases.



Fig. 5. Swelling profiles for initially dry gelatine layers immersed in deionised water (16.5 °C, pH =5.6) and pH 11.0 solution. Symbols – gravimetric data: square – pH 5.6, triangle – pH 11 (a) Gravimetric measurements of water volume fraction (ε) against logarithmic time; solid and dotted lines show fitted power law models for pH 5.6 and 11 respectively. (*b*) Water volume fraction (ε) against thickness, gravimetric tests. (*c*) Swelling profile (δ vs. *t*) in suction. Solid and dotted lines are ZFDG measurements for pH 5.6 and 11, respectively.



Fig. 6. Swelling profiles for gelatine layers immersed in aqueous solution. Data are decimated for clarity (1 point per 40 is plotted). 16.5 °C, suction mode, \dot{m} = 0.33 g/s.



Fig. 7. Comparison of swelling models with ZFDG experimental data for gelatine layers at pH 11.6, 16.5 °C. (*a*) power law; (*b*) second order; (*c*) diffusion-relaxation. Data decimated for clarity. Symbols: ejection – circle, solid loci; suction – triangle, dashed line. \dot{m} = 0.33 g/s, initial dry thickness 58 ±10 µm.

After 5000 s the extent of swelling at pH 11.6 was about 50% greater than that at pH 11. By comparison, the extent of swelling at pH 11 after 5000 s (~ 0.8 mm in Fig. 6) was about 50% greater than that at pH 5.6 (~ 0.55 mm). Gordon *et al.* (2012) reported similar differences between pH 11.6 and 9.4 (achieved using 0.03 M buffer solutions). The large increase in swelling at pH 11.6 is attributed to deprotonation of the amine groups on proline and hydroxyproline driving charge repulsion within the layer. It is noticeable that the amount of extra repulsion was relatively weak at pH 11. This could be due to exhaustion of the H⁺ supply as the solvent diffuses into the layer and is consumed in protonation steps: a high concentration is needed to drive the reaction to completion (Mercade-Prieto *et al.*, 2008).

The three semi-empirical models were fitted to the experimental data. Fig. 7 shows a swelling profile obtained for gelatine layer contacted with pH 11.6 solution. The models were fitted to data after the hydration stage and all three give excellent agreements over the time period studied ($R^2 > 0.96$).

For the power law model, the diffusion index, n, is close to 0.5 (Table 1) which indicates Fickian diffusion. When the solvent penetration rate is much slower than the polymer chain relaxation rate, the value of n can be less than 0.5 (Dengre *et al.*, 2000). Table 1 shows that the diffusion indices follow an increasing trend, rising from 0.27 to 0.50 with increasing pH: there is a transition between 'sub Fickian' behaviour, which is dependent on the relative contribution of penetrant diffusion and polymer relaxation, and Fickian with increasing pH.

The second order model (Fig. 7 (b)) shows good agreement except in the initial stages of swelling (t < 400 s), where it over-predicts the layer thickness. The equilibrium thickness of the gelatine layer, δ_{∞} , can be extrapolated from the model parameters, giving $\delta_{\infty} = 2.36 \pm 0.02$ mm and 2.27 ± 0.02 mm for the ejection and suction case, respectively. These values are four times the initial wet thicknesses of the layers, which suggest that the polymer wants to adopt a more open configuration.

Table 1. Fitting parameters obtained from least square regression of swelling models to ZFDG data at different pH (including Figure 7) at 16.5 °C.

pН	$(\delta_{\infty} - \delta_0) * k_1$	n	R^2
5.6	0.055	0.282	0.997
6.8	0.023	0.383	0.995
8.5	0.020	0.422	0.999
9.4	0.036	0.351	1.000
10.5	0.020	0.431	0.998
11	0.017	0.462	1.000
11.6	0.023	0.488	0.999

The diffusion-relaxation model (Fig. 7 (c) proved to be superior, giving a good fit ($R^2 > 0.99$) for all the datasets

tested. It is noteworthy that it contains the same number of adjustable parameters as the power-law and second order models.

Influence of gauging flowrates

The gauging flow does exert a deformative stress on the surface being gauged. The impact of this stress was investigated by using different gauging flow rates (\dot{m} = 0.33 g/s-0.67 g/s) in notionally identical tests. Fig. 8 shows the evolution of gelatine layer thickness for suction operation under tap water conditions. There is a small difference in measured thickness, with the largest value recorded for the highest flow rate and thus largest tensile stress which would cause the layer to swell. This confirms that the gel has an elastic response to stress. Further work will include systematically switching between suction and ejection to determine the mechanical properties of the interface.



Fig. 8. Swelling profiles of gelatine layers with initial thickness ~ 80 ±10 µm obtained with different gauging flowrates. Square, \dot{m} = 0.33 g/s; triangle, \dot{m} = 0.50 g/s; cross, \dot{m} = 0.67g/s. pH 6.8 (tap water), suction mode.

Future work will include the study of the swelling and removal characteristics of other soft solid materials related to fouling and cleaning, including potato starches, polymerized grease, PVA glue and biofilms. Simulations of coupled-flow deformation will also be performed.

CONCLUSIONS

- 1. The concept of automated zero discharge fluid dynamic gauge is demonstrated using a stainless steel nozzle with i.d. 1.78mm. Calibration tests show that a useful measurement range lies between $0.05 < h_0/d_t < 0.20$;
- 2. The device described here provides a maximum resolution of \pm 5 µm, and accuracy of \pm 10 µm for the thickness measurements;
- 3. ZFDG has significant advantages over other measurement techniques, including (*i*) the samples do not have to be removed from solution; (*ii*) more frequent measurements can be made (and with scanning, more sites tested), (*iii*) softer layers can be studied reliably; and (*iv*) measurements are not influenced by the user's experimental technique.

- 4. The swelling characteristics of thin gelatin layers were investigated. FDG and gravimetric data indicate 'sub-Fickian' diffusion at low pH: the contribution of polymer relaxation increases as pH of gauging liquid increases;
- 5. Swelling profiles for pH from 5.6 to 11.6 all fitted the power law, second order and diffusion-relaxation models well, with the latter giving the best description.
- 6. Flowrate does not affect the swelling much but there is a slight higher initial swelling rate for higher flowrate due to higher penetration depth induced;

NOMENCLATURE

Roman		
$C_{\rm d}$	Discharge coefficient	-
di	Inner diameter	m
$d_{\rm t}$	Diameter of nozzle throat	m
d _o	Outer diameter	m
h	Nozzle-laver clearance	m
h_0	Nozzle-substrate clearance	m
i.d.	Internal diameter	m
k	Rate constant. Equation (7)	$kg m^{-2}s^{-1}$
k_1	Rate constant for power law model	s ⁻ⁿ
<i>k</i> [']	Area factor, Equation (6)	m^2
k_2	Rate constant for second order	kg ⁻¹ s ⁻¹
k_{3}	Rate constant for relaxation-controlled	s-0.5
5	transport	
k_3	Rate constant for relaxation-controlled	m.s ^{-0.5}
5	transport	
k_{4}	Rate constant for diffusion-controlled	s ⁻¹
	transport	
k_4	Rate constant for diffusion-controlled	$m.s^{-1}$
	transport	
L	Length of stainless steel tube	m
т	Mass	kg
m_s	Initial dry mass of model layers	kg
m_t	Mass in time <i>t</i>	kg
m_{∞}	Mass in the equilibrium state	kg
'n	Mass flow rate of gauging liquid	kg/s
n	Diffusion index	-
ΔP	Differential pressure	Pa
R^2	Coefficient of determination	-
Re_{t}	Reynolds number at nozzle throat	-
$S_{\rm int}$	Internal specific boundary area	m^2
t	Time	S
We	Nozzle rim thickness	m
W _r	Nozzle rim width	m
z	Nozzle-surface clearance	m
Greek		
δ	Layer thickness	m
δ_0	Initial dry layer thickness	m
δ_∞	Final layer thickness	m
3	Water/solvent volume fraction	-
θ	Nozzle converging angle	0
ρ	Density of gauging liquid	kg/m [°]

Acronyms

DAQ Data acquisition

PT Pressure transducer

ZFDG Zero discharging fluid dynamic gauging

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