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UNDERSTANDING THE EFFECT OF COKE MICROSTRUCTURE UPON COKE QUALITY USING IMAGING AND MODELLING TOOLS[1]

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Abstract
We present preliminary work that examines the effect of components of the coke microstructure, such as voids, inerts and microfissures, upon the strength and reactivity of the coke. Using imaging of coke samples, we obtain sample information about coke microstructure, including identification of most features of the void space as well as location, size etc of inerts and microfissures. We use a 3D finite element model to calculate mechanical properties and locations of stress concentration within the structure, giving an identification of the coke strength. We also calculate gas permeability, which is an important determinant of the coke reactivity. We use a modelling approach to understand the effects of size, shape and distribution of the individual components in the coke on the strength and permeability of the coke, in order to better understand their effect on coke quality. Ultimately, we wish to relate these back to the properties of the coal charge.
Key words: Coke microstructure; Strength; Reactivity; Permeability; Finite elements.

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1 INTRODUCTION

In assessing the value of coke for use in a blast furnace, measures of coke quality including mean coke size and size distribution, coke strength and reactivity are desirable quantities to know. It is relatively straightforward to directly measure coke size distributions using screens or imaging technology, however it is not so straightforward to obtain accurate indicators of coke strength and reactivity. Commonly used indicators of these latter quantities, such as CSR, CRI and drum indices are actually *indirect* measures, in that they measure some other property and relate that property to coke strength or reactivity. This leads to some difficulty in assessing the value of these measures, even if they are generally accepted within the industry. Clearly, it would be desirable to obtain more *direct* measures for coke strength and reactivity, and this is the aim of the work presented here.

Modern technology for imaging, in both 2D and 3D, has the capacity to provide a great amount of detail about the coke microstructure, down to very small scales (of the order of a few microns)[1,2]. At this level of resolution, it is possible to distinguish virtually all of the features of the microstructure. Once the details of the microstructure are known, it is then possible, using various computational techniques, to determine direct estimates of certain quantities of the composite structure from the properties of the individual components of the structure. One particular method used is finite element analysis, which allows the solution of appropriate mechanistic equations, such as mechanical deformation, gas permeation, thermal and electrical conduction etc, based on dividing the microstructure geometry[3] into a regular mesh, with each element of the mesh having properties pertaining to the local component of the microstructure. In this way, the approach can estimate the elastic moduli of the material or its permeability. The elastic moduli (Young's modulus, bulk modulus and shear modulus) are indicators of material strength, while the gas permeability is a key component in determining the reactivity of a porous solid. In particular, for coke, transport of gas to the internal coke surfaces is the rate controlling step for gas reactions in the blast furnace [4], so the gas permeability is a key indicator of coke reactivity. Tsafnat et al[5] have shown the potential for calculating mechanical properties of coke from a knowledge of the 3D microstructure obtained from X-ray micro-CT scans.

Once a knowledge of the microstructure of coke is available, it becomes possible to create simulations of the microstructure, with the simulations having similar properties to the real microstructure. By combining such simulations with a finite element analysis, we are able to modify properties of the individual components of the microstructure in order to assess how individual components affect the strength and reactivity of the coke. This provides us with the opportunity to provide feedback into the preparation of coal charges, which may produce desirable properties of the final coke. In this paper we examine the effect of varying the mean bubble size, mean inert size, amount of bubble overlap (coalescence) and quality of the mosaic/inert interface on coke quality indicators using this simulation technique.

2 IMAGING OF COKE MICROSTRUCTURE

We are interested to determine as much information as possible about the microstructure of the formed coke, using imaging technology. The most common method is using high-resolution microscopy of cut and polished coke samples. Figure 1 shows two examples, for two different cokes. The resolution of the features

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in the images is very high, and we can clearly distinguish the pores within the coke, in particular. The pores appear to consist of a collection of more-or-less deformed circular shapes, as well as more complicated shapes. The majority of these pores are due to the bubbles formed during the plastic phase of carbonization [6], with the more complicated shapes being due to the partial coalescence of bubbles as they grow [7,8]. The shapes are "frozen in" to the microstructure when the plastic phase re-solidifies. Close examination of these images indicates that there are pores having a wide range of sizes. Moreover, the coke in Figure 1(a) has fewer pores, but larger mean size than the coke in Figure 1 (b) and this is confirmed by image analysis (see Table 1). Another, less obvious feature of the two microstructures is the presence of so-called *inertinites* – elements of the original coal that did not soften during carbonization. These form a significant proportion of the volume of many cokes (see Table 1) and presumably play a role in determining the properties of the coke.

We wish to determine key properties of the microstructure from these images, which are 2D in nature, but the microstructure itself is 3D. An alternative that is becoming more accessible is to use X-ray micro-CT imaging which can provide 3D images of the microstructure with resolution as low as 3μm. An example of the result of such imaging is provided in Figure 2. This capability provides an enhanced method for determining the 3D structure of the coke, which we plan to use to examine a range of cokes in order to investigate the variation in the properties of the microstructure and their role in determining coke quality. At this early stage we do not have 3D microstructure information, so the remainder of our analysis is based on interpretation of the microstructure derived from the 2D images of Figure 1.

Table 1. Measured properties of the cokes shown in Figure 1. The data on pores only includes individual pores having an area exceeding 300 sq. mm

	Coke from High Vol blend	Coke from Low Vol blend
Coke Porosity (%)	54.5	55.5
Coke Avge pore area (sq mm)	15.3×10^3	1.6×10^3
Coke Pore density (#/mm^2)	36	352
Coke ASTM Stability (%)	51.2	61.9
Coke ASTM Hardness (%)	68.4	64.0

3 MODELLING OF COKE MICROSTRUCTURE

In order to create a model microstructure, we consider the coke to comprise 4 different components, which we shall call *inerts*, *matrix*, *bubbles* and *gaps*. The term *inerts* is short hand for inert derived maceral components (IDMC), while the *matrix* is considered to be the reactive derived maceral components. These latter components are those that softened and resolidified during carbonization. *Bubbles* are modelled as spherical voids, some of which can overlap. The overlapping accounts for bubbles which have partially coalesced during the softening and resolidification. The *gaps* are small voids adjacent to the surface of the *inerts*. They are assumed to have been caused by the differential shrinkage between the *inerts* and the *matrix* [6].

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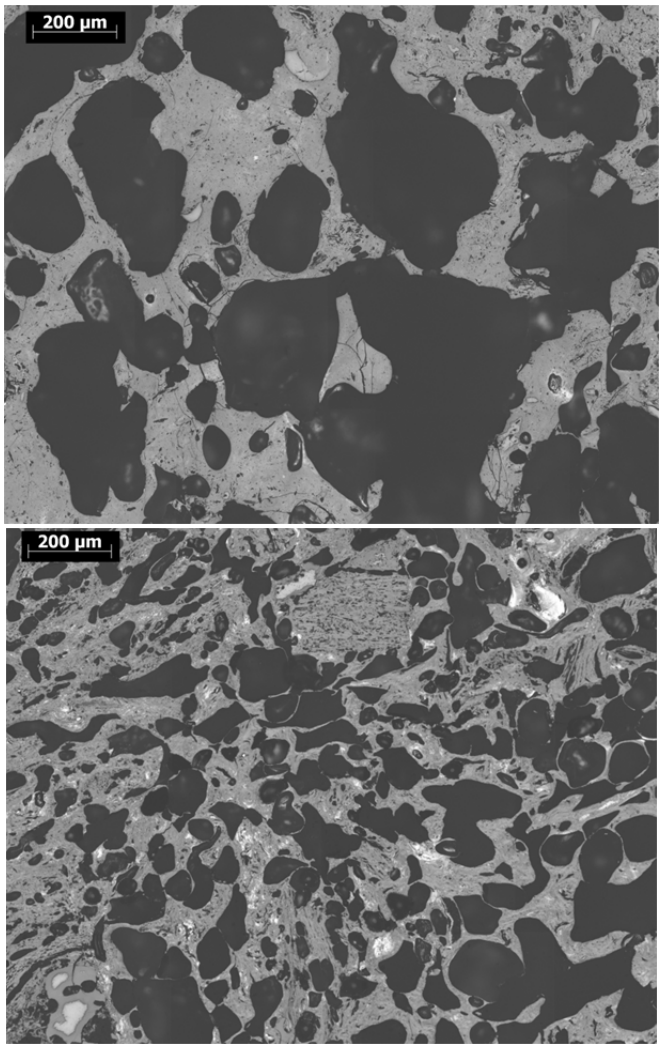

Figure 1. Optical microscope images of coke produced using (a) High volatile blend and (b) low volatile blend.

We do not distinguish between organic and inorganic inert components in the model microstructure. Inert particles are modelled as a collection of circular cylinders, having length and diameter that are chosen independently from gamma distributions with a specified mean and variance. Similarly, the bubbles are a collection of spheres whose diameter is chosen from a gamma distribution having a specified mean and variance.

The process of forming the microstructure can be summarized by the following:

- STEP 0: start with an empty cube comprising N x N x N voxels.
- STEP 1: Place the collection of *inerts* (with random dimensions) by locating their centres at random locations, having random orientation, within the cell, insuring they do not overlap.
- STEP 2: locate *gaps* as individual voxels, randomly located on the surface of *inerts*.
- STEP 3: Place a subset of the *bubbles* by locating their centres at random locations within the cell, ensuring they do not overlap with other *bubbles* or *inerts*.
- STEP 4: Place the remainder of the *bubbles* by locating their centres at random locations within the cell, ensuring they do not overlap with *inert* particles. They can, however, overlap with existing *bubbles* or *gap* voxels.

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The reason for the two-step process for placing bubbles is to control the amount of overlap.

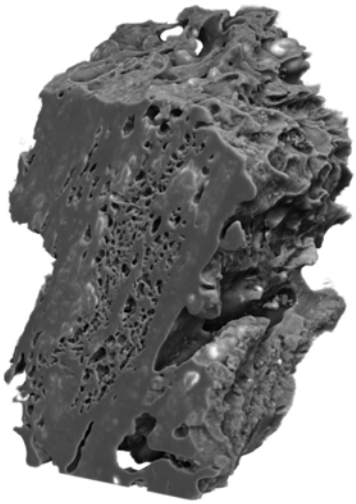

Figure 2. A rendered image of a sample of coke produced using an X-ray micro-CT device. The sample size is approximately 3mm and the resolution of the image is approximately 3μm.

In each case, a voxel is considered to be fully comprised of a particular component if the centre of the voxel is within the volume of the component. Also, if some part of the inerts or bubbles lies outside of the cube, then we assume periodicity in all 3 directions, so that the remainder of the volume 'enters' the cube at an opposing face. In this way the cube represents, in some sense, a much larger volume.

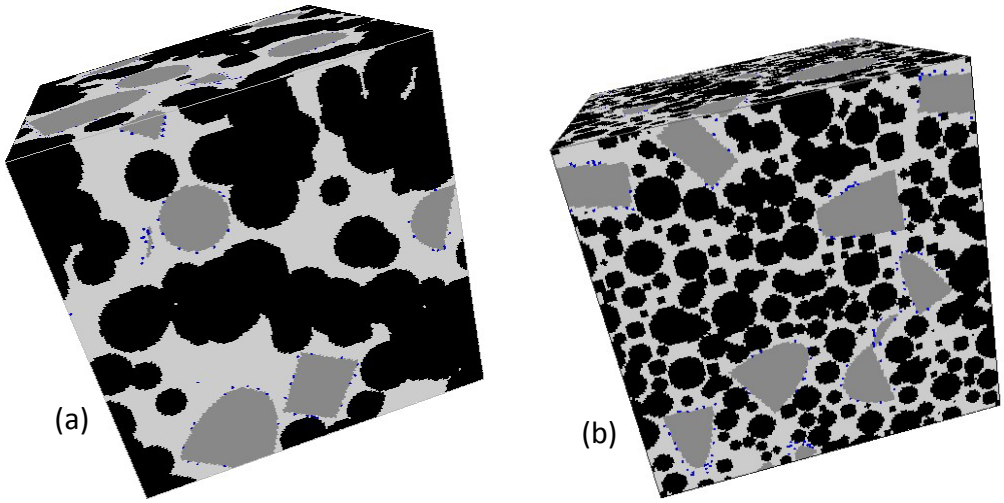

Figure 3. Calculated 3D microstructures of coke, containing inert particles (dark grey), mosaic (light grey), bubbles (black) and interface gaps (blue). The volume fractions of each component are the same in each case, but the mean bubble size in (a) is 3 times the mean bubble size in (b).

Figure 3 shows two examples of model microstructure created using this procedure, with N=160. In both examples, the volume fraction of each component is the same, namely *inerts* 16%, *bubbles* 50%, *gaps* ~2%, *matrix* ~32%. The only difference between the two examples is that the mean bubble size in Figure 3(a) is 3 times that in Figure 3(b). In each example, as many non-overlapping bubbles as possible were placed in STEP 3, before the remaining bubbles were placed to make up the total bubble volume. For the smaller mean bubble size, it is possible to locate a larger

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volume fraction of non-overlapping bubbles than for the larger mean size, using the method developed here.
These particular configurations were chosen in order to mimic the microstructures of the two coke images shown in Figure 2. The total porosity in the model microstructures is somewhat lower, in order to account for porosity that is (a) below the resolution of the model and (b) encapsulated within the inert particles. For this work, we do not consider these two contributions to the porosity.
Superficially, there are similar features between the model and real microstructures – there are larger coherent structures – both of pores (collections of bubbles) and matrix components in Figure 3(a) than in Figure 3(b) and the cluster of bubbles appears similar. It would be desirable to make the comparison more quantitative.

4 PROPERTIES OF COKE MICROSTRUCTURE

The model coke microstructure can be used as input to a proprietary finite element code *MesoProp* which is designed to calculate specific material properties of a composite material [3]. For the coke simulations, we consider the voids (*bubbles* and *gaps*) to have the properties of air, while the *matrix* and *inerts* have the mechanical properties of an isotropic solid. Using this software, we calculated the elastic moduli and the permeability of the simulated microstructures, this time produced with N=80, due to computational limitations. The three elastic moduli are **Young's modulus**, which we relate to bulk fracture of the coke, **bulk modulus**, which we relate to the compressive strength of the coke and **shear modulus**, which we relate to the abrasive strength of the coke. In general, we perform several simulations for each property, in order to account for the heterogeneity of the material.

4.1 Comparison with Observed Microstructure

We considered a set of microstructure simulations having the same properties as those in Figure 3, but we vary the mean size of the bubbles. In this way, we hope to replicate the variation seen across the range of cokes from our high blend situation to the low volatile blend situation. A key difference between these two extremes is understood to be the size of the bubbles in the coke and the concomitant overlap (coalescence) of the bubbles. The results are summarized in Figure 4.
The cases corresponding to the high volatile blend microstructure shown in Figure 3(a) and the low volatile blend microstructure shown in Figure 3(b) are highlighted on the graph. It is more difficult to pack non-overlapping bubbles into the volume as the mean bubble size increases, and as a result the amount of bubble overlap increases as the mean bubble size increases. The value of all the moduli increases as the mean bubble size decreases. The relative increase is most pronounced for the bulk modulus, and least pronounced for the shear modulus. These results indicate that not only the average properties of the coke are important in determining coke strength, but the distribution of the individual components within the structure.

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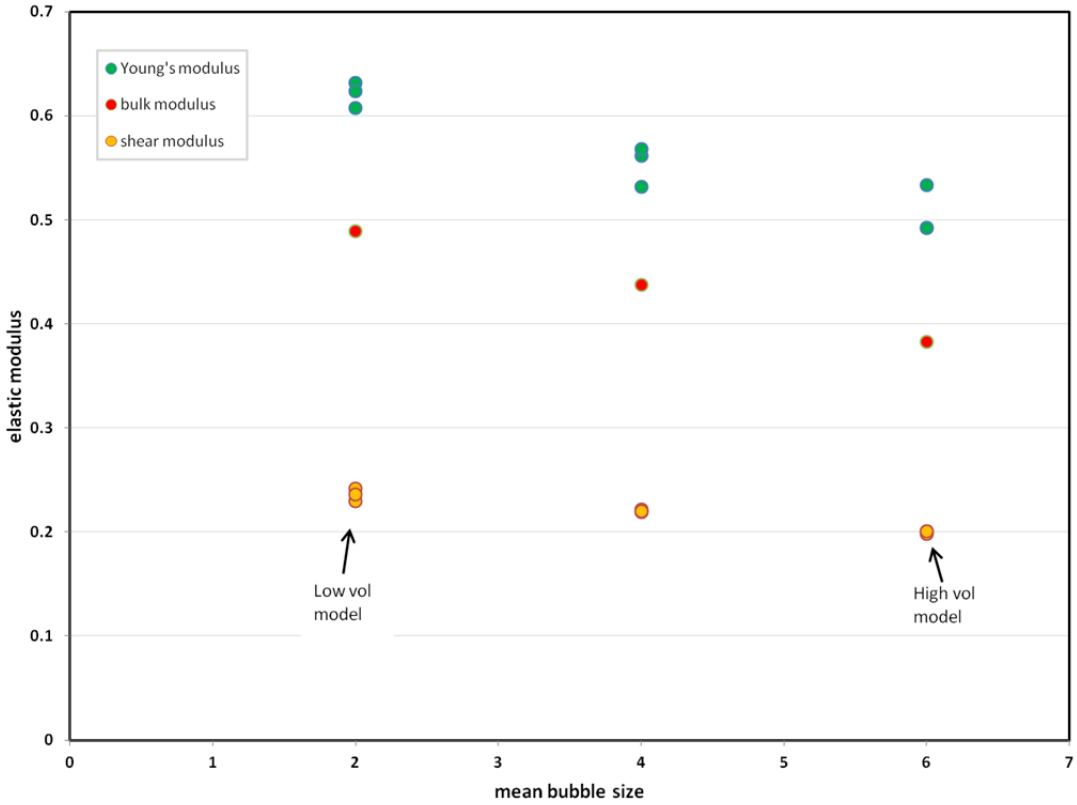

Figure 4. Calculated elastic moduli of the coke microstructure, for varying mean bubble size. Note that there are 3 values for Young and shear modulus per simulation, but only one for the bulk modulus.

4.2 Effect of Bubble Overlap

It has been postulated [7-9] that the coalescence of bubbles is an important issue affecting the coking pressure during carbonization as well as aspects of coke quality. When bubbles coalesce in a fluid having a low viscosity (such as water) they will rapidly form a new single bubble. However, as the viscosity decreases, the time taken to form a new sphere increases in inverse proportion to the viscosity. As a result, it appears that there are many instances in the coke microstructure where bubbles have overlapped, but not had sufficient time to form a spherical bubble before re-solidification, resulting in the "frozen-in" shapes shown in Figure 1.
Therefore, we wish to investigate the effect of the amount of bubble overlap on coke quality. We do this by varying the number of bubbles that are forced to be non-overlapping in STEP 3 of our microstructure formation algorithm, while keeping all other aspects of the microstructure, and in particular the total porosity, the same. A simple indicator of the amount of overlap of bubbles is the specific surface area of the pores, which decreases as the amount of overlap increases, for a specified constant porosity. Figure 5 shows the calculated elastic moduli of a set of simulated microstructures, having 16% *inerts*, 58% *pores*, ~2% *gaps* and ~24% *matrix*. The mean size of the pores is the same in all simulations, which is different to the previous example. In this way we can isolate the effect of the overlap upon coke properties.

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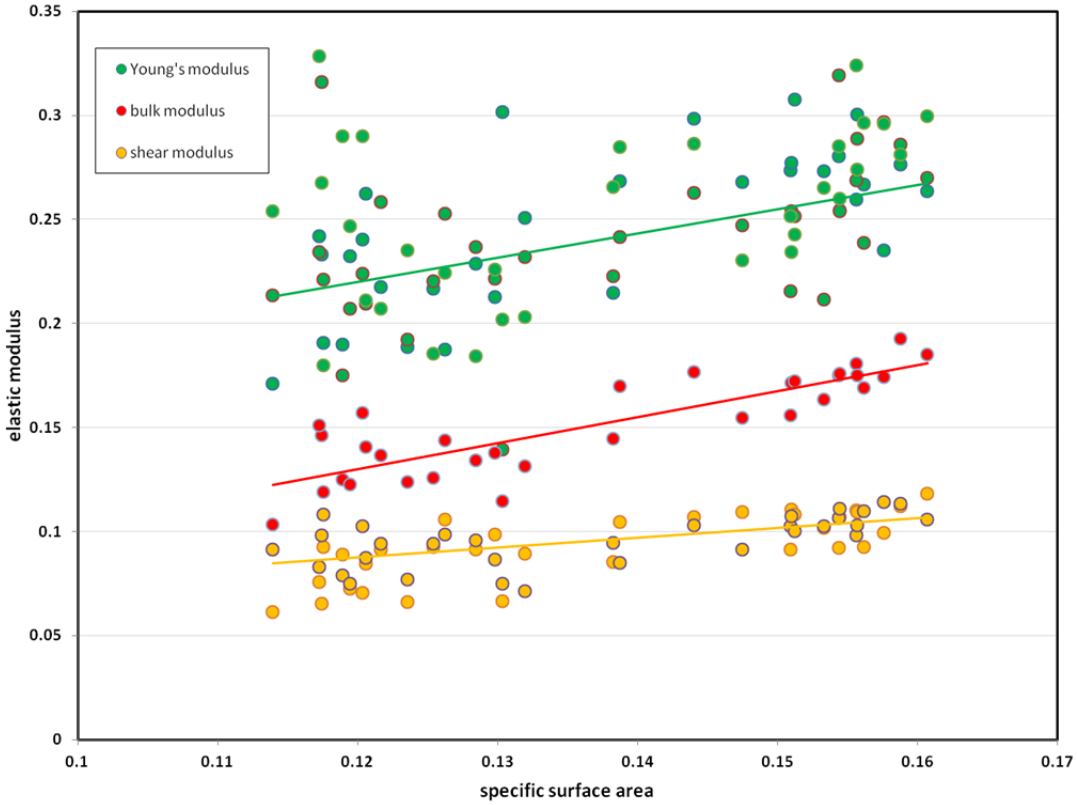

Figure 5. Calculated elastic moduli for coke microstructure having the same amounts of the various components comprising the microstructure. The specific surface area of the bubbles is an indicator of the amount of overlap of the bubbles in the microstructure.

There is a clear trend in the results showing that the elastic moduli increase as the amount of overlap decreases. This occurs because there are fewer, larger pore structures as the overlap increases, which tends to decrease the elastic moduli. Correspondingly, it would be expected that this would reduce the coke strength. In contrast, we found that increasing the amount of overlap, with porosity held constant, made effectively no difference to the gas permeability and hence coke reactivity.

4.3 Effect of Inert Mean Size

Experiments have shown [10] that the coke strength can be affected by the size of the inert rich fraction of the coal charge. We examine this effect here by considering a situation by performing a set of simulations where we vary the mean size of the inerts in the microstructure, while keeping the volume fractions of all the components the same. Figure 6 shows the results of the simulations. Most interestingly, the elastic moduli all increase significantly when the inert size is sufficiently small. It is clear that the smaller inert size causes a change in the distribution of pores (bubbles) within the microstructure. The large number of small inerts leaves only small spaces between them for bubbles, which leads to less overlap and more individual bubbles. As we saw in the previous section, less overlap leads to larger elastic moduli. The specific surface area of the bubbles generally increases as the inert mean size is increased in these simulations. The inference we draw is that it is better to have inert particle size larger than the mean bubble size of the coke. As the inert particles approach the mean bubble size the coke is weakened. This may explain the result of Mahoney, O'Brien and McGuire [10] where the effect of inert size on coke strength is positive for some coals and negative for others.

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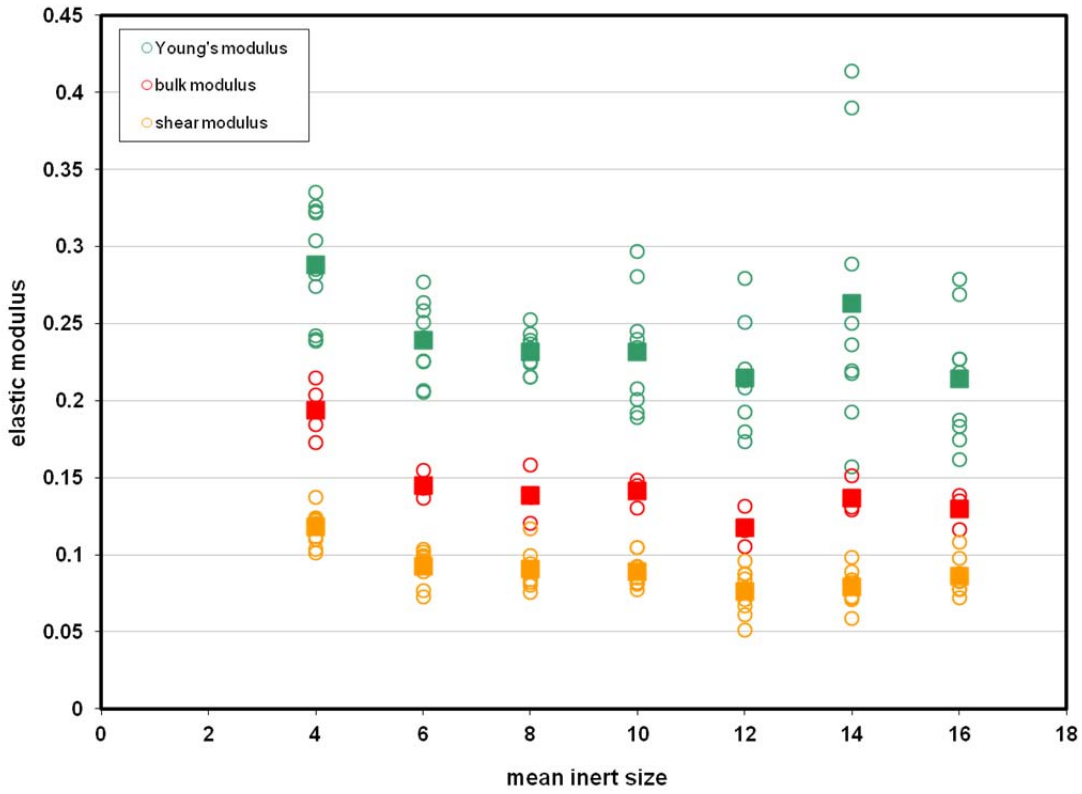

Figure 6. Calculated elastic moduli for coke microstructure having the same volume fractions of the various components comprising the microstructure, but differing mean inert size. The filled squares represent the average value at each mean size.

4.4 Quality of Inert/Mosaic Interface

In this case, we vary the coverage of the surface of the inert particles by “micro gaps” (the blue elements in the microstructure of Figure 3). So, although the volume fraction of bubbles is held constant, the total porosity will slightly increase as the number of gaps increases. These gaps could be formed by micro-fissuring, due to the differential shrinkage between inert and softened particles, or perhaps by factors such as the degree of wetting of the inert by the mosaic [11,12]. We interpret the coverage of the inert surfaces by micro gaps as an indicator of the quality of the interface between the softened particles and the inerts. Figures 7 and 8 show the calculated elastic moduli and gas permeability for a range of coke microstructures. In each case, the properties of the microstructure are the same, except for the fractional coverage of the surface of inerts by micro gaps. The results indicate that the quality of the inert/mosaic interface has only a small effect on the coke strength, since the elastic moduli vary only slightly over a large range of surface coverage, but the gas permeability is more significantly affected. Thus this effect is more likely to affect coke reactivity, and hence coke strength after reaction, rather than coke cold strength.

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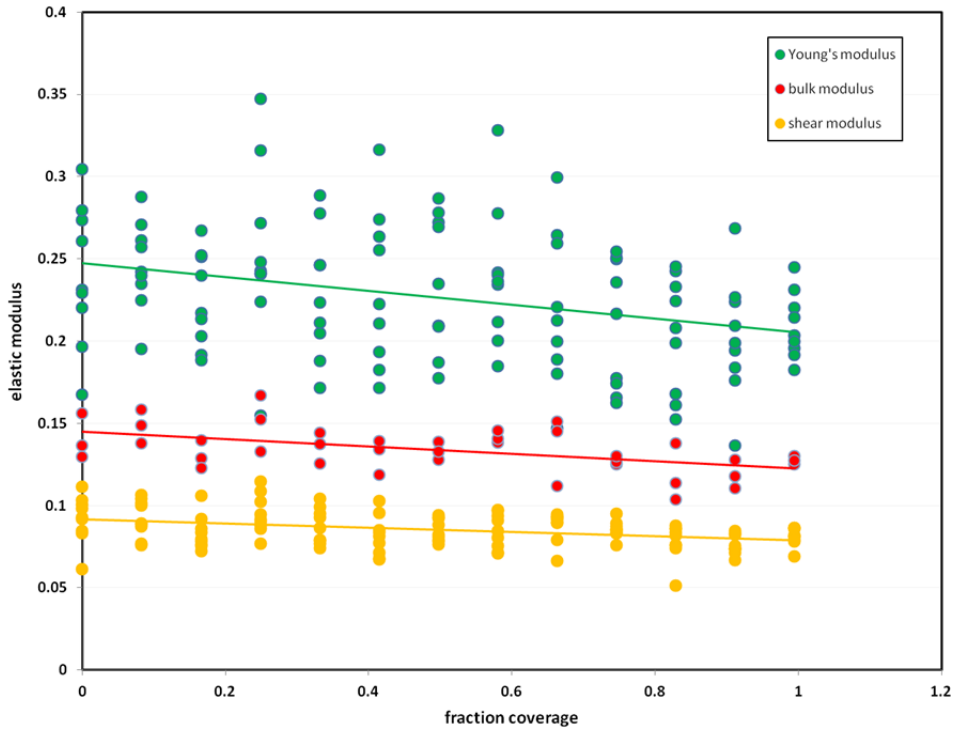

Figure 7. Calculated elastic moduli for coke microstructure having different amounts of coverage of the surface of inert particles by gaps.

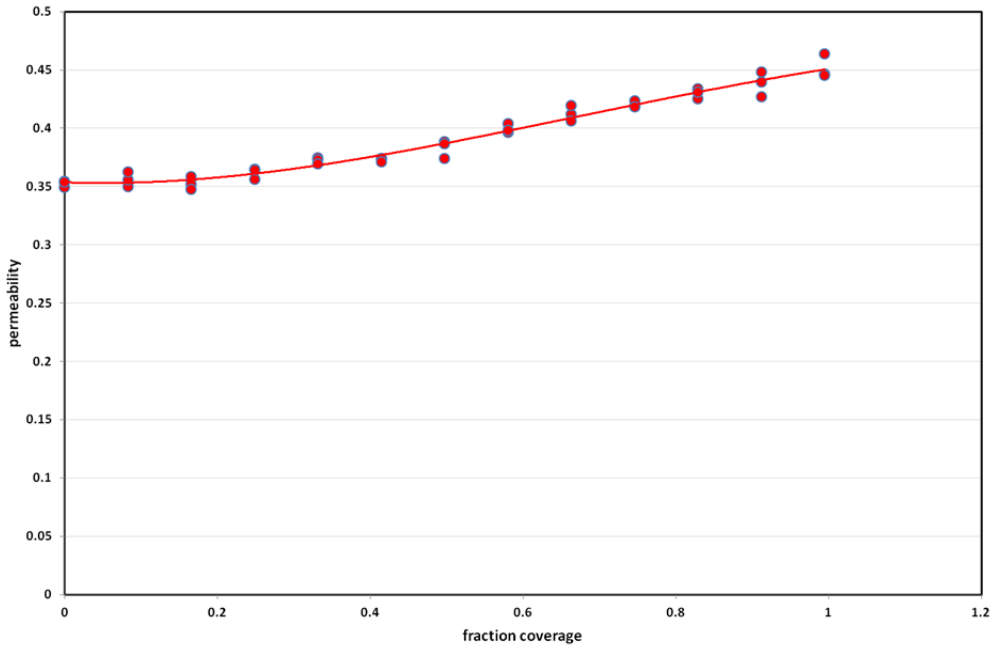

Figure 8. Calculated permeability for coke microstructure having different amounts of coverage of the surface of inert particles by gaps.

5 DISCUSSION

Our results provide a useful insight into the ability to estimate coke strength and permeability properties from detailed information of coke microstructure. At the present stage of development, we have used 2D images to collect information about the microstructure, but we are currently developing 3D micro-CT capability to obtain microstructure details for a range of cokes produced from Australian coking coals.

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Ultimately, we anticipate this research will allow improved strategies for optimizing coke quality.

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