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Recovery and Concentration of Buoyant Cenospheres using an Inverted Reflux Classifier


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Abstract

Cenospheres are hollow, low-density particles found in power station fly ash. They have many commercially-useful properties which make them a valuable by-product. However, recovering cenospheres from fly ash is difficult due to their low concentration and fine size. Experiments were performed to test the novel approach of using an Inverted Reflux Classifier. In this configuration, the particles are fluidised by adding wash water from above which helps to wash any entrained dense material from the overhead product. Inclined channels are mounted at the base to minimise the loss of buoyant cenospheres in the waste underflow stream.

Experiments were performed at both laboratory scale (80 mm × 100 mm cross-section) and pilot scale (300 mm × 300 mm cross section) using mixtures of cenospheres and silica, all nominally less than 100 μm in size. In batch tests, the bed expansion behaviour of the positively-buoyant cenospheres in the Inverted Reflux Classifier was found to be analogous to the behaviour of negatively-buoyant particles in the standard configuration. Continuous steady-state experiments were performed using feeds with suspension solids concentration varying from 0.3 to 9.5 wt% solids and a buoyant cenosphere grade of 0.5 to 65 wt%, with a range of fluidisation wash water rates, and degree of volume reduction (ratio of volumetric feed to product rate). Both units delivered high recoveries and product grades. An increase in volume reduction (decreasing overflow rate for a given feed rate), caused a drop in recovery and an improvement in grade. The throughput advantage compared to a conventional teetered (fluidised) bed separator was over 30 in some cases. Both laboratory and pilot-scale units displayed similar behaviour and the results were also consistent with existing correlations for negatively-buoyant particles in the standard Reflux Classifier. Hence this technology has clear potential for recovering and concentrating cenospheres from fly ash.

Key Words:

Inverted Reflux Classifier; Cenospheres; Fly ash; Density separation; Buoyant particles.
1.0 Introduction

Cenospheres are hollow spherical alumina-silicate particles formed during coal combustion. The effective density of individual cenospheres can vary from that of silica (~2600 kg/m³) down to 200 kg/m³ or even lower, depending on the number and size of the gas inclusions in each particle [1]. However, in this work we use the term “cenosphere” to refer to only those particles with an effective density less than water i.e. specific gravity less than 1.0. Cenospheres are free flowing, strong, low in density and have low thermal and electrical conductivity. These properties make them a valuable by-product which is finding increasing use in a range of industries, such as aero-space, building and chemical, where they provide advantages such as low weight, increased heat resistance, better electrical insulation, low shrinkage, microwave shielding and reduced water absorption [e.g. 2-7]. Therefore, the recovery of cenospheres from power station fly ash is attracting growing interest, with various wet and dry processes having been trialled [e.g. 8-12].

There are some major challenges in efficiently recovering cenospheres. Their surface, magnetic and electrical properties are similar to many of the other components found in fly ash. Thus methods such as flotation and electrostatic separation that are routinely used in mineral processing for separating fine particles, and have been successfully applied to remove combustible material from fly ash, cannot be effectively applied to recovering cenospheres. Gurupira et al. [10] do report using a triboelectric separator to recover cenospheres from fly ash, but rely on a definition of cenosphere as particles of specific gravity less than 2.0, whereas most commercial interest is focused on cenospheres with specific gravity less than 1.0, Hence, the only practical method available appears to be density-based separation. However, cenospheres are generally present at low concentrations, typically 0.01 to 5 wt %, and are small in size, typically in the range 50 to 250 μm diameter [13]. Gravity-based separation processes struggle to perform well down in this size range [e.g. 14].

It is in theory simple to separate positively buoyant cenospheres from the denser components of fly ash by float-sink separation in water. Indeed the usual approach to producing commercial grade product involves scooping the buoyant cenospheres from the surface of fly ash tailings ponds [9]. However, this requires large holding ponds, results in significant losses and the product includes a high level of contamination, so it needs further upgrade. Attempts to separate cenospheres via a conventional elutriation device inevitably also entrain a significant amount of high density silica slimes in the overhead product.

The objective of the present study is to evaluate the potential for an Inverted Reflux Classifier (IRC) to efficiently separate and recover cenospheres from fly ash. The standard Reflux Classifier (RC) is a separation device that consists of a fluidised bed with a set of parallel inclined channels mounted
above (Figure 1a). The inclined channels provide a much larger effective settling area than a standard teetered (fluidised) bed separator due to the Boycott effect [15]. The selective resuspension of low-density particles that settle on the channel walls suppresses the effects of particle size on the separation, thus making the separation more sensitive to density difference [16]. Recent work has further shown that when narrow channels are used to promote laminar flow with high shear rates, the separation becomes almost entirely based on density difference [17-18], especially in the intermediate range of particle Reynolds numbers (1 < Re < 500).

Figure 1: Diagram of the (a) standard and (b) Inverted Reflux Classifier.

The standard Reflux Classifier design has been in commercial use for about 8 years, initially in the coal industry but increasingly also in the mineral processing industry, where it is used to separate valuable material from gangue based on differences in their particle density [19]. In all the commercial applications to date, the particle species being separated have both been denser than the water used to fluidise the unit. However, cenospheres are buoyant in water, and only present at low concentrations. Thus their recovery and concentration from fly ash in a standard RC would result in an unacceptably high level of ultrafine dense silica being entrained in the overhead product stream. So instead, it was decided to trial an alternative approach of inverting the entire apparatus so that the fluidising water is added from above, and the inclined channels are located at the base (Figure 1b). The downward flow of fluidising (or wash) water near the exit at the top should help prevent the elutriation of dense silica particles in the overhead product, whilst the enhanced
effective “settling” area of the inclined channels at the base should help prevent the buoyant cenospheres being lost in the underflow. This is a novel application of the Reflux Classifier concept. The focus of this paper is on preliminary experiments designed to test its potential.

The background theory of how standard Reflux Classifiers work is outlined in Section 2. Section 3 gives details of the experimental methodology of the present study. There were two stages in the work. Firstly, laboratory-scale experiments were performed in a unit with a horizontal cross-sectional area of 80 mm × 100 mm. Then pilot-scale experiments were performed in a unit with cross-sectional area of 300 mm × 300 mm. The results of these experiments are presented and discussed in Section 4, with final conclusions in Section 5.

2.0 Theory

This paper is focussed on the recovery of positively buoyant cenospheres that float, whereas existing theory was developed for negatively buoyant particles that settle. So in this section the existing theory is summarised, but then some further comments are provided on potential additional factors that may come into play when separating buoyant cenospheres from negatively-buoyant particles.

For fine particles, the terminal free settling velocity \( u_s \) of a spherical particle of diameter \( d \) can be predicted using Stokes law:

\[
    u_s = \frac{d^2 (\rho_p - \rho_f)g}{18 \mu}
\]  

Here \( \rho_p \) and \( \rho_f \) are the particle and fluid densities respectively, \( \mu \) is the fluid dynamic viscosity and \( g \) is the gravitational acceleration. The correct particle density to use is its apparent density i.e. the total mass of the particle divided by the total occupied volume including any gas cavities. For cenospheres, this value can be anywhere from 1000 down to 200 kg/m\(^3\) or even lower. For positively buoyant particles where \( \rho_p < \rho_f \) the terminal free “settling” velocity predicted by Eq. (1) is negative, indicating that particles rise in the upwards direction.

When a suspension of many particles is present, their settling (or rise) velocity is lower. The semi-empirical Richardson and Zaki [20] equation describes this effect for a single-component suspension:

\[
    u_h = u_s (1 - \phi_s)^n
\]  

where \( u_h \) is the hindered settling velocity of particles with a terminal free settling velocity \( u_s \), when present in a suspension with a solids volume fraction \( \phi_s \). In Stokes settling regime \( n = 4.65 \).
Within an inclined section, particles have only a relatively short vertical distance to fall before they settle against the lower surface of the channel, from where they can slide down to the base of the channel. In the case of buoyant particles such as cenospheres, they will instead rise to the upper (downward facing) surface of the channel and then slide up to the top. This so-called Boycott effect [15] produces an increase in the effective settling area compared to a purely vertical column with the same footprint. Hence parallel inclined channels are beneficial in lamellae thickeners [21], offering very high hydraulic loadings. In lamellae thickeners the objective is to simply capture all of the solid, often flocculated, particles onto the inclined surfaces, allowing clear supernatant to pass through. We may define the \textit{throughput advantage}, $F$, of inclined channels as the ratio of the superficial upwards velocity of the fluid in the vertical section, $U$, divided by the terminal free settling velocity, $u_t$, of the largest particle that can in principle be elutriated from the inclined channels assuming that the solids concentration is very low:

$$ F = \frac{U}{u_t} \quad (3) $$

The well-known PNK theory [22, 23] predicts the increase in the hydraulic capacity of inclined channels over conventional thickeners. For plug flow and with the assumption that any particle that settles onto the lower channel surface will slide back down into the fluidized chamber, the PNK theory predicts a theoretical throughput advantage of [16]:

$$ F_{\text{theory}} = 1 + \left( \frac{L}{z} \right) \cos \theta \sin \theta \quad (4) $$

Here, $\theta$ is the angle of inclination relative to the horizontal, $L$ is the channel length and $z$ is the channel width perpendicular to the direction of fluid flow (see Figure 2). The group $L/z$ is termed the \textit{aspect ratio} of the channels.
Equation 4 predicts that $F$ should increase indefinitely as $L/z$ increases. However, in reality the throughput advantage does not increase indefinitely because particles that settle onto the incline do not always slide all the way back into the vertical section (or rise to the top in the case of cenospheres). Re-suspension may occur due to fluid shear, turbulent mixing, etc. The segregation efficiency, $\eta$, is defined as the ratio of the actual to the theoretical throughput advantages:

\[
\eta = \frac{F}{F_{\text{theory}}} = \frac{U/u_i}{1 + \left(\frac{L}{z}\right) \cos \theta \sin \theta}
\]

(5)

Laskovski et al. [16] fitted the following empirical expression to the segregation efficiency obtained in batch Reflux Classifier experiments:

\[
\eta_{\text{pred}} = \frac{1}{1 + 0.133 \cos \theta Re_{\text{l}}^{1/3} (L/z)}
\]

(6)

where $Re_l = \rho u d / \mu$ is the particle settling Reynolds number. The relevant particle diameter used to calculate $Re_l$ corresponds to the particles that have a partition probability of 50 %, $d_{50}$.

Although the re-suspension phenomenon reduces the throughput advantage, the selectivity on the basis of particle density is improved. Particles with a density far from the fluid density are less likely to be re-suspended than particles with a density close to that of the fluid. Therefore the efficiency of density separation is higher than in a purely vertical elutriation column.
Most of the experiments of Laskovski et al. [16] involved relatively large channel spacings where the flow exhibited some turbulence. More recent work with narrow channels has found that laminar flow is even more beneficial for promoting density separation [17]. In laminar channel flow of average velocity $U'$, the local fluid velocity $u$ at a distance $d/2$ from the wall in a channel of spacing $z$ is given by [24]:

$$u = \frac{3}{2} \frac{U'}{z} \left( 2 - \frac{d}{z} \right) \lim_{d/z \to 0} \frac{3U'd}{z}$$

Hence in the limit for particles that are small compared to the channel thickness ($d/z \to 0$), the conveying velocity experienced by a particle becomes proportional to its size ($u = k \cdot d$). Hence large particles experience a greater upwards force than small particles. The critical condition is assumed to occur when the downwards component of a particle’s settling velocity parallel to the channel surface $u'_t = u_s \sin \theta$ equals the upwards component $u$ of the local fluid velocity. Friction and hydrodynamic resistance associated with the wall are neglected here. Figure 3 illustrates this phenomenon.

![Figure 3: The parabolic velocity profile in laminar channel flow results in a near-linear velocity profile $u$ experienced by particles at the channel wall. Hence larger particles experience a proportionally larger local fluid velocity. $u'_t = u_s \sin \theta$ is the component of terminal settling velocity](image)
parallel to the direction of fluid motion. The critical condition occurs when \( u = u_t' \) (adapted from [17]).

Hence for particles in the Stokes settling regime (Equation 1), the effect of particle size on elutriation velocity is reduced from \( d^2 \) to a \( d^1 \) dependency. This model gives an excellent match with experimental elutriation results [17]. The most recent theoretical model also includes consideration of the shear-induced lift force on the particles, which gave an even closer match to the observed experimental data [18].

All of the above theory for negatively buoyant particles in the standard Reflux Classifier (Figure 4a) should be equally applicable to the Inverted Reflux Classifier when used with only buoyant particles (Figure 4b). In both these cases, all of the particle species present tend to rise or settle in the same direction. However, in the current work on separating cenospheres from silica, both positively and negatively buoyant particles are present simultaneously, and thus there will be layers of particles forming on both the upper and lower surfaces of each inclined channel (Figure 4c). Thus there is potential for interaction that may cause different behaviour to that predicted by the existing theory.

![Figure 4](image_url)

**Figure 4:** Flow patterns of negatively (●) and positively (○) buoyant particles in the (a) standard and (b & c) Inverted Reflux Classifier configuration.

Intuitively it might be expected that the interactions between upwards rising positively buoyant particles and downwards settling negatively-buoyant particles would always lead to increased hindered settling and thus a reduced efficiency of separation. However, this is not always the case. The first suggestion of this was in work by Whitmore [25] using neutrally buoyant particles. Weiland and MacPherson [26] extended this using mixtures of positively and negatively buoyant particles.
They found that this could lead to an accelerated settling rate of both species. This was caused by an initial lateral movement of particles such that the component present at the lowest volumetric concentration formed clusters or vertical fingers (streams) within a suspension of the other component. These streams then experienced a buoyancy-driven convective flow which enhanced the particle settling rate. Reasonably accurate models have been developed to predict the settling rates with and without streaming [27-29].

A certain minimum critical concentration is required before streaming behaviour occurs. Below this limit, the presence of a second species does indeed reduce the settling rates [28]. For systems with roughly equal volume fractions of both species, streaming has only been observed in systems with total solids volume fractions greater than 15 vol.% [30-31]. If one species is only present at low concentrations, then much higher solids volume fractions are required before streaming is initiated. There has been both analytical modelling and numerical simulation work on the flow stability of mixtures of particles driven in opposite directions, which shows some promise to be able to predict which regime will occur [30,32]. Given the fact that the buoyant cenospheres are present at such low concentrations in fly ash, it might be questioned whether this phenomenon is relevant to the operation of the Inverted Reflux Classifier. However, at steady state the concentration of cenospheres inside the unit can build up to much higher levels than in the feed, due to the refluxing action of the inclined channels and the downwards flow of the wash water producing hold up of particles inside the unit.

3.0 Materials & Methods

3.1 Materials

Due to supply availability, two different grades of Q-CEL Hollow Microspheres (Potters Industries) were used in this work. According to the manufacturer’s specifications, these both had a nominal size range of 5 – 90 μm and “mean size” of 45 μm (as measured by laser light scattering). The 7040S grade of microspheres used in the laboratory-scale work was quoted as having a typical effective density of 430 kg/m³ (measured by liquid displacement). The 5020FPS grade microspheres used in the pilot-scale work had a quoted typical density of 210 kg/m³. Volume frequency size distributions of these particles were measured using a Malvern Mastersizer 2000, and are shown in Figure 5. The volume arithmetic mean and Sauter mean sizes were 39 and 24 μm for the 7040S grade and 31 and 19 μm for the 5020FPS grade respectively. There is a clear bi-modality, with the main peak near 25 μm, and a smaller peak at 4 μm.
Figure 5: Volume frequency size distributions of the as-supplied hollow microspheres and silica flour, as measured by a Malvern Mastersizer 2000.

Not only do these commercial microspheres vary in size, but they also vary in density. Float-sink separation in water showed that around 35% of the particles in both grades were denser than water. For the 5020FPS grade, the average densities of the floats and sinks fractions were found by water pycnometry to be 400 kg/m$^3$ and 2410 kg/m$^3$ respectively (significantly higher than the manufacturer's specification of 210 kg/m$^3$). Malvern laser size analysis showed that the sinks fraction contained particles with a peak size at 20 μm, compared to the floats fraction with a peak at a larger size of 50 μm (Section 4). So whilst the two grades are clearly different, they cover a similar size range and have similarly shaped size distributions.

For some of the later laboratory-scale experiments, the concentration (grade) of cenospheres in the feed was lowered to make it similar to that of raw fly ash. This was done by adding silica flour (Sibelco, 400 G) as the gangue material. The manufacturer's specification sheet quotes a typical silica flour density of 2670 kg/m$^3$ with 98% in the size range 1 to 45 μm. Its size distribution is also shown in Figure 5.

3.2 Equipment

3.2.1 Laboratory-scale

The laboratory-scale Inverted Reflux Classifier was built from Perspex. The upper section was a vertical column with internal cross-sectional area of $B = 80$ mm and $W = 100$ mm, and a height of 1000 mm. The lower section had a length of $L = 1000$ mm inclined at an angle of $\theta = 70^\circ$ to the
horizontal. Inside the inclined section there were 7 parallel, equally-spaced stainless-steel plates of thickness $t = 0.7$ mm. This created 8 channels of perpendicular spacing $z = 8.8$ mm (see Figure 2 for definitions of the key dimensions).

There was a pair of pressure tappings spaced 475 mm apart set in the top half of the vertical section. These were connected to a differential pressure transducer, thus providing pressure gradient data for calculating the overall solids volume fraction and monitoring whether the system was at steady state.

Fluidising (wash) water was supplied from the mains water supply and the flowrate measured using a rotameter. It entered through a pyramid shaped distributor. The inner chamber of this distributor had internal dimensions of 100 mm x 80 mm at the base (to match the vertical section), was 110 mm in height and narrowed to a 20 mm ID outlet at the top. There were $56 \times \varnothing 1$ mm holes distributed evenly over the sloped walls. Four hoses (one on each side) were connected externally to supply fluidising water to the outer chamber which would then flow into the main chamber via these 52 holes.

In continuous experiments, feed was prepared in a stirred 300 L tank and pumped into the unit at a height 370 mm above the entrance to the inclined channels. The underflow was pumped out the base at a controlled rate and the overflow exited at the top of the column (Figure 6).
Figure 6: Schematic of the set up used for the continuous experiments (not drawn to scale). Experiments were run in closed-loop configuration, so the underflow and reject were returned to the feed tank. Some water needed to be discarded from the system to balance the addition of fluidization water. In the laboratory-scale work, the discard water was removed from beneath a distributor at the base (a), whereas in the pilot-plant scale, it was removed at a Y junction in the underflow line (b) and passed through a small lamella thickener to minimise loss of solids from the system.

Except when samples were being collected, both the overflow and underflow streams were directed back into the feed tank in order to provide enough feed for long continuous experiments to be performed. To operate in this closed-loop manner, it was necessary to remove water at the same rate as fluidising wash water was being added, otherwise the feed would gradually become more dilute and the feed tank would eventually overflow. Hence a discard water stream was removed via a distributor plate located at the base of the unit. This distributor had the same dimensions as the one at the top of the system. The small holes helped to reduce the rate of loss of solids in this stream, but this was not perfect and some solids were observed in the collected discard water.

3.2.2 Pilot-scale

The pilot scale Inverted Reflux Classifier was built from stainless steel. It had a square cross-sectional area of 300 mm × 300 mm. The height of the vertical section was 2.7 m. There were 38 inclined channels with a nominal channel spacing of \( z = 6 \) mm, length \( L = 1.2 \) m at an angle of \( \theta = 70^\circ \) to the horizontal.

The fluidising water was again taken directly from the mains water supply. Feed was prepared in a 1300 L stirred tank. The feed was pumped into the unit about 500 mm above the entry to the inclined channels. Underflow was pumped out the base and the overflow exited at the top of the column.

Similar to the laboratory-scale rig, the underflow and overflow streams were directed back into the feed tank except when samples were being collected. In order to discard water to compensate for the added fluidisation wash water, a side stream was removed from the underflow and passed upwards through a lamella thickener to minimise loss of any solids in the discard water stream. This lamella thickener (visible in Figure 7) had a cross-sectional area of 80 mm × 100 mm, with 23 stainless steel plates creating 24 channels with a perpendicular spacing of about 2.6 mm in each one. This was much more effective at removing solids from the discard water than the method used in
the laboratory scale unit. However, a small quantity of solids was still observed in the collected discard water.

3.3 Method

3.3.1 Bed Expansion Experiment

Doroodchi et al. [34] performed batch experiments with negatively-buoyant particles in a standard Reflux Classifier. With increasing fluidisation rate, the bed expanded until it reached the base of the inclined channels. After that there was very little bed expansion with further increases in fluidisation rate because the particles started to be captured by the inclined channels and were returned to the bed. Some preliminary experiments were performed to measure the expansion characteristics of a bed of cenospheres in the Inverted Reflux Classifier to test whether analogous behaviour occurred. A 1.0 kg batch of cenospheres was charged into the laboratory-scale column, then water was added and the cenospheres fluidised from above at 500 mL/min for 2.5 h. Another 1.0 kg was then added to the column and the combined material fluidised at 500 mL/min for another 2.5 h. This procedure was intended to elutriate the ultrafines and any non-buoyant material to obtain a more uniform mono-disperse suspension for performing bed expansion experiments. A total of 1.213 kg of ultrafine elutriated underflow solids were collected, so it is estimated that 0.787 kg of larger buoyant material remained in the column.

Samples of the floats solids left in the bed after this time were measured by water pycnometry and found to have an average density of 315 kg/m$^3$. Malvern size measurement showed that 90 % of the volume was in the particle size range 40 ±20 μm. The depth of the “settled” layer of floating cenospheres was 0.738 m.

Then a series of bed expansion experiments was performed. In these batch experiments, there was no feed or overflow stream. Fluidising water was passed down through the column at a known rate. When conditions were steady, the bed pressure drop was measured. This value was used to estimate the solid volume fraction (see Section 3.4.1). The rate of fluidisation was then altered a number of times, and the measurements repeated each time once steady state was reached. During this series of experiments at progressively higher and higher flowrates, it was observed in between experiments that the thickness of the “settled” bed at zero flow gradually dropped from 0.738 to 0.500 m, a 30 % reduction. This indicates that there was a gradual loss of material.

3.3.2 Continuous Tests
The arrangement used in the continuous experiments is shown in Figure 6. A feed suspension of the required solids concentration and cenosphere grade was first prepared and agitated in the feed tank until it was well-mixed. Wash water was then turned on at the required rate and used to fill the Inverted Reflux Classifier, with the valve on the main underflow return line shut. Then the variable-speed pump on the discard water side stream was turned on and carefully adjusted until the rate of removal of discard water equalled the rate of fluidisation wash water addition. After that, the feed pump was turned on to start adding feed suspension to the unit. The pump on the main underflow return line was then turned on and adjusted until the desired overflow rate was achieved. The laboratory unit used variable speed Masterflex peristaltic pumps rated at 0 to 3 L/min. The pilot plant unit used King Cobra hose pumps rated at 0 to 25 L/min. Figure 7 shows a photograph of the pilot-scale unit.

Figure 7: Photograph of the pilot-scale Inverted Reflux Classifier experimental rig.
The laboratory-scale system was deemed to be at steady state when the pressure transducer reading was steady. Once the system was at steady state, timed samples of the feed, overflow product and underflow reject were then collected for analysis. Samples of the full cross-section of each stream were collected by diverting it into a beaker or bucket. The overflow and underflow streams were sampled before the feed stream, so as to avoid the disturbance of the feed affecting the subsequent measurements.

Operation of the pilot-scale Inverted Reflux Classifier was carried out in a similar manner to the laboratory-scale unit, but with larger flowrates. However, there were no pressure readings taken on the pilot scale unit. So instead, the unit was operated for at least 2 h before samples were collected, to ensure that it had reached a steady state. Small samples of the overflow were collected periodically and allowed to stand in a measuring cylinder. The relative thickness of the floats “sediment” layer was monitored to judge whether conditions were steady.

### 3.4 Analysis

#### 3.4.1 Solids Volume Fraction

In the bed expansion experiments, the solids volume fraction was estimated by measuring the pressure drop $\Delta P$ between tappings a vertical distance $\Delta H$ apart. The average bed density $\rho_{\text{bed}}$ was then calculated by:

$$\rho_{\text{bed}} = \frac{g \Delta P}{\Delta H} \tag{8}$$

Knowing the density of the cenosphere particles $\rho_p$ then enabled the volume fraction to be determined via:

$$\phi_s = \frac{\rho_{\text{bed}} - \rho_f}{\rho_p - \rho_f} \tag{9}$$

#### 3.4.2 Float-Sink Analysis

The samples of the feed, product and reject streams were float-sink separated in 1 or 2 L separating funnels (Figure 8). The sample was added, stirred vigorously for 5 to 10 s to minimise the rafting of
any silica particles, and then left to separate. The silica-rich sediment in the base of the funnel had a tendency to compact and set if left too long, so the stop cock at the base was initially opened every few hours to bleed out some of the sediment. After a total settling time of over 24 h the floats and sinks fractions were recovered separately, dried and weighed.

Figure 8: Separating funnel used to perform float-sink separation. Note the valve at the base was opened periodically to prevent the silica bed from setting.

The mass flowrates and compositions of all the streams into and out of the system were calculated from the measured sample masses and collection times. For a steady state system, the mass flow rate of feed into the unit divided by the combined rate of the exiting product and reject streams should equal one. A similar balance should also hold for each of the two density fractions. However, in practice calculations based on the raw data usually do not exactly satisfy these mass balances due to random sampling and measurement errors.

It is important to have an internally consistent set of data so that system performance parameters, such as recovery, can be calculated unambiguously. Therefore mass balance reconciliation was used to objectively obtain a self-consistent set of data. There were three measurements for each stream, the total mass rate (i.e. water plus all solids), the rate of float solids (cenospheres) and the rate of sink solids (silica). There were three streams analysed, the feed, product and reject, so there were a total of nine measurements for each experiment (NOTE: the mass rates of wash water and discard water were not included as these were physically balanced before the start of each experiment). Errors were assigned to each of these nine measurements, and the true value was assumed to be the measured value plus the error term. An objective function was defined equal to the sum of the
squares of the relative errors of each measurement. Then the Solver numerical search routine (Microsoft Excel) was used to minimise this objective function, by varying all the individual error terms, subject to the constraint that the adjusted values must satisfy all of the mass balance equations for steady state conditions, namely that the mass of each component (water, cenospheres and sinks) into the system must equal the total mass rate out.

Experiments where the ratio of measured solids mass rate in divided by measured solids mass rate out varied from 1.0 by more than 40% were not analysed further. In addition, experiments where any of the nine raw measurements were adjusted by more than 20% during mass-balance reconciliation were also omitted. This left 15 laboratory-scale and 14 pilot-scale experiments that were retained for further analysis. The tabulated results in Section 4 report the average absolute value of the percentage adjustment of the nine raw measurements in each experiment and also the maximum absolute adjustment.

Once a consistent set of data was obtained, the recovery of cenospheres was calculated from the mass rate of floats exiting in the overhead product divided by the mass rate entering in the feed. Solids concentration is defined as the total mass of solids divided by the total mass of suspension, expressed as a weight percent solids. The grade of cenospheres in each stream was calculated from the mass of floats divided by the total mass of floats and sinks solids in that stream. Upgrade is defined as the grade of cenospheres in the product divided by their grade in the feed.

The other important parameter reported is the volume reduction, which is defined as the volumetric flowrate of the feed divided by the volumetric flowrate of overhead product. This is based on the raw measurements of volumetric flowrate, since volumetric information was not used in the mass-balance data reconciliation. Note that volumetric flow rates are reported in Section 4 in terms of the equivalent superficial velocity in the vertical section of the column, namely the volumetric rate divided by the vertical column cross-sectional area. This enables direct comparison between the laboratory and pilot scale results.

### 3.4.3 Size Distribution Analysis

In some of the pilot-scale experiments, representative samples of the floats and sinks fractions of each stream were also analysed using a Malvern Mastersizer 2000. If the material was dried, it formed a cake that was then difficult to re-disperse. So these samples were kept in wet form.
The laser sizer purports to measure the volume size distribution of the particles. At steady state, the volume of solids in each size fraction of each density interval into the unit should equal the combined volume leaving in the two outlet streams. Similar to the mass balance reconciliation procedure outlined in Section 3.4.2, errors were assigned to each measured volume fraction for all three streams. These were varied using the Solver routine to find the set of errors that had the least overall sum of square relative error, subject to satisfying the volume balance constraints in each size and density interval. Only results where the average absolute adjustment to the volume fraction in each size interval of all three streams was less than 10% are reported in Section 4.

From this reconciled data, the total recovery of all floats material to the product could be calculated. It was also possible to construct the size partition curve for the floats fraction, where the partition number is the probability of a floats particle of that size reporting to the overhead product stream.

It is well known and accepted that in gravity separation performance tends to deteriorate as particle size becomes smaller, so it is unrealistic to expect high recoveries of the finest particles. This poor performance at the fine end of the size distribution may obscure good recoveries being obtained for the larger particles. Therefore the volume-based recovery of particles larger than 20 μm in size,  \( R_{>20} \), was also calculated using the two-product formula [14]:

\[
R_{>20} = \frac{x_p}{x_F} \left( \frac{x_F - x_R}{x_R - x_R} \right)
\]

where \( x_p, x_F \), and \( x_R \) are the fractions of the particles larger than 20 μm in each of the feed, product and reject streams respectively, as measured by the Malvern Mastersizer.

4.0 Results and Discussion

4.1 Bed Expansion Characteristics

In a conventional fluidised bed consisting of particles of one size and density there is a unique relationship between the fluidisation velocity and the steady state volume fraction of the particles. As the velocity increases, the solids volume fraction decreases. This can be predicted by the Richardson-Zaki equation (Equation 2). However, in the standard Reflux Classifier, this relationship only corresponds to the early part of the curve, before the bed has expanded enough to reach the inclined channels. Once particles start to enter the channels, they are captured and returned back to the bed. Thus for a given initial mass of solids, there reaches a point where the bed stops expanding significantly, with further increases in fluidisation velocity producing relatively little effect on the solids volume fraction [33].
Batch bed expansion experiments were therefore performed to confirm whether analogous behaviour occurs when positively-buoyant particles are downwards fluidised in an Inverted Reflux Classifier. Figure 9 shows the results of the experiments described in Section 3.3.1. The results are also compared with the theoretical prediction from Equations 1 and 2. Clearly, the behaviour does not follow the normal expansion behaviour for a fluidised bed (Eq. 2). Rather it is similar to that observed by Doroodchi et al. [33] in the standard Reflux Classifier. Once the cenospheres started entering the inclined section, they were captured and returned to the vertical section above. Thus, the solids volume fraction in the vertical section only decreased very slowly with increases in the fluidisation velocity from 0.002 to 0.0012 m/s. Most of this decrease can be explained by the gradual loss of 30% of the finer and/or denser solids from the system (Section 3.3.1). Even at fluidization velocities twice the nominal terminal rise velocity of the particles, the volume fraction of solids in the Inverted Reflux Classifier remained finite and significant.

![Figure 9: Volume fraction of solids in the vertical section as a function of the downwards fluidisation velocity.](image)

Experimental values were determined from the measured bed pressure drop (Equation 9). Theoretical predictions of terminal free settling velocity \( u_t \) and bed expansion behaviour are based on Equations 1 and 2 for spherical particles with size and density of 40 \( \mu \)m and 315 kg/m\(^3\) respectively. These quantities were found to characterise the expansion in the vertical section. Error bar shows expected drop in solids volume fraction due to the entrainment of relatively fine particles, equivalent to 30% of the original solid mass.
These results show that the Inverted Reflux Classifier applied to positively-buoyant particles gives equivalent behaviour to that obtained using negatively buoyant particles in the standard Reflux Classifier. This is important because it shows that the inclined channels at the base can be relied on to prevent cenospheres being entrained downwards into the underflow. This phenomenon should be able to be combined with a downwards flow of wash water to remove entrained high density fly ash while preventing a large reduction in cenosphere recovery.

4.2 Continuous Experiments

The laboratory scale mass-balance results are shown in Table 1. Different combinations of fluidisation, feed and underflow removal rates were investigated. Note that the first seven experiments used feed solids concentrations of about 1.5 wt% solids, with about 60 % grade of cenospheres. These show the potential of the device to further concentrate cenospheres scooped from the surface of fly ash tailings ponds. Experiments 8 to 15 used much higher feed concentrations of around 8.5 wt % solids, but with a cenosphere grade of only about 1.5 wt% down to as low as 0.5 wt%. These experiments demonstrate the potential of the device to recovery and concentrate cenospheres directly from a raw fly ash feed.

The pilot scale mass-balance results are shown in Table 2. Note that due to the logistical difficulties of providing enough feed to run a continuous pilot-scale experiment until it reached steady state, these experiments were all performed at quite low feed concentrations less than 1 wt% solids, and cenosphere grades in the range 40 to 75 %.

Tables 1 and 2 report the consistency of the raw data, in terms of the ratios of the measured mass rate of material in the feed divided by the measured mass rate out in the combined product and reject streams. In both the laboratory and pilot scale experiments, the mass balances on total slurry mass rates were very accurate, with discrepancies usually less than ± 3 %. However, there were significantly larger discrepancies in the individual component balances on the cenosphere and silica rates, reflecting the difficulty in performing precise sink-float separations of the sample streams. This is also to be expected due to the low concentrations of these two components, so that the effects of random sample variability, losses during handling and imprecision in weighing the samples were magnified. Also reported are the average adjustment and the maximum single adjustment of the raw data during the reconciliation process.
Table 1: Mass-balance reconciled results obtained from the laboratory-scale experiments. Consistency of mass balances based on raw data is indicated, as well as the maximum and average adjustment to the raw data during reconciliation. Note that volumetric flowrates and volume reduction are based on raw measurements.

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Table 2: Mass-balance reconciled results from the pilot-scale experiments. Consistency of raw data mass balances is indicated as well as the maximum and average adjustment to the raw data during reconciliation. Note that volumetric flowrates and volume reduction are based on raw data. Final eight rows show results of calculations based on the volume-balance reconciled size distribution measurements from Malvern Mastersizer.

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Calculations based on Volume-Balance Reconciled Size Distribution Data from Malvern Mastersizer

| | | | | | | | | | | | | | |
| Total Recovery of Cenospheres (vol.%) | 87.3 | 87.5 | 92.7 | 68.2 | 77.7 | 82.2 | 83.7 | 84.3 | 77.2 |
| Recovery of Cenospheres >20 μm in Size (vol.%) (Eq. 10) | 95.4 | 90.2 | 95.8 | 80.7 | 84.9 | 88.6 | 90.3 | 90.7 | 84.0 |
| Partition Cut Point, d50 (μm) | 20 | | | | | | | | |
| Experimental Throughput Advantage, F (Eq. 3) (Assumes particle density 400 kg/m3) | 25.2 | 25 | 25 | 23 | 22 | 19 | 21 | | |
| Experimental Segregation Efficiency, η (%) (Eq. 5) | 39 | 38 | 39 | 41 | 42 | 46 | 47 | | |
| Predicted Segregation Efficiency, ηpred (%) (Eq. 6) | 44 | 38 | 39 | 41 | 42 | 46 | 43 | | |
| Average (absolute) Adjustment during Volume-Balance Reconciliation of Size Data (%) | 4.6 | 3.6 | 2.9 | 2.3 | 6.8 | 4.4 | 4.1 | 3.9 | 5.7 |
Due to the similar behaviour of the laboratory and pilot scale, these two sets of results are now discussed together in parallel. The following sub-sections focus on different aspects of performance, including: size partition, recovery, volume reduction, feed concentration and wash-water rate.

4.2.1 Size Partition Performance

In some of the pilot-scale runs, representative samples of the floats and sinks fractions from the three streams were sized using a Malvern Mastersizer 2000. Typical size distributions are shown in Figure 10. The size distributions of the feed, product and reject stream sinks fractions were virtually identical (Figure 10a), with a peak at around 50 μm and signs of a slight perturbation at around 4 μm. There is minimal size classification of these particles. Because there was so little difference between the three distributions, it was not possible to perform volume-balance data reconciliation on the sinks fraction size data.

Figure 10: Volume frequency distributions of (a) sinks and (b) floats fractions from the feed, product and reject streams of pilot-scale Run 17 (unreconciled raw data).

Figure 10b shows that there were three peaks in the floats material size distributions: a small peak in the reject stream at 4 μm, a peak in the feed and reject streams at approximately 20 μm and a peak in the feed and product streams at approximately 50 μm. The 20 μm peak was almost non-existent in the product stream, whereas the 50 μm peak was almost absent in the reject stream. Hence a size classification has clearly taken place, with more of the fine floats material reporting to the reject underflow stream and more of the coarse floats material reporting to the overflow product. This reflects the greater difficulty in recovering finer particles due to their lower rise velocities.

This classification is shown in the size partition curve obtained from the volume-balanced size distribution data, shown in Figure 11. Floats particles between 40 and 90 μm in diameter had a greater than 90% chance of reporting to the overhead product stream, whereas particles around 20 μm in diameter had only a 25% chance of reporting to the overhead product. It is well known in size classification devices that fine particles have a tendency to follow the water split, due to their low settling velocities [e.g. 14]. In Runs 17-19, only 9 - 10% of the water added in the feed and fluidising streams reported to the overhead product, so only approximately 10% of ultrafine particles should be expected to report to the overhead product due to water split. The fact that the floats partition
was higher than this shows that even for the fine particles, some density based classification was still occurring. The apparent improvement in recovery as particle diameter drops below 20 μm and the drop in recovery as particle diameter becomes larger than 100 μm are unexpected, but may simply be a numerical artefact caused by the very small amounts of material in these size ranges (Figure 10b) giving a much higher uncertainty in the partition calculations.

Figure 11: Floats partition to overhead product versus particle size for pilot scale Runs 17-19. Results based on volume-balance reconciled data.

The particle cut size inferred from the partition curve enables calculation of the throughput advantage which can then be compared with the prediction from the Laskovski correlation (Eq. 6). Consider Run 17, for example. The 50 % partition size was approximately 25 μm (Figure 11). If we assume an average density of 400 kg/m³ for these particles, then their terminal settling velocity was 

\[ u_t = \left(25 \times 10^{-6} \text{ m}^2\right)(400 - 1000 \text{ kg/m}^3)(9.81 \text{ m/s}^2)/18/(0.001 \text{ Pa s}) = -2.0 \times 10^{-4} \text{ m/s} = -0.74 \text{ m/h} \]  

with a terminal particle Reynolds number of 

\[ Re_t = (1000 \text{ kg/m}^3)(2.0 \times 10^{-4} \text{ m/s})(25 \times 10^{-6} \text{ m})/(0.001 \text{ Pa s}) = 0.005. \]  

For this experiment, the downwards superficial velocity through the vertical section was approximately 17 m/h. Hence the throughput advantage was 

\[ F = (17 \text{ m/h})/(0.74 \text{ m/h}) = 23 \]  

(Eq. 3).

Note that hindered settling effects have been neglected, which is justified based on the extremely low solids concentrations (less than 0.5 wt%) in the underflow reject stream. The theoretical throughput advantage was 

\[ F_{\text{theory}} = 1 + (1.2 \text{ m})/(0.006 \text{ m})\cos(70^\circ)\sin(70^\circ) = 65 \]  

(Eq. 4), so the efficiency was \( \eta = 35 \% \) (Eq. 5). The efficiency predicted by the Laskovski correlation was \( \eta_{\text{pred}} = [1 + \)
0.133 \cos(70^\circ)(0.005)^{1/3}(1.2 \text{ m}/0.006 \text{ m})^{-1} = 39 \% \text{ (Eq. 6)}. Similar close agreements between the predicted and experimental segregation efficiencies were obtained in most of the other experiments for which the size distributions of the feed, product and reject were measured (Table 2). This close agreement further demonstrates that the behaviour of buoyant particles in the Inverted Reflux Classifier is analogous to that of negatively buoyant particles in the standard configuration. Hence correlations developed for one can be reliably used for the other.

The experimental throughput advantages \( F \) varied from 16 to over 30 (Table 2). This highlights the large increase in the hydraulic capacity of Reflux Classifiers compared with conventional teetered/fluidised bed classifiers.

### 4.2.2 Measures of Cenosphere Recovery

Figure 12 shows the recovery of the cenospheres based on particle volume versus the cenosphere recovery based on particle mass for all the pilot-scale experiments. The volume-based recoveries are significantly higher than those based on mass. One possible explanation for this systematic bias is that the small cenospheres may on average have had a higher density than the larger cenospheres. If this was so, then the small cenospheres represented a greater fraction of the total mass of floats material than they did of the total volume of floats. So the lower recovery of the finer cenospheres seen in Figure 11 would cause a greater drop in the recovered mass than in recovered volume. Indeed if smaller cenospheres do have a higher density, then this would help to further explain the reduced recovery of the finer floats seen in Figure 11, since it is harder to recover higher density material to the overflow product. More detailed investigation of cenosphere density as a function of size would be required to confirm this.
Figure 12: Pilot plant floats total and > 20 µm volume-based recoveries calculated from the reconciled Malvern size distribution data versus the mass-based recovery calculated from the reconciled mass rate data.

Figure 12 also shows that the volume-based recovery of cenospheres greater than 20 µm in diameter (via Eq. 10) is on average 9 % higher than the volume-based recovery over the entire size range down to zero. This improvement is expected due to the fact that gravity separation performance deteriorates at finer particle sizes [e.g. 14]. Again this is reflected in the lower partition values for fine cenospheres seen in Figure 11. Overall, the fact that the recovery in many cases exceeded 90 %, is an excellent result when considering that there was also a significant upgrade and volume reduction (Table 2).

4.2.3 Effects of Volume Reduction

Figures 13 and 14 show plots of cenosphere recovery versus volume reduction for all the laboratory and pilot-scale runs respectively. A similar trend was obtained at both scales, namely that as the volume reduction was increased, the recovery of cenospheres to the overflow decreased. This trend is to be expected, since reducing the rate of material reporting to the overflow results in an increase in the underflow rate, which thus increases the likelihood of misplacing cenospheres to the reject. The slight suggestion in the pilot-scale data that recoveries start to rise at high volume reductions is believed to be an artefact of the extremely low stop-start product flowrates that occur at such large volume reductions, which make it hard to obtain representative flow rate measurements.
Figure 13: Cenosphere recovery to product versus volume reduction in the laboratory-scale reflux classifier.

Increasing the volume reduction should also improve the upgrade, which is indeed the case as shown in Figure 15 for laboratory-scale Experiments 1 to 7 where the upgrade was close to the maximum possible. In the pilot scale work, most experiments generated products with a cenosphere grade greater than 84 % (Table 2), and often well in excess of 90 %. These grades, which are on a mass basis, would be far closer to 100 % on a volume basis. Figure 16 shows the cenosphere upgrade versus volume reduction obtained in the pilot-scale unit, together with one set of results from the laboratory-scale unit for comparison. Again the general trend is that upgrade increases with increasing volume reduction.

Figure 14: Recovery versus volume reduction for all experiments in the pilot-scale Inverted Reflux Classifier. One set of laboratory-scale results from Figure 13 is included for comparison.
Figure 15: Cenosphere upgrade versus volume reduction in the laboratory scale unit for all runs done at low feed solid concentrations (Experiments 1 to 7). Note that the maximum possible upgrade in these experiments was approximately 1.6.

Figure 16: Cenosphere upgrade versus volume reduction for all the pilot-scale Inverted Reflux Classifier experiments. One set of laboratory-scale data from Figure 14 is shown for comparison.
The laboratory and pilot scale work covered a wide range of operating conditions and also used slightly different cenosphere feed stocks. Given this variation, the similarity in both qualitative trends and quantitative values between laboratory and pilot-scale results shown in Figures 14 and 16 illustrates the robustness of this technology. This agreement provides a high degree of confidence in the likely performance of full-scale units, and the potential for scale-up to meet the needs of industry.

4.2.4 Effects of Feed Grade and Solids Fraction

Laboratory-scale Experiments 8 to 12 had a much higher feed concentration of 8 wt % solids, but only a very low feed cenosphere grade of order 1.5 wt%. These experiments were designed to assess the ability of the system to recover cenospheres directly from low grade fly ash (rather than from the floats material skimmed off tailings ponds). The Inverted Reflux Classifier performed well (Table 1). The recoveries in these runs were 34 to 55 wt%, and the product grade was 45 to 60 wt%, with an upgrade of over 30. Experiments 13 to 15 dropped the feed cenosphere grade down even lower to 0.5 wt%, and obtained upgrades as high as 160, with 55 % recovery of cenospheres, an exceptional result.

4.2.5 Effects of Wash Water Rate

In laboratory-scale Experiments 5 to 7 the fluidisation wash water rate was varied from 0.38 to 3.0 m/h. This variation had almost no effect on the recovery of cenospheres. Similarly, in the pilot-scale unit, increasing the wash water rate from 0.17 to 1.33 m/h (Runs 17 to 19), had almost no effect on the floats size partition curves, as shown in Figure 11. There was a slight increase in the product grade from 93.0 % to 96.8 % in the three laboratory scale experiments (Table 1). This might reflect the better removal of entrained silica material with higher wash water rates, however, this trend was not clearly duplicated in any of the sets of pilot-scale tests (Table 2) where fluidisation rate was varied with other conditions held constant. The best that can be said is that fluidisation wash rate did not have a strong effect on product grade in the range of conditions studied in this work.
5.0 Conclusions

The recovery and concentration of cenospheres from fly ash are challenging given that technologies such as flotation or electrostatic separation are unable to target a property that distinguishes the cenospheres from the denser silica. Gravity separation is arguably the only option, but has significant capacity constraints. A novel device, the Inverted Reflux Classifier, was therefore investigated at both laboratory and pilot scale. The Inverted Reflux Classifier efficiently separated positively-buoyant cenospheres from negatively-buoyant silica particles. High recoveries and upgrades were obtained, with similar performance achieved at both scales. Results were consistent with the existing correlations developed for negatively buoyant particles in standard Reflux Classifiers. The throughput advantage compared to a conventional teetered bed separator was over 30 in some cases. Hence this technology has the potential to efficiently recover and concentrate cenospheres from power station fly ash, thus creating a valuable by product. Further research is required using real fly ash samples to confirm this potential.
6.0 Acknowledgements

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7.0 References


