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EFFECTS OF POLYMER FLOCCULATION UPON SLUDGE DEWATERING IN POTABLE WATER TREATMENT

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ABSTRACT

This study quantifies the effect of polymer flocculation (at the head of the plant) upon the dewatering of sludges generated in conventional drinking water treatment. A rigorous theoretical framework was used to extract two dewatering parameters that fully characterise basic operations from clarification and thickening to centrifugation and filtration.

Coagulation and flocculation were carried out under various conditions representative of full-scale operation. Two high-molecular-mass polymers were used: one weakly cationic, the other weakly anionic.

The results indicate that flocculation has little effect on the equilibrium parameter, 'compressive yield stress', suggesting the internal aggregate structure was unchanged. Moreover, flocculation typically led to a twofold improvement in the dynamics, represented by the 'hindered settling function'.

As industrial processes tend to be rate-limited, the latter enhancement is expected to translate directly into commensurate increases in throughput.

INTRODUCTION

The conventional potable water treatment process is well-established. Two key steps are addition of a precipitating 'coagulant', such as aluminium sulfate ("alum") or a ferric salt, and 'clarification' to separate the water from the resultant sludge, which is usually by gravity settling. The sludge that results from clarification/thickening is a by-product of little value, and is generally viewed as a waste. In recent times financial and environmental impetuses have increased the prevalence and extent of further sludge dewatering — achieved by *e.g.* thickening, centrifugation, or filtration — which maximise water recovery from the sludge, and minimise the amount or volume of sludge to be disposed of or 'reused'.

Since the 1960's it has been common to dose a polymeric flocculant at the head of the plant, usually just after the coagulant, in order to enhance the separation [Young (1968), Bolto (1995), Ogilvie (1997)]. The polymers are intended to combine the coagulated aggregates into larger 'flocs' by a charge-based mechanism or by hydrophobic interaction. The motivation is to improve the kinetics, with enhancements reported for settling [Durand-Piana *et al.* (1987), Dixon *et al.* (2004)] and filtration [Chang *et al.* (1997)] that have been associated with increases in floc size and porosity over polymer-free aggregates. There are suggestions that this may be counteracted by a decrease in the equilibrium compactibility [Chang *et al.* (1997), Dillon (1997), Gregory (1997), Dixon *et al.* (2004)].

The objective of this study was to quantify the effect of polymer flocculation at the head of the plant* upon sludge dewatering.

In contrast to other work, this study used a rigorous theoretical framework [Landman & White (1994, 1997), Bürger *et al.* (2000)] to extract two dewatering parameters — the compressive yield stress and the hindered settling function — that apply to operations from clarification and thickening to centrifugation and filtration [Verrelli (2008)].

The compressive yield stress, P_y, is an equilibrium parameter representing a static mechanical strength. Physically it describes the stress which a given sample can withstand without undergoing dewatering. The hindered settling function, R, is a kinetic parameter representing a hydrodynamic resistance. Physically it is easiest to understand as being inversely related to the sample permeability [Verrelli *et al.* (2009)]. Both of these parameters are material properties which have been shown to depend only upon the state of the material. It is usual to express these parameters as explicit functions of the solids concentration. The fundamental theory itself is written in terms of the volume concentration of solids, or 'solidosity', but in the present work these are converted to solid mass concentrations, C, which are more readily measurable and are more familiar to industry personnel. (It should be noted that dissolved solids are deemed part of the liquid phase.) Hence the dewaterability of a given sludge type is

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* The present work follows scientific convention wherein 'flocculation' refers to the action of the flocculant [La Mer (1964)]; this differs from sometime use in industry, in which the term is intended to mean gentle agitation, and is also distinct from polymer 'conditioning' of thickened sludge. 'Flocs' are the aggregates resulting from flocculation.

completely characterised by $P_y(C)$ and $R(C)$ together. One especially important piece of information contained within that data is the value of the 'gel point', which is the concentration at which the flocs have just approached close enough to form a connected assemblage, or 'network'. This is estimated as the value of C in the limit of P_y approaching zero.

EXPERIMENT

The particulars of the methodology have been set out in detail previously [Verrelli (2008), Verrelli *et al.* (2009, 2010)]. For brevity a more concise description is presented below.

Sludge generation

Coagulation and flocculation were carried out in the laboratory under conditions representative of full-scale operation.

Raw water was obtained from a local reservoir supplying Melbourne, which had relatively low true colour and turbidity. Due to the large volume of water required to produce sufficient sludge for characterisation at low alum doses, it was not practicable to use the same raw water sample in each case. Key data for the specific samples of interest are presented in **Table 2**.

The water was treated in a 48 litre baffled tank that was mixed using a Rushton turbine impeller, following the 'standard' configuration of Holland & Chapman [Holland & Chapman (1966)]. The basic protocol was to apply rapid mixing for the first two minutes to disperse the dosed chemicals throughout the volume, and then to mix more gently for a further 20 minutes in order to promote particle aggregation. The mixing conditions are summarised in **Table 1** in terms of the characteristic velocity gradient, G, and the Reynolds number, Re.

Each of the samples was first dosed with alum at one of two doses, 5 or 79 mg(Al)/L, referred to as "low" and "high" respectively. After 60 seconds of mixing sodium hydroxide was added to attain the target pH, which was 6.0 for all cases. After a further 30 seconds a flocculant was added.

Two polymers of high molecular mass were used:

- *Zetag 7623* — a 10% cationic copolymer of acrylamide and a quaternary ammonium salt, and
- *Magnafloc 338* — a 10% anionic copolymer of acrylamide and sodium acrylate.

The concentration of the solution added was 1000 mg/L, made up from refrigerated stock solution (5 g/L) and well mixed to ensure homogeneity. The flocculant dose for the 79 mg(Al)/L sludges was 1.23 mg(flocculant)/L, expected to be equivalent to approximately 3.5 kg(flocculant)/t(dry solids), roughly in line with conventional recommendations [Berné & Richard (1991), Blakemore *et al.* (1998), Kawamura (2000), Crittenden *et al.* (2005)]. For the 5 mg(Al)/L sludges the flocculant dose was reduced to 0.62 mg(flocculant)/L, due to the decreased amount of dry solids resulting from coagulation, although this still yielded a rather high ratio of 26 kg(flocculant)/t(dry solids). Further details of the sample generation conditions are presented in **Table 2**.

After flocculant addition a further 30 seconds of rapid mixing was applied. This is shown in **Table 1**.

Flocs were allowed to settle in the tank overnight to form a shallow layer of sludge at the base. The supernatant was decanted and the sludge recovered for characterisation of its dewaterability.

Table 1: Standard dosing and mixing sequence.

Time [min:s]	Chemical	G [1/s]	Re [–]
<0:00	–	254	41000
0:00	$Al_2(SO_4)_3$	254	41000
1:00	NaOH	254	41000
1:30	flocculant	254	41000
2:00	–	13	6000
>22:00	–	~0	~0

Dewaterability assessment

Sludge dewaterability was assessed in two regimes of C. At low C the sludge was allowed to settle under gravity, from an initially homogeneous state, in glass measuring cylinders. The decline of the sludge bed over time was monitored, and the initial and equilibrium values of C measured. By application of a numerical solution algorithm [Lester *et al.* (2005)] $P_y(C)$ and $R(C)$ data were extracted.

To obtain high-C data the sludge was filled into a customised dead-end filtration rig and subjected to a sequence of constant stresses [de Kretser *et al.* (2001)]. The consolidation rate was monitored electronically, and again the initial and equilibrium values of C were measured. Whereas a continuously varying profile of C with height is obtained at equilibrium in gravity settling, in filtration a uniform C is attained at equilibrium, so extraction of $P_y(C)$ and $R(C)$ data required only simple mathematics.

For $P_y(C)$ a reasonable estimation of the values at intermediate C can be obtained by interpolation. Typically the shape of the $R(C)$ curve makes interpolation more difficult for this parameter.

Table 2: Generation conditions for alum sludge samples used to investigate flocculation.

Sample:	High	High + M.	High + Z.	Low	Low + M.
Raw water properties					
Raw water collection date	2006-01-05	2006-01-05	2006-01-05	2004-01-08	2005-11-16
True colour [mg/L Pt units]	22	22	22	10	27
Absorbance at 254 nm [10/m]	1.14	1.14	1.14	0.66	1.23
Total solids [g/kg]	0.05	0.05	0.05	0.08	0.07
Total dissolved solids [g/kg]	0.05	0.05	0.05	0.07	0.07
Treatment conditions					
Coagulant dose [mg(Al)/L]	79	79	79	5	5
Coagulation pH [–]	6.1	6.0	6.0	6.0	6.0
Flocculant	None	*Magnafloc 338*	*Zetag 7623*	None	*Magnafloc 338*
Flocculant dose				–	
[mg/L(mixture)]	–	1.23	1.23	–	0.62
[g/kg(dry solid)]	–	3.4	3.4	–	26

DISCUSSION AND RESULT ANALYSIS

Dewatering parameters

On the basis of published articles, the dosing of flocculant shortly after coagulant and alkali addition is expected to lead to faster dewatering, at the expense of a reduced maximum extent.

Experimental results systematically comparing the effects of flocculant addition upon dewatering properties are presented in **Figure 1** (compressive yield stress) and **Figure 2** (hindered settling function).

The similarity of the compressive yield stresses (**Figure 1**) for the three high-alum-dose sludges is remarkable. In fact, it surpasses the concordance previously found between even duplicate runs of a given material [Verrelli (2008)]. It strongly suggests that across all concentrations the flocculant has nil effect on the equilibrium condition.

The $P_y(C)$ data for the two low-alum-dose sludges shows more variation, with the flocculated sample exhibiting marginally weaker behaviour in filtration, but a lower gel point. Although this might be due wholly to the flocculant, it is likely that the different nature of the two raw waters has also affected the results. This premise is based firstly on the lack of observable difference in the high-alum-dose sludges. Secondly, it is based on the inter-relation between alum dose and raw water NOM (natural organic matter), indicated in **Table 2** by the surrogates true colour and ultraviolet absorbance. Reduction in NOM and increase in coagulant dose could be expected to yield similar results, namely a greater influence of precipitated coagulant on the sludge properties — although experimental results showed the physical consequences to be more complicated [Verrelli *et al.* (2009)]. Here the unflocculated low-dose sludge was generated from relatively uncoloured raw water, so that the precipitated coagulant makes a greater contribution to the sludge composition. This may be reflected in the similarity between the $P_y(C)$ curve for this sludge and for the three high-alum-dose sludges. This inference is consistent with observations previously reported for this system, which established that — in the absence of flocculant — precipitated coagulant controls sludge dewaterability for alum doses above 5 to 10 mg(Al)/L [Verrelli (2008), Verrelli *et al.* (2009)].

There are differences in the $R(C)$ curves shown in **Figure 2**. There is a small but fairly consistent tendency for the flocculated sludges to have reduced values of R, implying less resistance to dewatering. This is the trend that would generally be expected (if any). The trend is consistent across the two types of flocculant used, and across the range of observed C, except for the portion of the low-alum-dose curves at low concentration.

For the gravity settling tests, the initial concentration loaded was subject to significant uncertainty for two of the three high-dose tests, and so the differences at low C cannot necessarily be relied upon. Nevertheless, the excellent concordance of the $P_y(C)$ curves, even at low C, suggests that the estimates used were good, despite the experimental uncertainty. (Uncertainty in initial C for the low-alum-dose sludges was negligible.)

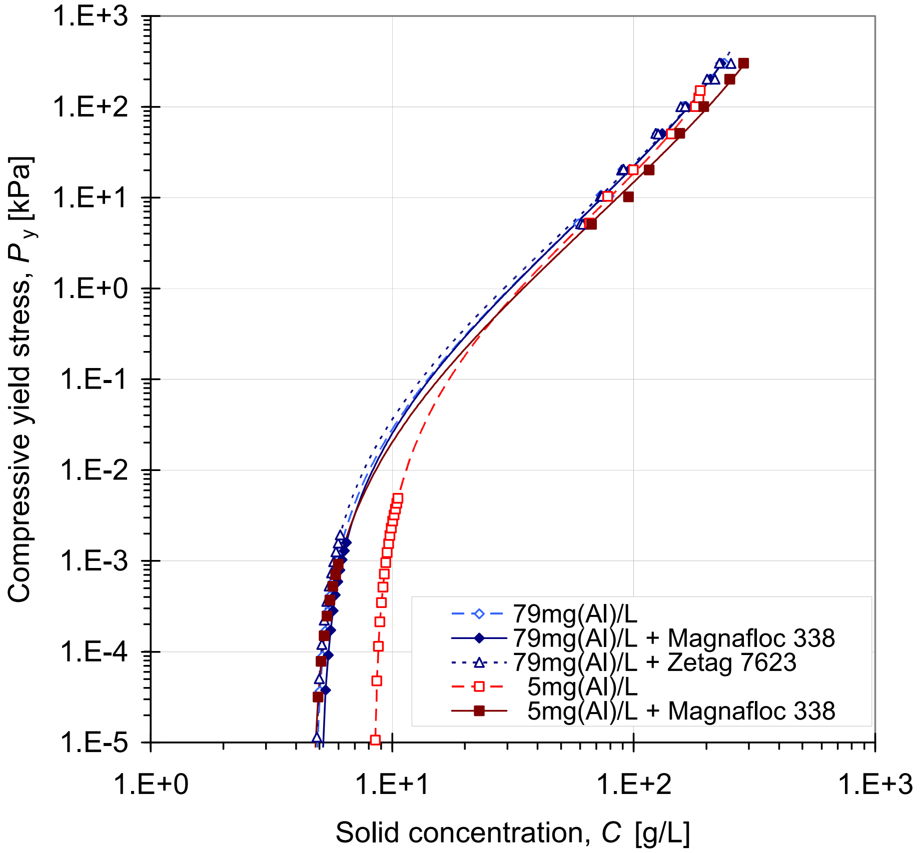

Figure 1: Compressive yield stress for two sets of sludges where flocculants were added to promote aggregation, compared to 'controls' with no flocculant.

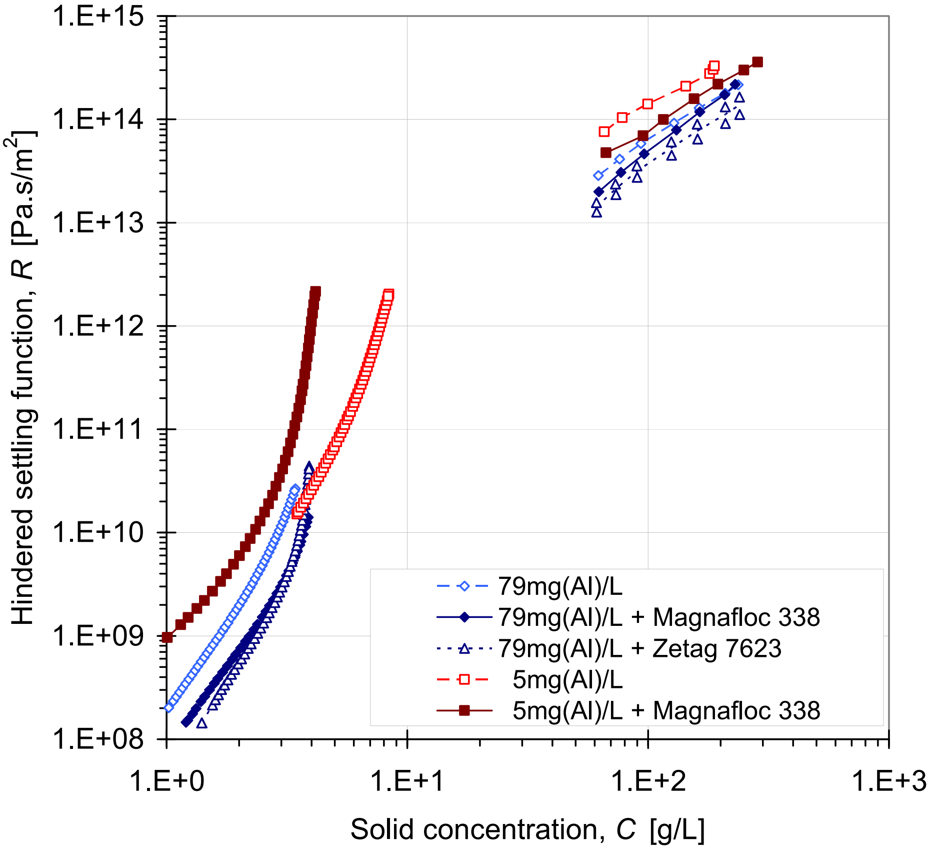

Figure 2: Hindered settling function for two sets of sludges where flocculants were added to promote aggregation, compared to 'controls' with no flocculant.

Physical mechanism

It was observed that for the most part the addition of flocculant did not affect the equilibrium condition, even though the dewatering kinetics changed significantly. These effects are characteristic of certain physical changes in the nature of the flocs. It is of interest to assess the physical mechanisms by which flocculation might be accountable for such changes.

The conundrum faced is that the unaffected $P_y(C)$ seems to suggest an unchanged internal structure of the particulate network and constituent aggregates, while some sort of structural change is necessary to explain the change in $R(C)$. Based on the experimental results, the two options are that the structural changes causing the reduction in $R(C)$ either cause mutually cancelling changes in $P_y(C)$ or directly cause a negligible change in $P_y(C)$. With regard to the former option, an example might be increased strength within an aggregate balanced by decreased strength between aggregates.

An illustrative example of the latter option is presented in **Figure 3**. For this simplified arrangement the solid concentrations, C, of each system are identical. The vertical resistances to mechanical stress are also the same, to a first approximation. (In practice the presence of lateral restraints helps to support 'columns' of primary particles oriented in the major principle stress direction against buckling [Adams *et al.* (1994), Oda *et al.* (1998), Muthuswamy *et al.* (2006)] [*cf.* Trappe *et al.* (2001)].) However, applying a Poiseuille-like analysis, the resistance to flow in configuration "a" is much greater than that in configuration "b". Application of Ockham's razor suggests that this simpler alternative is to be preferred over the more complex mode that requires maintenance of a perfect balance of two countervailing effects across almost two orders of magnitude in C.

In the particular form of the illustration chosen, the aggregates in each system have the same properties, including the same inherent strength. The difference between the two systems lies only in the packing of the aggregates, or flocs. A mechanism of this nature is compatible with the experimental protocol adopted in the present work. Two key considerations are the timeline of floc formation, and the persistence of macroscopic structure.

The experimental protocol (**Table 1**) dictates that the alum had time to dissociate, hydrolyse, and start to adsorb and precipitate before the flocculant was added to the tank. Those early stages in the coagulation process are sufficiently fast to have developed significantly even in that short space of time [Stol *et al.* (1976), Livage *et al.* (1988), Nordstrom & May (1996)]. (Flocculation kinetics can be quite slow in dilute systems [Gregory (1988)], although the long slow-mix period provides ample time for eventual action.) It is implausible that flocculant would be able to penetrate pre-existing particle clusters to any great extent.

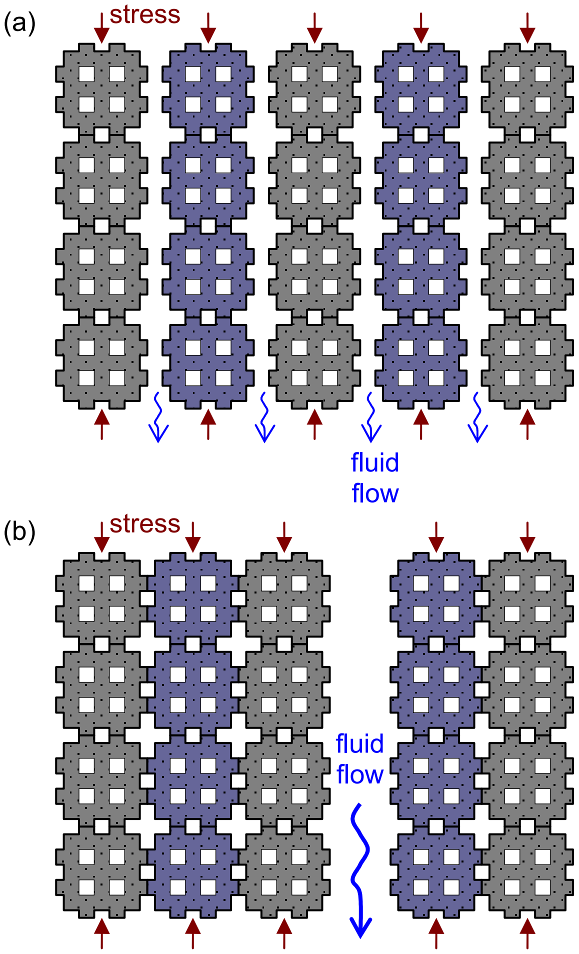

Figure 3: Schematic illustration of the effect of macroscopic sludge structure on strength and hydrodynamics.
(a) Uniformly spaced stacks of aggregates.
(b) Unevenly spaced stacks of aggregates.

For the given model, persistence of macroscopic structure is required because differences in R are still seen at high values of C, which could only be reached after the sludge has been progressively dewatered into a reasonably firm cake. A simple argument in favour of such persistence is that major flow paths can be self-preserving. Numerous reports on settling systems have described the evolution of major flow paths, wherein the 'scouring' action of the rapid fluid flow widens, straightens and maintains the channels [Michaels & Bolger (1962), Poon *et al.* (1999), Starrs *et al.* (2002)]. Such action could carry on into regimes of higher C, such as in filtration, although this has not been confirmed through direct observation.

The logical synthesis of the foregoing arguments is that the flocculant is predominantly to be found on the periphery of particle clusters, aggregates, or flocs, and that the resultant alteration of the external surface affects the binding ability of the objects, and hence the packing efficiency, thereby modifying the characteristic hindered settling function associated with the particulate system at any given C.

It should be mentioned that in the free settling regime P_y is identically zero. This low-C regime was not probed in the present work. Anecdotal reports indicate that addition of flocculant allows the formation of flocs that are larger than the aggregates formed by coagulation alone, although the correlation is less clear in some experimental studies. The initial formation of larger flocs is consistent with the proposed theory, with flocculant acting to bind pre-existing aggregates together in the low-C regime. At low C larger flocs would settle faster than smaller aggregates with the same compactness, because the weight grows faster than the drag[†]. Although the size of primary particles is known to affect the compressive yield stress of certain model gels [Miller *et al.* (1996)], it can be expected that P_y will be insensitive to floc size *per se*. This may be seen by recognising that P_y only takes non-zero values above the gel point, at which concentrations all of the constituent flocs are connected anyway, forming what is commonly called a sludge 'bed' or a sludge 'cake', but which for the purposes of the present discussion could be termed a 'super floc'.

Industrial implications

Although the sludges were processed in two very specific operations to obtain the $P_y(C)$ and $R(C)$ data, these two parameters are material properties, and thus do not depend on the type of dewatering operation used to obtain the estimates. (Notwithstanding the fact that due care must be taken to assure the accuracy of the estimates.) As the trends in P_y and R were consistent across the range of C investigated, the dewaterability observations are relevant to a wide range of industrial unit operations including thickening, centrifugation and filtration.

An important characteristic of the industrial operation of dewatering processes is that they tend to be rate-limited [Verrelli *et al.* (2010)]. What this means is simply that the sludge is not exposed to the dewatering 'force' for long enough to come to equilibrium. Instead, the dewatering is terminated at a state dictated by the available time and the dewatering rate. Thus, although $P_y(C)$ did not seem to be affected by flocculation, the results obtained herein indicate that prior flocculation would result in a greater extent of dewatering in practice. That is, a larger C would be attained over the same period of time. Alternatively, the decrease in resistance, $R(C)$, could be exploited to enhance throughput by reducing the processing time — equivalent to increasing the flowrate for continuous processes — to achieve the same output C.

One of the interesting features of the present findings is that the dewaterability enhancement due to flocculation at the head of the plant would carry through to all subsequent stages of dewatering. This suggestion needs to be tempered with a recognition of additional factors that become important on a full-scale plant. Of particular relevance is the potential for flocs to have significant exposure to high shear flows in the pipework and channels between unit operations. Separate experimental work has indicated that exposure to high shear can disrupt aggregate structure down to a threshold lengthscale, so that dewatering occurring up to a limiting degree of compaction is adversely affected [Verrelli (2008)].

As a consequence of such practical issues, further doses of a 'conditioning' polymer are often implemented immediately upstream of a dewatering device. Such conditioning of settled/thickened sludge further downstream has previously been reported to have a greater effect on dewaterability than flocculation does [Young (1968)], although the basis for comparison may not be 'fair'. The effects of polymer conditioning were not considered herein; an experimental evaluation has been reported elsewhere [Verrelli (2008)].

In all industrial settings a complete assessment of the potential advantages of flocculant dosing needs to encompass both downstream benefits due to improved dewaterability and an evaluation of upstream operating costs, including chemical purchase costs. Due to regional variation and site-specific factors, the best solution may vary from plant to plant.

[†] Even though water treatment flocs have an approximately fractal structure [Lagvankar & Gemmell (1968), Tambo & Watanabe (1979)], the rule is still expected to apply in practice.

CONCLUSION

Measurements of compressive yield stress, which characterises the maximum (equilibrium) extent of dewatering for a given stress, showed no significant difference between sludges generated with and without flocculant. Measurements of the hindered settling function, which is inversely related to permeability, showed a clear difference between the two sets of sludges. They differed typically by a factor of two, in favour of the flocculated samples, across the full range of concentrations (*i.e.* for all of the unit operations) for each of the flocculants.

Physically these results suggest that, in forming flocs, the polymers attached to the outside of the coagulated aggregates, or particle clusters, and did not affect their internal structure. Instead, the packing of flocs was altered. In contrast to previous reports, the results show improvement in dewatering dynamics with no significant degradation of the equilibrium state.

In industrial dewatering processes, which are generally rate-limited, flocculation at the head of the plant can be expected to result in increased throughput, following directly from the improvements in $R(C)$.

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