P2-22

THE USE DYNAMIC AVALANCHING AND FRACTAL ANALYSIS TO CHARACTERISE URANIUM OXIDE POWDERS

JW Hobbs* and D Rhodes
Research and Technology, British Nuclear Fuels, Sellafield, Seascale, Cumbria, UK CA20 1PG

ABSTRACT

Direct thermal denitration is an attractive method of co-converting mixed-metal nitrate solutions of plutonium and uranium into oxide because of its apparent simplicity. Such benefits are often marred by the relatively poor powder quality and handling characteristics, which can be overcome by modifications to the process chemistry. To ensure that powder synthesis routes under assessment require the minimal further processing it is necessary to be able to characterise the powder fully in term of the key fundamental properties. This paper will demonstrate the use of a dynamic avalanching technique, fractal analysis and morphology to assess processing behaviour. The use of dynamic avalanching to uniquely characterise the chaotic flow properties of urania powders has proved successful and results have shown that this technique is capable of detecting small differences in processing behaviour due changes in morphologies and particle size distribution. This technique has promise for being able to provide nearly instantaneous feedback to the powder generation process being monitored (e.g. calcination, milling, mixing). The use of fractals to describe powders is an interesting characterisation tool when combined with morphological shape factors and the flow index.

INTRODUCTION

This paper discusses the use of a recently developed instrument, the API Aeroflow analyser, for testing and quantifying the rheology of bulk powder samples. The motivation for this investigation is to quantify an index, a number, by which the flowability of one powder may be compared to that of another and to try and establish a link with morphological shape factors and in particular the fractal index. The Aeroflow analyser relies on a technique referred to as dynamic avalanching to deduce how well a powder flows. At the heart of the analyser is a rotating drum into which bulk powder samples are placed. The tumbling of powders in the sample drum is in marked contrast to many of the standardised tests, Jenike shear cells or funnel flow test, which have been conceived throughout the years.

Early investigations of dynamic powder avalanching involved forming a heap of the powder to be tested on a platform adjacent to an adjustable ramp (1). As powder flows from the hopper onto the growing heap, a series of avalanches are initiated. The weight and time between successive avalanche is data logged as the powder cascades down the ramp and into a bin on a balance. As the avalanches progress, the data is acquired in real–time. The length of experimental runs, however, is limited by both the available amount of powder and the capacity of both the feed hopper and the collection bin. Since the apparatus is fairly bulky and not self–contained, it is difficult to control ambient temperature, humidity or the loss of fines. Also depending on its size, the ramp technique can require large powder samples.

The AeroFlow differs from the early ramp approach in that it is fully automated, self–contained and is sealed which prevents the loss of dispersible fines. Further, the rotating drum approach employed utilises comparatively small powder samples, roughly 60 ml, which could be important from a criticality perspective if the technique was ever exploited for PuO_2 powders. The equipment consists of a light–tight cabinet in which the powder flow measurements are carried out in a disk–shaped rotating drum (see Figure 1). The drum is partially filled with a power sample to be tested. As the drum rotates, the powder rides up the drum wall and cascades down in a series of avalanches. Powder movement is monitored by a light and photocell arrangement. As the powder tumbles down the drum wall, the degree of masking of the photo cells changes, hence, the voltage output from the photocells varies. The changing detector output forms the basis for recording dynamic powder flow, allowing the time between successive avalanches as well as the magnitude of each event to be recorded by the data acquisition software.

Figure 1. The sample drum and photocells

It is generally regarded that geometric shape descriptors by themselves e.g. aspect ratio, convex roughness, are not particularly sensitive measures of particle morphology. However, when used in combination with the fractal dimension or flow index could possibly discriminate between types of UO_2 powder or even indicate batch to batch variation. Trials with alumina have previously proved promising (2). The boundary fractal dimension is a measure of the ruggedness/irregularity of a particle profile and does not give any information on the overall geometric shape. In simplistic terms the fractal dimension of a boundary is generated by measuring the perimeter at a series of yardstick lengths. The number of steps multiplied by the step length, added to any remaining incomplete boundary segment, is an approximate boundary length $L(\varepsilon)$. As the dividers opening become smaller and smaller the values of $L(\varepsilon)$ converge rapidly for a circular shape. However for fractal curves the observed $L(\varepsilon)$ tends to increase without limit, to yield,

$$L(\varepsilon) = M\,\varepsilon^{(1-D)}$$

Where M is a constant and D is the fractal dimension of the boundary. By plotting Log L (ε) against log ε known as a Richardson plot, the fractal dimension can be obtained by adding one to the modulus of the gradient of the resulting data-line. The structured-walk investigation of a particle profile is not suitable for practical characterisation purposes as it too time consuming. Thus image analysis based techniques have been developed and for this study the method proposed by Flook was used (3). For particulates two linear portions are usually observed on the Richardson plot. The region at the smallest step-lengths is called the textural fractal, whereas the structural fractal, at coarser step-lengths, represents the gross boundary of the particle. Previous work has shown that it is the structural fractal dimension that is useful for characterisation purposes.

METHOD

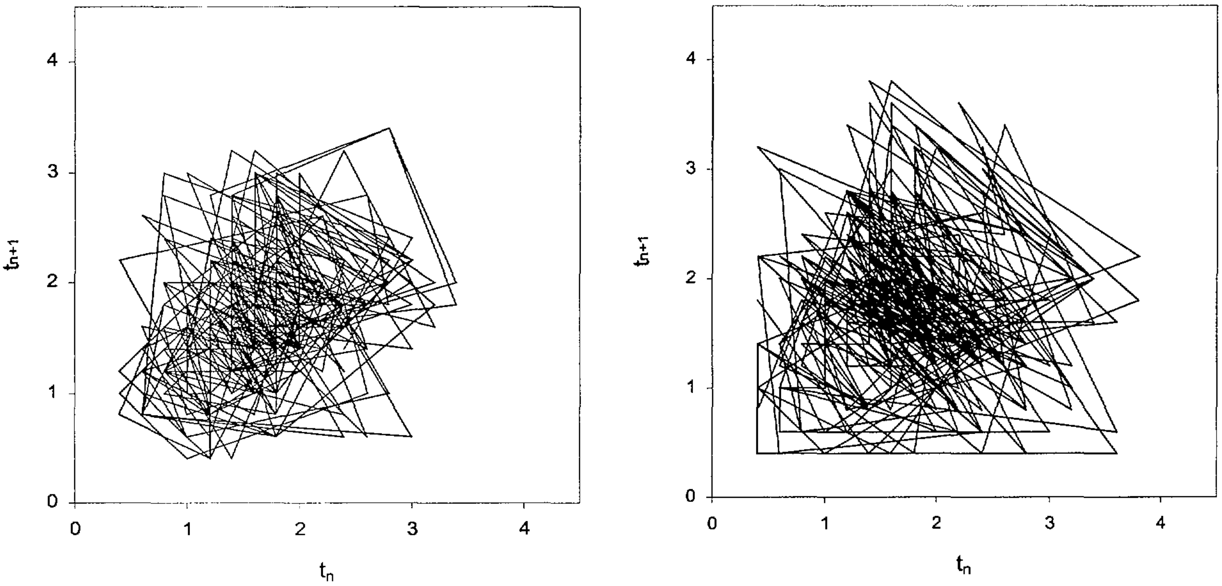
The UO_2 powder sample, Integrated Dry route (IDR) and ammonium uranyl carbonate (AUC) derived material were riffled into a 60 ml container. This provided an aerated standard volume of powder for testing The powder sample was then placed in the sample drum which was previously washed and allowed to fully dry prior to each run. The inside of the drum body was lined with a metal mesh strip which reduces powder slippage. Treatment of the Perspex surfaces with an anti-static agent was also used to reduce the amount of *powder adhering the surface of the Perspex window as the latter reduces the signal to noise ratio.* Tests were conducted at a drum speed of 1 revolutions per minute. The experiments were sixty minutes in duration but for analysis of the data only the last 10 minutes, representing 3000 data points, was sampled. This data is fed into an Excel spread sheet developed by the author.

SEM images of UO_2 derived from ammonium uranyl carbonate (AUC) precipitation and Integrated Dry route (IDR) powders were obtained and their outlines extracted using a manual perimeter tracing procedure. Forty individual particle images were collected for each powder to maximise the statistical significance of the results. The resultant profiles were then digitised by a Kontron IBAS 2000 image analysis system via video camera, onto a 512x512 square pixel rastor. The image analysis routine consisted of two parts, the Euclidean section, which determines the standard geometric attributes e.g. circularity, aspect ratio and size of the particles and the fractal section.

RESULTS

The raw Aeroflow data may be presented in several ways. The raw data consist of a time against intensity plot from which it is difficult to extract useful data. An informative graphical depiction of powder flowability, called an attractor diagram (4), is presented in Figure 2 for both UO_2 powders. The diagram is constructed by plotting the points t_n and t_{n+1}, where n is the event number, for a series of successive avalanches. This creates a cluster of points, the spread of which is a measure of how free–flowing a given powder is. A highly flowable powder will exhibit a compact cluster of points, whereas a powder that does not flow as well will have a less tightly grouped set of points. The centroid of the data cloud (the mean) is referred to as the attractor point and the scatter is called the strange attractor pattern. Analysis of the time attractor results would indicate that the IDR powder is more free flowing than the fine precipitated AUC material. This is demonstrated by a much more concentrated cluster and confirmed by the higher number of avalanche events with a lower mean time between those events as summarised in Table 1. The results were reproducible when repeated with fresh powder charges and thus differences between the powders was real and significant.

Figure 3. Phase-space attractor diagrams for IDR and AUC UO_2



The data, however, can not be considered purely in terms of the time attractor and the intensities of the avalanche events must be examined. These can also be investigated by similarly constructing intensity attractor plots, the data from which is also summarised in Table 1

Table 1 Summary of time and intensity attractor data sets

		Time			Intensity			
	No. of events	Mean	Max	σ_{n-1}	Mean	max	σ_{n-1}	Flow ranking
IDR	359	1.669	3.4	0.556	26.03	55	13.671	1
AUC	342	1.750	3.8	0.692	11.51	40	7.080	2

The intensity data is a measure of how much powder is displaced by an event. These results again indicate the superior flow properties of the IDR material with significantly more powder moving for each event . It should be realised that too much powder moving at one time could be an indication of poor flow, slumping, and would be more of a concern if the overall number of events was not significantly higher than for AUC.

The image analysis parameters for the two powders were cross correlated against each other using a least squares linear regression technique, to check for simple relationships which may indicate the non-independence of the morphological measures. The results show a high degree of correlation between the values of fractal dimension and circularity, but were found to be independent of particle size. The values for the mean and standard deviation are summarised in Table 2 which confirm the significance of circularity and fractal dimension. Using the w/s test for normality of a frequency distribution, the sets of values for the morphological parameters were each found to individually conform to the Gaussian distribution. The significance level, α, was found to be at 0.1 the highest level of significance for the fractal index, where as the circularity was deemed less significant. Use of this technique on a series of alumina powders has shown a much stronger dominance of the fractal dimension to individually characterise powders and it is intended to expand this study to other urania powders and increase the number of particles included in the studies.

Table 2. Summary of image analysis results

Powder	Circularity		Aspect ratio		Fractal dimension	
	mean	σ_{n-1}	Mean	σ_{n-1}	mean	σ_{n-1}
IDR	0.381	0.17	0.729	0.10	1.124	0.08
AUC	0.615	0.16	0.705	0.12	1.038	0.05

The link between flow, morphology and fractal dimension was not as predicted with the more rugged IDR powder flowing more predictably. The study with alumina powders had shown a strong relationship between fractal index and flow which had led us to believe that such a relationship could be established for UO_2 powders. It is believed that the tumbling action of the drum may be having a mild granulating effect on the powder increasing its flow and minimise the effect of surface roughness. The moisture content of the powder was unfortunately not recorded. This behaviour had been witnesses for a particularly cohesive alumina powder which had visibly granulated on extended tumbling in the analyser drum. The ruggedness of the powers also give a mechanism by which powders may lock together to form agglomerates which may also improve flow.

CONCLUSIONS

The AeroFlow analyser is capable of detecting differences between UO_2 powders. The two powders examined had quite varied morphologies and particle size distributions which the AeroFlow has distinguished differences between. More work is required to establish a link between flow and morphological characterisation factors as has been established for alumina powders. The fractal dimension has shown promise as a promising characterising tool and more work will be undertaken indexing relationships by combining this parameter with circularity and flow to track powder behaviour in terms of data clouds in multi-parameter data space.

REFERENCES

1. GJ Brown. The avalanching characterising of the handleability of blended coal fuels, Powder technology, 198 1(1), 157-165.
2. JW Hobbs and D Rhodes. Unpublished results.
3. AG Flook. The use of dilation logic on the Quantimet to achieve fractal dimension characterisation of textured and structured profiles., Powder technology, 1978, 21, 295-298.
4. B.H. Kaye, J. Gratton–Liimatainen, N. Faddis, 'Studying the Avalanching Behaviour of a Powder in a Rotating Disc.' Particle System Characterisation, 1995, 12, 232-236.