Coal Combustion and Gasification Products is an international, peer-reviewed on-line journal that provides free access to full-text papers, research communications and supplementary data. Submission details and contact information are available at the web site.

© 2015 The University of Kentucky Center for Applied Energy Research and the American Coal Ash Association

Web: www.coalcgp-journal.org
ISSN# 1946-0198
Volume# 7 (2015)

Editor-in-chief: Dr. Jim Hower, University of Kentucky Center for Applied Energy Research

CCGP Journal is collaboratively published by the University of Kentucky Center for Applied Energy Research (UK CAER) and the American Coal Ash Association (ACAA). All rights reserved.

The electronic PDF version of this paper is the official archival record for the CCGP journal.

The PDF version of the paper may be printed, photocopied, and/or archived for educational, personal, and/or non-commercial use. Any attempt to circumvent the PDF security is prohibited. Written prior consent must be obtained to use any portion of the paper’s content in other publications, databases, websites, online archives, or similar uses.

Suggested Citation format for this article:

Multistage Concentration of Cenospheres in Fly Ash Using the Inverted Reflux Classifier
A. Kiani*, J. Zhou, K.P. Galvin

Centre for Advanced Particle Processing and Transport, Newcastle Institute for Energy and Resources, University of Newcastle, Callaghan, NSW 2308, Australia

ABSTRACT

Cenospheres are one of the most valuable components found in the fly ash waste of coal-fired power stations, consisting of hollow, aluminosilicate spherical shells and offering very low bulk density, high strength, and high thermal resistance. Although wet processing methods offer the best prospect for achieving recovery and concentration of the cenospheres, significant impediments remain for achieving satisfactory economics. This work was concerned with the recovery and concentration of cenospheres from fly ash using a novel system, the inverted reflux classifier (IRC), covering single and multistage processing. The system consists of a series of parallel inclined channels mounted underneath an inverted liquid fluidized bed. This work demonstrated a high separation performance due to the so-called Boycott effect, which arises in inclined settling. In preliminary experiments, a fly ash feed with a very low cenosphere concentration was examined. After two stages, the product grade and total recovery were unsatisfactory. However, in the main part of the study, examining a fly ash feed with about 0.9% cenosphere concentration, a three-stage IRC was used to achieve almost pure cenosphere product, with the cenosphere grade increasing to around 97 wt% (almost pure on a volume basis). In the second stage, which involved a much more dilute feed, the more neutrally buoyant cenospheres remained in the stream to tailings. Hence, with three stages, the recovery fell to 50%. It is concluded that accelerated separation of the positively buoyant cenospheres is promoted by the presence of a high concentration of the negatively buoyant fly ash, and therefore a single separation stage is preferable.

Journal homepage: www.coalcgp-journal.org

ARTICLE INFO

Article history: Received 2 July 2015; Received in revised form 6 October 2015; Accepted 7 October 2015

Keywords: inverted reflux classifier; recovery; cenospheres; fly ash; upgrade

1. Introduction

Fly ash is a waste that is generated as a by-product from burning coal in power stations worldwide. The production of fly ash in the United States alone was estimated to be around 52 MT in 2012. A proportion of this waste is directly reused in different areas such as concrete products, cement, and mining applications. However, about 50% of the fly ash waste is discarded to the lands around the power stations (American Coal Ash Association, 2014). In addition to the environmental issues, fly ash has the potential to cause respiratory diseases in humans.

One of the fine components found in fly ash is cenospheres, consisting mostly of silica and alumina, which are valued at up to $2000 per metric ton. Owing to useful properties such as low bulk density, these fine particles have been used in different applications such as light-weight composites and insulations. The concentration of cenospheres in fly ash has decreased in recent years owing to the modifications made to the burners of power stations to reduce NOx emissions (Huang et al., 2003). Therefore, the conventional method of separation, scooping the cenospheres from the surface of the ash pond, is inefficient. Furthermore, in the conventional methods, fine cenospheres with very low rise velocities are not recovered, and also the ultrafine fly ash particles contaminate the cenosphere product due to entrainment.

Flotation is the most common method used in separating fine particles; however, it is ineffective here given the similar surface properties of cenospheres and fly ash. Wet gravity separation can be effectively applied due to the significant difference between the

* Corresponding author. Tel.: (+61) 2 40339251. E-mail: Ali.Kiani@uon.edu.au
densities of the particles and water. However, cenospheres are very fine with low rise velocities; thus, a method for increasing their segregation velocities needs to be used in order to provide a significantly high throughput.

Application of the principal of inclined settling, first reported by Boycott (1920), can effectively increase the rate of segregation. Galvin invented the reflux classifier, which provides a remarkably more powerful approach over conventional fluidized beds in the separation of particles based on their density and size (Laskovski et al., 2006; Galvin et al., 2009; Walton et al., 2010). The other phenomenon that considerably increases the rate of segregation is the bulk streaming formation effect, which was comprehensively investigated by Batchelor and Van Rensburg (1986). This phenomenon involves positively and negatively buoyant particles within specific concentration regimes. In such suspensions, the particles gather together and form clusters moving as a bulk in the suspensions, resulting in a significant enhancement in the particle convective driving force and hence an effective increase in their velocities.

Previously, Kiani et al. (2015a) employed the combined effects of inclined settling and bulk streaming to obtain a high recovery of cenospheres in the inverted reflux classifier (IRC). Later, Kiani et al. (2015b) comprehensively investigated the cenosphere separation process in a one-stage pilot-scale IRC. In the present study, a series of IRCs was applied to upgrade the cenosphere concentration and separation in the fly ash. In fact, in this article, we report for what is to our knowledge the first time, findings from the multistage application of IRCs involving fly ash feeds with different cenosphere concentrations. Thus, this article provides further insight into the optimization in terms of the separation of the cenospheres from fly ash, covering the product grade, recovery, and throughput.

2. Experimental Methods

2.1. Inverted reflux classifier

In this work, the IRC, a system of parallel inclined channels installed underneath an inverted liquid fluidized bed, was applied to separate cenospheres from fly ash. Laboratory- and pilot-scale IRCs were used in this study. At the laboratory scale, the fluidized bed section was 1 m long with a cross-sectional area of about 86 cm². The inclined section consisted of eight channels with perpendicular spacing of 9.5 mm. The inclined section was mounted below the fluidized bed section at an inclination of 70° to the horizontal. The much larger pilot-scale IRC consisted of a 1.7-m-long fluidized bed section located above a 1.2-m-long inclined section. The cross-sectional area of the vertical fluidized bed section was about 900 cm². The inclined section contained 38 channels with 6-mm perpendicular spacing. Two pressure transducers were installed along the vertical section, providing information on the density of the formed suspension. A portion of the tailings was discharged from a level of 1 m above the top of the IRC, at atmospheric pressure, ensuring the IRC system was maintained under positive pressure. Figure 1 shows a schematic representation of the IRC used in this work.

2.2. Materials and methods

The fly ash waste used in this work was sourced from a power station in Australia. Feed slurry was prepared by mixing the solid fly ash with a specific amount of water and then was kept stirring in a mixing tank. The feed was pumped to the vertical fluidized bed section of the IRC. On entry, some of the cenospheres moved upward through the vertical section, while the rest were entrained toward the inclined section. The high segregation rate along the inclined channels permitted the cenospheres to return to the vertical section, preventing their loss to the tailings stream. A fluidization chamber was installed at the top of the IRC to distribute the fluidization water and in turn to wash away the ultrafine gangue materials from the cenosphere product. At the end of each experiment, representative samples were taken from all streams and analyzed.

A multistage arrangement was used to upgrade the cenospheres in the fly ash. In a preliminary work, a series of two laboratory IRCs was applied to upgrade the cenospheres in fly ash feed containing only 0.33 wt% cenospheres. In the main part of the study, however, a fly ash feed with around 0.9 wt% cenosphere concentration was examined using a series of three IRCs. The large pilot-scale IRC was used as the first stage, delivering a significant quantity of partially upgraded cenospheres to feed the additional stages. In this first stage, the yield was adjusted to obtain the highest possible recovery. Then, the product of the first stage was sent to the laboratory-scale IRC, the aim being to deslime the product. The third-stage IRC was then used to further purify the product of the second stage. Samples of all streams in each stage were analyzed. A simple representation of the three-stage process is shown in Figure 2.

2.3. Data analysis

Sink-float separation was used to quantify the compositions of the samples. Each sample was poured into a sink-float separating funnel and left for 24 hours. The dense particles were discharged from the base of the funnel. Some fine cenospheres were entrained with the sinks and were consequently separated and returned to the funnel. When a layer of clear water was observed between the cenospheres and fly ash in the funnel, the fly ash particles were discharged, and then the cenospheres left in the funnel were washed several times and collected. The separated fly ash and cenospheres were placed in an oven for drying, providing the mass of dried solids for the calculation of the product grade and then the recovery of the cenospheres.
A laser scattering technique (Malvern Mastersizer 3000) was applied to measure the size distributions of the cenospheres and fly ash. The errors in all experimental data were minimized using a mass balance reconciliation method. The cumulative size distributions of the cenospheres and fly ash are shown in Figure 3. It was evident that 26 vol% of the fly ash was finer than 20 μm, making the upgrading process difficult. Furthermore, about 67 vol% of the cenospheres was measured to be finer than 100 μm, making them difficult to recover. The average particles size was calculated to be 95 μm for the cenospheres and 64 μm for the fly ash. The size of the particles in both fly ash samples used in this study was almost the same. The density of the cenospheres and fly ash in the main feed was measured to be about 843 and 1887 kg/m^3, respectively, using the gas pycnometry method. However, the cenosphere density in the preliminary work was measured to be slightly lower at about 807 kg/m^3. Scanning electron microscopy analyses of the fly ash and cenosphere feeds used in the preliminary and main experiments are shown in Figure 4.

3.3. Results and Discussion

In the preliminary experiments, a series of two IRCs was used to upgrade the cenospheres in the low-grade fly ash. The optimum feed solids concentration for a fly ash with 1 wt% cenospheres (Kiani et al., 2015a) was established prior to commencing the multistage separations in the main part of the study. At the optimum, the pilot-scale IRC was used to process approximately 1 metric ton of fly ash and generate a large quantity of product to feed to the next stages. A series of IRCs was employed to purify the cenosphere product, while aiming for the maximum possible recovery. Error bars were defined to show the discrepancy between the experimental data and the mass-balanced data. As the errors were found to be negligible for the product grades results, the error bars are only shown for the recovery data points.

3.1. Preliminary study on the multistage separation of cenospheres from fly ash

A fly ash feed containing about 0.33 wt% cenosphere concentration was first processed in the laboratory-scale IRC, aiming to recover most of the cenospheres. The product obtained from this stage was then sent to the other IRC to be upgraded. The same operating conditions were adjusted for both stages. The volumetric split ratio was about 40%, and the feed and fluidization water volumetric fluxes were about 7.3 and 0.88 m^3/(m^2 h), respectively. As indicated in Table 1, the cenospheres were upgraded from 0.33 wt% to about 3.8 wt% in stage 1. The recovery of this stage was around 82.5%. After processing the product of the first stage in the second stage, a final product grade of 21.3 wt% and a recovery of 79.3% were achieved. Lower split ratios could be used in the process for obtaining a higher product grade; however, the recovery would be lower in this case. As listed in Table 1, the overall product grade and recovery were calculated to be 21.3% and 65.4%, respectively. A higher product grade but a lower overall recovery could be achieved using another stage of the IRC. It was concluded that the multistage IRC process was inefficient in separating cenospheres from the low-grade fly ash.

Previously, Kiani et al. (2015a) found the optimum feed solids concentration in the cenosphere separation process to be around
In that study, a product grade of 64% and a recovery of about 90% were achieved using a one-stage IRC (Kiani et al., 2015a). The single-stage pilot-scale separation of cenospheres from fly ash was then comprehensively investigated by Kiani et al. (2015b), and a very good agreement between different scales of the IRC was achieved. In the main part of the present study, the pilot-scale IRC at the optimum feed solids concentration and a solid throughput of about 4.0 t/(m² h) operating at a split ratio of 20% was used as the first stage of a three-stage IRC process in order to obtain the highest possible recovery and a satisfactory grade.

### Table 1

<table>
<thead>
<tr>
<th>Stage</th>
<th>Feed grade</th>
<th>Product grade</th>
<th>Upgrade</th>
<th>Recovery</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stage 1</td>
<td>0.33</td>
<td>3.8</td>
<td>11.4</td>
<td>82.5</td>
</tr>
<tr>
<td>Stage 2</td>
<td>2.7</td>
<td>21.3</td>
<td>7.8</td>
<td>79.3</td>
</tr>
<tr>
<td>Whole process</td>
<td>0.33</td>
<td>21.3</td>
<td>64.6</td>
<td>65.4</td>
</tr>
</tbody>
</table>

![Fig. 5](image)  
**Fig. 5.** Product grade and cenosphere recovery at different stages of the process.
3.2. Maximizing cenosphere product grade using the multistage IRC

3.2.1. Product grades and cenosphere recovery

At the optimum feed solids concentration, three stages of IRC were applied to upgrade the cenospheres in the fly ash. In the first-stage IRC, the primary aim was to operate at a relatively high feed rate, while ensuring high recovery of the cenospheres. Here, an upgrade of about 20 was deemed satisfactory. The focus of the latter stages was on the further upgrading of the cenospheres from the fly ash. Figure 5 shows the product grade and the cenosphere recovery obtained at different stages of the process.

The product grade of the first stage was relatively low at about 17%, but the recovery was significant at about 80%. This stage of the process was carried out at the optimum feed solids concentration of about 39% and a solids throughput of about 4.0 t/(m² h). The fluidization water flux was around 0.83 m³/(m² h), and the product rate was set at about 20% of the feed volumetric rate. In the second and third stages, however, lower feed fluxes and higher split ratios were used. In the second stage, the product grade of cenospheres was raised to about 77%; however, the recovery obtained from this stage was about 69%. In the third stage, the cenospheres grade was increased to about 97%, and cenosphere recovery of around 92% was achieved.

Figure 6 shows the cumulative grade and recovery during the multistage processing. The combined grade and recovery of the cenospheres in the multistage IRC were about 97% and 50%, respectively. In fact, as the second stage of the process was conducted at a relatively low feed rate and a high split ratio (volumetric product to feed ratio), a higher recovery of cenospheres was expected. However, the low solids concentration of the feed, about 10.3 wt%, could be the reason for the relatively low recovery of this stage. According to the literature (Fessas and Weiland, 1984), in a mixture of positively and negatively buoyant particles, the presence of a high concentration of negatively buoyant particles can cause the streaming formation and hence promote an increase in velocity of those particles. The feed solids concentration in stage 2 was perhaps much lower than the critical concentration needed to induce the streaming effect.

Figure 7 shows the product grade and cenosphere recovery obtained from the third stage of the process at different product fluxes. At constant feed and fluidization water fluxes, the product grade decreased, and the cenosphere recovery increased by increasing the product flux from 0.38 to 1.30 m³/(m² h).
Figure 8 shows the grade-recovery curves for each stage. It was appropriate to operate stages 2 and 3 at relatively low feed volumetric fluxes given the significant upgrade following stage 1. As the feed volumetric fluxes of stages 2 and 3 were relatively low, their grade-recovery curves were higher than for stage 1.

3.2.2. Size and density of cenospheres in the multistage IRC process

The size and density of the cenospheres in the product of each stage were measured to investigate the performance of the multistage IRC in detail. Figure 9 presents the size distributions of the cenospheres in the products from all three stages. The size distribution of the first and second stages had a significant difference, reflecting the loss of considerable proportions of fine cenospheres to the tailings in stage 2. However, the size distribution of the cenospheres exhibited no change from stage 2 to stage 3.

Using the particle size distributions of the feed, tailings, and product, partition curves were plotted for each stage of the process. As shown in Figure 10, the second stage showed a poorer size separation compared with the first stage due to the lack of the bulk streaming phenomenon, reflecting the loss of fine and high-density cenospheres to the tailings in this stage. On the other hand, the third stage of the process, which involved a very dilute feed containing less dense and larger cenospheres than the second stage, provided a sharper separation. Table 2 presents the imperfection and $d_{50\%}$ (Wills, 1997), indicating the efficiency of different stages of the process.

Figure 11 shows the density of the solids and the cenospheres in the product obtained from each stage compared with the average density of the cenospheres in the feed. The density of solids in the product decreased from about 1413 kg/m$^3$ in stage 1 to about 778 kg/m$^3$ in stage 2 and then almost levelled off. These data reflected the difficulties in the recovery of high-density cenospheres in stage 2. This difficulty was not apparent in stage 1 due to the high suspension density involved.

It was previously explained that the occurrence of the bulk streaming motion depended on the concentrations of cenospheres and other solids in the suspensions. In the second stage of the multistage IRC, the total solids concentration was about 10 wt%, obviously lower than the threshold concentration reported by other investigators (Fessas and Weiland, 1984; Batchelor and Van Rensburg, 1986). In fact in the second and third stages, the separation was only promoted by the Boycott effect, while the first stage separation was promoted by both the Boycott and streaming effects.

### Table 2

<table>
<thead>
<tr>
<th>Stage</th>
<th>$d_{50%}$</th>
<th>Imperfection</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stage 1</td>
<td>36</td>
<td>0.21</td>
</tr>
<tr>
<td>Stage 2</td>
<td>54</td>
<td>0.35</td>
</tr>
<tr>
<td>Stage 3</td>
<td>40</td>
<td>0.06</td>
</tr>
</tbody>
</table>

4. Conclusions

The concentration and recovery of cenospheres from fly ash were studied using the IRC. In order to achieve a high upgrade of the cenospheres in the fly ash, a multistage IRC process was used. The preliminary results demonstrated the ineffectiveness of the multistage IRC arrangement in processing a low-grade fly ash. However, in the main part of the study, involving a fly ash feed with around 0.9 wt% cenospheres, a product grade of about 97% was achieved. However, the recovery dropped to about 50% at the end of the multistage process. The size and density analysis suggested that the second stage of the process was less effective in recovering the fine and dense cenospheres. The low total solids concentration of the feed in the latter stages and, hence, the lack of the bulk streaming formation effect were the likely reason for the lower overall recovery. These results suggest that a one-stage process, conducted at a reduced feed rate, may provide the best approach for achieving the target grade and recovery.

Acknowledgments

The authors would like to acknowledge the financial support received from Vecor Australia and the Australian Research Council.
Kiani et al. / Coal Combustion and Gasification Products 7 (2015)

References


