Jurnal Sains Nuklear Malaysia, 2012, 24(1): 81-90
ISSN: 2232-0946

BIAXIAL TESTING FOR NUCLEAR GRADE GRAPHITE BY BALL ON THREE BALLS ASSESSMENT

Mohd Reusmaazran Yusof, Yusof Abdullah and Gary Kipling*

Malaysian Nuclear Agency
Bangi, 43000 Kajang, Selangor, Malaysia.

* Department of Engineering,
University of Hull, U.K.

ABSTRACT

Nuclear grade (high-purity) graphite for fuel element and moderator material in Advanced Gas Cooling Reactors (AGR) displays large scatter in strength and a non-linear stress-strain response from the damage accumulation. These responses can be characterized as quasi-brittle behaviour. Current assessments of fracture in core graphite components are based on the linear elastic approximation and thus represent a major assumption. The quasi-brittle behaviour gives challenge to assess the real nuclear graphite component. The selected test method would help to bridge the gap between micro-scale to macro-scale in real reactor component. The small scale tests presented here can contribute some statistical data to manifests the failure in real component. The evaluation and choice of different solution design of biaxial test will be discussed in this paper. The ball on-three ball test method was used for assessment test follows by numerous of analytical method. The results shown that biaxial strength of the EY9 grade graphite depends on the method used for evaluation. Some of the analytical methods use to calculate biaxial strength were found not to be valid and therefore should not be used to assess the mechanical properties of nuclear graphite.

Keywords: ball on three balls, biaxial, ey9, graphite, strength

INTRODUCTIONS

Graphite has been used since the start of nuclear power generation as a very effective moderation. It has been used extensively as moderator reactors for Advanced Gas-cooled reactors (AGR) and is still being used for the vigorous advancing High Temperature Gas Reactor (HTGR) (Albarhoum, 2011). The first pile constructed by Enrico Fermi, while no enrichment process was used, was moderated by graphite. Generally, graphite had been used in different types of nuclear reactors: UNGR (France, Spain), RBMK (Lithuania, Ukraine, and Russia), Magnox (Italy, Japan, Korea, and United Kingdom) and HTR (Germany, United-Kingdom, Japan, China and United States (Frechao et al. 2007).

An important characteristic of graphite is that its strength is stochastic which means that an individual specimen can show a large random fluctuation in strength from a population mean. Graphite also has a nonlinear stress–strain response because of distributed damage and damage accumulation within the material prior to rupture. This behaviour can be described as “quasi-brittle” or “ductile-like” which is contrast with the classically brittle materials, such as ceramics and glasses that fail abruptly without prior damage accumulation, although they similarly display large scatter in strength (Nemeth &

Correspondence author: reusmaazran@nuclearmalaysia.gov.my

Bratton, 2011). Other materials, such as fibre-reinforced composites, can accumulate significant damage prior to failure and have less scatter in strength than the individual constituents of the composite have. Graphite ruptures behaviour falls somewhere between the behaviours of ceramics and fibre-reinforced composites (Nemeth & Bratton, 2011). The stress-strain relationship of graphite is characterised by non-linearity, by hysteresis loop formed under cyclic stressing and by attendant residual deformation when applied stress in release (Seldin, 1966). When irradiated to even a small fluence the stress strain behaviour becomes linear and the hysteresis is lost. Therefore, the development of the design methods and validation to bridge this phenomenon of the graphite structures used in the reactor are very challenging and important. Quasi-brittle materials by their very character show deviation from a truly elastic material and so challenge some of the assumptions being made.

At present, in the nuclear industry, the prediction of when graphite bricks will crack in a nuclear core, due to the build-up of shrinkage and thermal stresses, is largely based on the measurement of mechanical properties from small irradiated samples, even though the volume of a typical brick is a factor of 10^4 greater than that of a typical flexural test sample (Neighbour, 2010). Determination of the strength of most brittle materials including graphite from data obtained from flexural strength tests or uniaxial or biaxial tensile tests. Biaxial strength testing has been used for many years using a wide variety of test configurations. Biaxial strength testing is claimed to have some benefits compare to uniaxial testing including ease of test piece preparation and testing of a large surface area free from edge finishing defect (Borger, 2002). There are many configurations available to determine the biaxial strength of material such as diametric compression test (Awaji et al. 2007), ball on three ball (Godfrey, 1985), ring on ring (Fett et al. 2006), ball on ring (Isgro et al. 2003), bulge test (Imaninejad et al. 2004 and arcan test (Doyoyo and Mohr, 2003). Ball on the three balls was chosen in this work with several analytical solution has been described by a number of authors (Godfrey, 1985; Ovri, 2000; Higgs et al. 2000; Pagniano et al. 2005; ASTM F394-78, 1996 and Borger et al. 2002). The advantages of ball on the three ball test were adaptable to different specimen geometries, non-potential for edge effects and suitability for brittle material.

Theory

Based on numerous biaxial tests on a cylindrical symmetrical thin-plate, an empirical equation for elastic materials was proposed by Kirstein and Woolley (1966), as given below;

$$\sigma = YL/T^2 \qquad \text{(Equation 1)}$$

Where σ is the biaxial stress, L is the apply load, T is thickness and Y is a geometric factor. Y values can be derived in several way depending on the test conducted for example sample geometry such as radius and thickness or design of apparatus used such as ball size and the distance from ball to centre. In ASTM S F394-78 (reapproved 1996) the Standard Test Method for Biaxial Flexure Strength (Modulus of Rupture) of Ceramic Substrates it has been suggested that the biaxial strength (S) are as follows (1);

$$S = -0.2387\, P.\left(\frac{X-Y}{d^2}\right) \qquad \text{(Equation 2)}$$

where;

$$\mathrm{Y}=(1+\upsilon_{\mathrm{d}})\left(1+\ln\left(\frac{\mathrm{A}}{\mathrm{R_d}}\right)^2\right)+(1-\nu_{\mathrm{d}})\left(\frac{\mathrm{A}}{\mathrm{R_d}}\right)^2$$

$$X = (1+\nu_d)\ln\left(\frac{A}{R_d}\right)^2 + \left[\frac{(1-\nu_b}{2}\right]\left(\frac{A}{R_d}\right)$$

(ν_d is Sample poisson’s ratio, A is Support Radius, R_d is Radius of disc (Specimen), ν_b is Ball poisson’s ratio)

This document recommended the support points are provided by three balls bearing positions at the angle of 120°. The load applied to specimen suggested is centre by a right circular cylinder of hardened steel (dowel) with its, end flat and perpendicular to axis. The area of applied load is therefore stated as the radius of the dowel. Specimen thickness was also not specified but required the limit of the deflection of the specimen centre must be one half on the specimen thickness at fracture. Ovri et al. (2000) found good agreement with ASTM F394-78 when he applied the same solution to alumina and glass, however, his analyses did not take the test apparatus material properties into account.

Pagniano et al. (2005) applied another solutions regarding to the issue of the uniform loading to the specimen. He did study the flexural strength of perodonthic ceramic material. He obtained the biaxial flexural strength using the ball-on-ring method. Each disk was centred on a ring of steel balls. The discs were loaded with spherical indenter centred on the top surface of the disc. He suggested that the radius of uniform radial and tangential stresses at centre is equivalent to the radius of the loading ball (R_b) as given in equation (3).

$$\sigma = 3L\,\frac{(1+\upsilon_d)}{4\pi T^2}\left[1+2\,ln\left(\frac{A}{R_b}\right)+\left(\frac{1-\nu_d}{1+\nu_d}\right)\left(1-\frac{R_b^2}{2A^2}\right)\left(\frac{A^2}{R_d^2}\right)\right] \qquad \text{(Equation 3)}$$

When calculating the biaxial strength, the local stresses at contact caused by the pressure between the load point and disc specimen are importance and must be addressed. Using the Hertzian theory (elastic body interaction) Godfrey (1985) tested of Silicon Nitride, Si_2N_3 using an approximation for the stress between loading ball and specimen, R. This is given by equation (4). The properties of test apparatus also were accounted for.

$$\sigma = 0.4775\left(\frac{L}{T^2}\right)(1+\nu_d)ln\left(\frac{A}{R}\right)+\frac{1}{2}(1+\nu_d)+\frac{0.25(1-\upsilon_d)(2A^2-R^2)}{R_d^2} \qquad \text{(Equation 4)}$$

$$R = 0.721\left[\frac{PD_b(1-\nu_b)}{E_b}+\frac{(1-\nu_d^2)}{E_d}\right]^{\frac{1}{2}}$$

Where R is Godfrey Contact Radius Approximations, D_b is Ball Diameter, E_d is Young Modulus (Specimen/disc), and E_b is Young Modulus (Ball)

The equation given in (4) is valid when the R value is greater than 1.7T. For the *R* value smaller than 1.7T, Higgs et al. (2001) and Borger et al. (2002) studied PMMA cement and Alumina respectively suggest to use Westegaard approximation contact area (5) that’s refer to equivalent area of contact, b and become the biaxial strength solutions as shown in equation (6), by using of the equivalent radius makes possible the calculation of the finite maximum stresses produced by a point loading.

$$\text{Westergaard Contact Radius Approximations } (b) = \sqrt{1.6R^2 + T^2} - 0.675T \qquad \text{(Equation 5)}$$

$$\sigma = 3L\,\frac{(1+\nu_d)}{4\pi T^2}\left[1 + 2\ln\left(\frac{A}{R}\right) + \frac{(1-\nu_d)}{(1+\nu_d)}\left(1 - \frac{R^2}{2a^2}\right)\frac{A^2}{R_d}\right] \qquad \text{(Equation 6)}$$

$*\quad a = R_d.T$

In this present work, the EY9 nuclear grade graphite was study in several analytical solutions suggested by several authors mentioned above. The purpose of this work is to study the validity of numerous analytical solution of biaxial strength. The ball on three balls test method was used as a test apparatus to assess the solution upon the graphite.

MATERIALS AND METHODS

The nuclear grade graphite (EY9) was purchased from the Morgan Crucible Company. It came in the form of cylindrical rod, and it is 67 mm in diameter. The graphite was first cut into disc plates with a lath machine in different thickness of 4, 5, 6 and 7 mm, in order to study the change of the biaxial strength of the different volumes. The specimen's surface are then smoothed with silicon carbide to remove the machining effect. The ball on 3 balls apparatus comprises of thin disc sample that is supported on three equally spaced ball bearings. The opposite face is centrally loaded, with stainless steel ball bearing to the orientation of the plane, as illustrated in Figure 1. The test was conducted using Llyods EZ 50 Universal Testing Machined, fitted with10 kN load transducer. Tests were started with a speed of 0.5 mm/minutes, and 2.0 N pre-load, and then compressed until it fractures. The fracture surface properties were examined under an optical microscope (Figure 2). The disc was broken by using fixtures with a support radius and ball bearing radius of 21.80 and 3.0 mm respectively. The elastic constant of ball bearing, E_b and specimen, E_d are 200 and 20 GPa respectively. The poisson ratio used for the ball, ν_b is 0.3 and the specimen, ν_d is 0.2. The load at this point (maximum load) is the value required to calculate the biaxial strength of the sample. The maximum principle stress (σ_{max}) in the disc, which occurs on the disc surface opposite to the centred loading ball, is used to define the strength. NEXYGEN MT materials testing software was used to display and record the properties of each test. The strength of the graphite specimen were then calculated using a number of solution proposed by Godfrey (1985), Ovri, (2000), Higgs et al. (2001), Pagniano et al. (2005), ASTM F394-78, (1996) and Roger et al. (2001).

As for comparison with uniaxial test, a compression test was set up for cylindrical specimen with dimension of 8 mm for diameter and 12 mm in height. The compression plate was equipped with spherical setting to prevent misaligned loading. The test was conducted at 0.5 mm/min crosshead velocity fitted with 10 kN of load transducer. The specimens were compress until it fractures. Strength for uniaxial then calculate by the equation;

$$\sigma_{max} = L_{max} / \pi r^2 \qquad \text{(Equation 7)}$$

Where = L_{max} is the applied max load and r is the specimen radius. The test was conducted at similar speed until the specimen fracture.

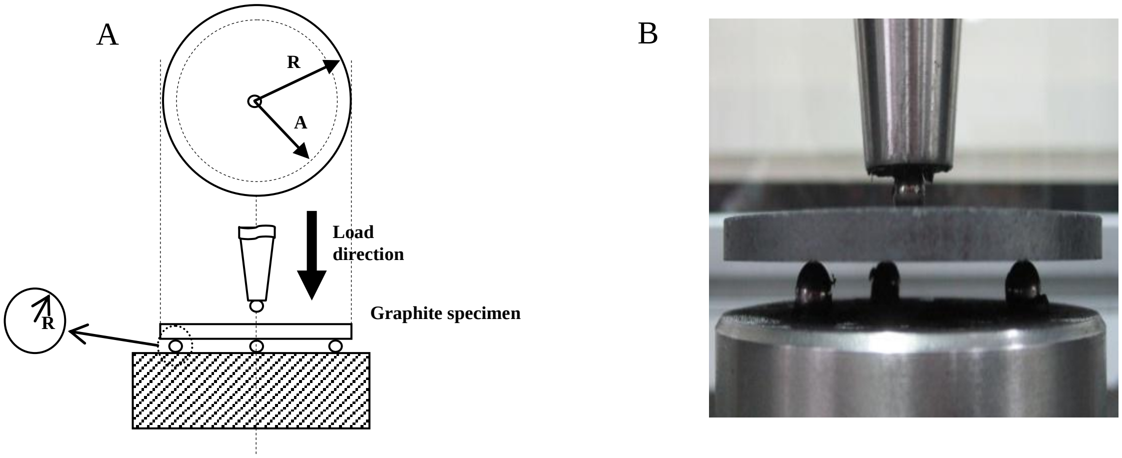

Fig. 1: (A) Schematic diagram of biaxial ball on 3 ball test apparatus with sample position and (B) Horizontal view of actual biaxial testing.

RESULT AND DISCUSSION

The advantage of the ball on the three balls strength test is that the area of the maximum tensile stress is located at the centre of the lower face of the plate. From observation, the entire fractured specimen's fracture is initiated in the tensile surface plane underneath the loading balls. The sample(s) break into 2 and 3 pieces (Fig 2). In all investigated cases, the fracture origin was at or close to the tensile's origin. Figure 3 (a) indicated that the surface faces the compression and tension site, whereas Figure 3b shows the course surface at the tensile site, compared to the compression site fracture surface of the fracture sample. In most cases, there are no fractures starting from the area around the loading ball; this early observation proves that the ball on three balls loading geometry is appropriate.

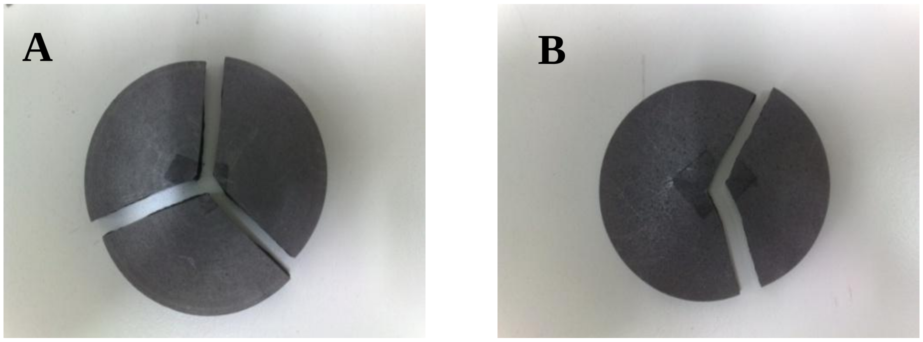

Fig. 2: Optical image shows the fracture pattern of EY9 graphite
(A) 3 pieces break and (B) 2 pieces break.

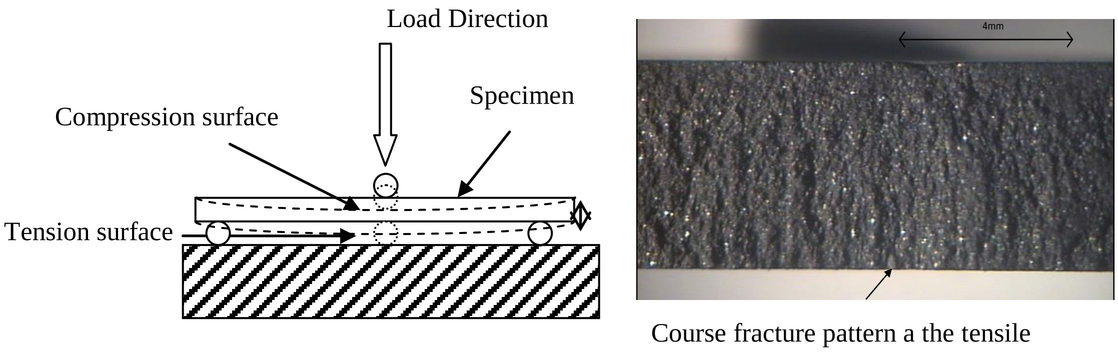

Fig. 3: (A) Schematic diagram demonstrates the tensile and the compression surface site.
(B) Optical image shows the texture pattern of fracture surface at tensile and compression site.

Figure 4 shows the histogram of the biaxial strength of EY9 graphite, calculated using the Godfrey, Ovri, Higgs, Pagniano, ASTM F394-78 and Roger solutions. Higgs, Ovri, Borger and ASTM-78 give the negative values of biaxial strength. These 4 solutions are considered invalid for EY9 graphite, and are thus excluded for the thickness study. However, it should also be noted that the ASTM have now withdrawn this standard. Godfrey shows the highest value, followed by Godfrey that includes Westegaard's approximation contact area, which was about 26.61 and 17.62 MPa, respectively, whereas the lowest biaxial strength of 13.5 MPa was obtained by Pagniano solution.

The biaxial strength of EY9 graphite, with different thickness of specimen is shown in Figure 5. Generally, Godfrey (WG) provide the higher value of biaxial strength for the all thicknesses, compared to Godfrey without the application of the approximation of the contact area, and Pagniano indicate the contrast of biaxial strength result with Godfrey after a 5 mm thickness. Godfrey (WG) shows the increase in strength when the sample thickness increases from 4 to 5 mm.

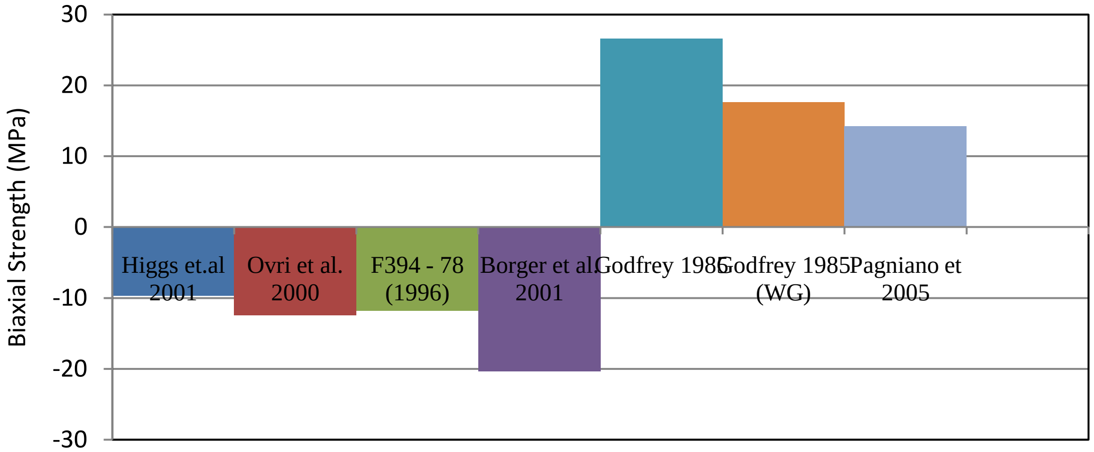

Fig. 4: Histogram of the biaxial strength obtained by numerous analytical solutions

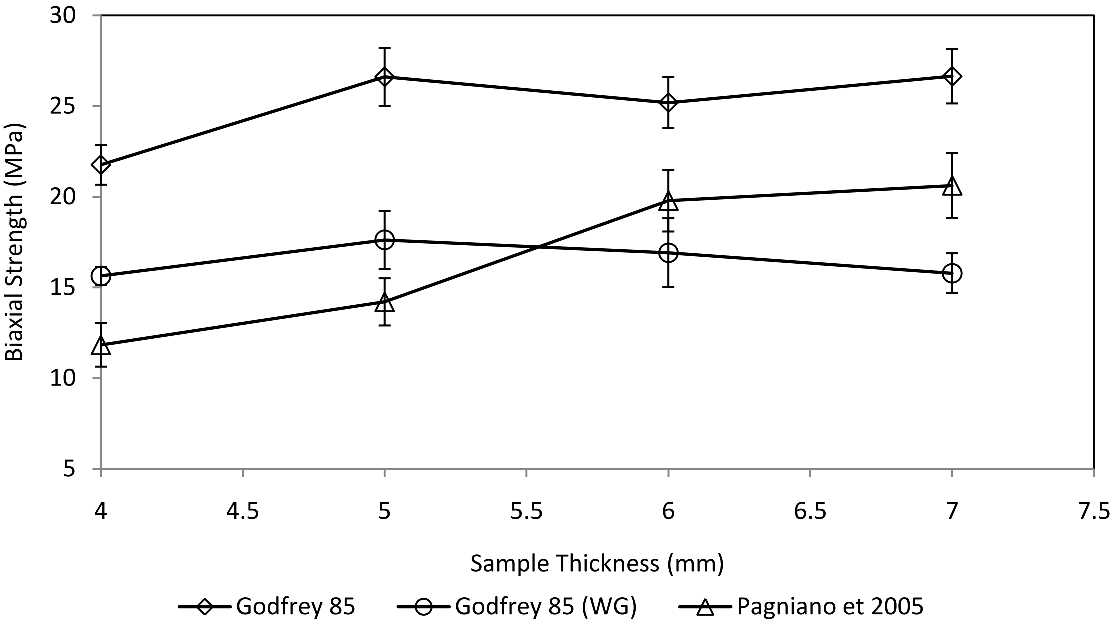

Fig. 5: Figure shows the effects of sample thickness to biaxial strength

The strength is constant with a slight increase when the sample's thickness increased from 5 to 7 mm. The trend shows almost the same pattern without using Westegaard approximation contact area in Godfrey solutions, but shows a constant strength for the 6 mm of specimen thickness. This observation is contrary to the statistical theory of fracture, which predicts lower stresses for larger sample. A number of factors are probably responsible for this observation, such as specimen's density, porosity, homogeneity and fracture mode as well. However, further work needs to be done in order to define the role of each factor, especially with regards to Pagniano solution, because it shows the decrease of biaxial strength after 5 mm thickness. Neighbour (2010) discovered that the drastic decrease in biaxial strength for nuclear grade graphite occurs when the thickness of the specimen was 8 mm. He also noted that a large amount of the compressive force is concentrated at the point of loading, compared to tensile surface, when higher volume specimens are used. This could be effect of the loading radius, when Pagniano suggested that the radius of uniform loading at the centre is equivalent to the radius of the loading ball

The biaxial strength of graphite tends to be lower than its uniaxial strength; it was about 80-85 % of the uniaxial strength (Brocklehurst et al. 1977). The data obtained from compression test shows the mean of the uniaxial strength is about 8.5 MPa, and suggest that the biaxial strength is actually higher than the uniaxial strength, contradicting general relations stated by Brocklehurst. Neighbour (2010) reported that the compression test on graphite, configured by Kennedy's suggestion to calculate uniaxial strength also discovered the uniaxial stress is lower than the biaxial strength, and this also contradicts Brocklerhurst works. It should be noted, however, that the characteristic of nuclear grade graphite is quasi–brittle, due to the presence of calcinations cracks, gas evolution pore, binder phase filler particle and fragmented filler particle, and this factor could very well influence the results. Figure 6 shows the uniaxial strength obtained by compression test.

The biaxial of strength of graphite tend to be lower than the uniaxial strength; it was about 80-85 % of the uniaxial strength (Brocklehurst et al. 1977). The data obtained from compression test show mean of uniaxial strength was about 8.5 MPa and suggest that the biaxial strength actually higher than uniaxial strength and it is contradict to general relations state by Brocklehurst. Neighbour (2010) reported that the compression test on graphite which configure by Kennedy (1993) suggestion to calculate uniaxial strength were also found the uniaxial stress is lower than biaxial strength and this also against the Brocklerhurst works. Should be note that the characteristic of nuclear grade graphite is quasi–brittle material due to the present of calcinations cracks, gas evolution pore, binder phase filler particle and fragmented filler particle that could influence the result. Figure 6 shows the uniaxial strength obtained by compression test.

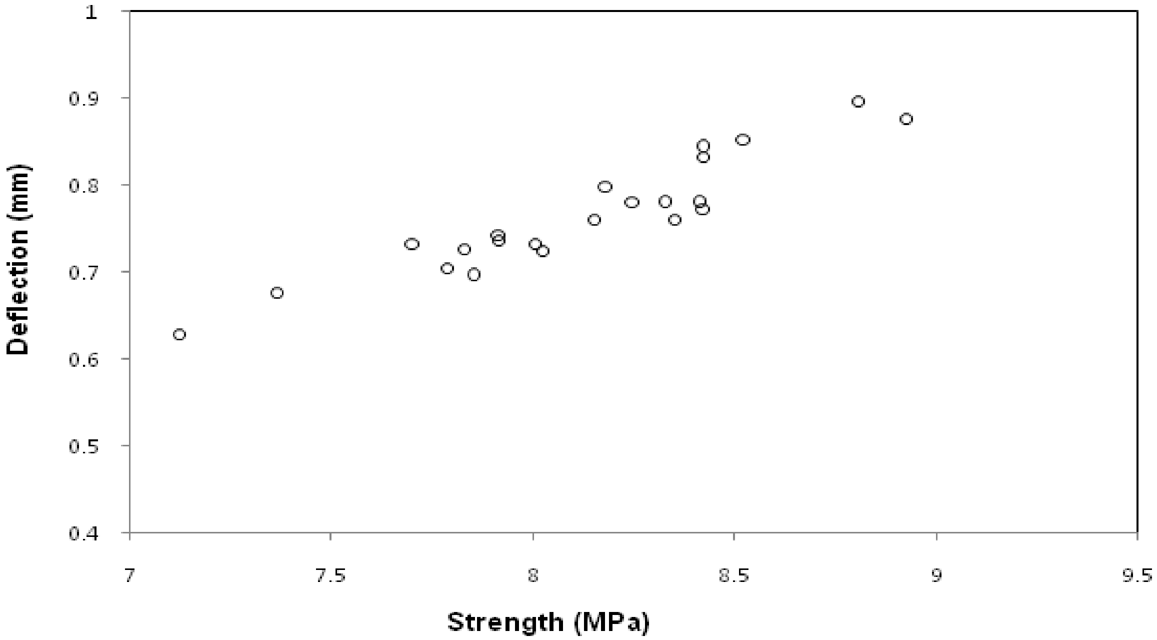

Fig. 6: Compression strength over the specimen’s deflection in uniaxial test

CONCLUSION

The biaxial strength obtained from various analytical solutions produced a range of strength of EY9 nuclear grade graphite. Some of the solution did follow the theory of brittle fracture and some did not. The relationship between biaxial strength and sample thickness was also indicated various trend depending on the analytical solutions. The parameters that were considered in ball on 3 ball test were contributed to different value of this biaxial strength. The biaxial strength values for EY9 grade graphite were also higher than uniaxial strength. The quasi-brittle behaviour of the nuclear graphite did influence much of the mechanical results, due to the presence of pores and other phases in the graphite. This would present a challenge in the future, especially when determining the significant mechanical test configuration in the future.

ACKNOWLEDGEMENT

The authors gratefully acknowledge the Department of Engineering, University of Hull which kindly provided the materials and facilities to complete the research of this paper.

REFERENCES

ASTM F394-78 (Reapproved 1996). Standard test method for biaxial flexure strength of ceramic substrates.

Albarhoum, M. (2011). Graphite Reflecting Characteristic and Shielding factors for Miniature Neutron Source Reactors, *Annals of Nuclear Energy* 38: 14-20.

Awaji, H., Sato, S., Kawamata, H., Kurumada, A., Oku T. (1987). Fracture criteria of reactor graphite under multiaxial stresses. *Nuclear Engineering and Design* 103: 291-300.

Brocklehurst, J. E. (1977). Fracture in Polycrystalline Graphite. *Chemistry and Physics of Carbon* 13: 145-279.

Börger, A., Supancic, P., Danzer, R., (2001). The ball on three balls test for strength testing of brittle discs: stress distribution in the disc. *Journal of the European Ceramics Society* 22*:* 1425-1436.

Doyoyo, M., Mohr, D. (2003). Microstructural response of aluminum honeycomb to combined out-of-plane loading. *Mechanics of Materials* 35: 865-876.

Fett, T., Ernst, E., Rizzi, G., Müller, R., Oberacker, R. (2006). A 3-balls-on-3-balls strength test for ceramic discs. *Journal of the European Ceramic Society*, (Corrected Proof, unspecified).

Frechou, C. And Degrpos, J.P. (2007). *Radiological Inventory of irradiated graphite samples. Journal of analytical and Nuclear Chemistry* 273: 677-681

Godfrey, D., J. (1985). Fabrication, formulation, mechanical properties, and oxidation of sintered Si_3N_4 ceramics using disc specimens. *Materials Science and Technology* 1: 510-515.

Higgs, R. J. E. D., Lucksanasombool , P., Higgs, W. A. J., Swain, M. V. (2000). Evaluating acrylic and glass- ionomer cement strength using the biaxial flexure test. *Biomaterials 22:* 1583-1590.

Imaninejad, M., Subhash, G., Loukus, A. (2004). Experimental and numerical investigation of free-bulge formation during hydroforming of aluminum extrusions. *Journal of Materials Processing Technology*, 147(2): 247-254.

Kennedy C.R. (1993). The Brittle-Ring Test for Graphite. *Carbon*, Vol. 31. No. 3 519-528

Kirstein, A.F., Woolley, R.M., (1967). Symmetrical bending of thin circular elastic plates on equally spaced point supports. *Journal of Research of the National Bureau of Standards 71C*: 1–10.

Neighbour, G. B. (2010). Biaxial Testing: Appropriate for Mechanical Characterization: Securing the Safe Performance of Graphite Reactor Cores, RSC Publishing.

Makuteswara, S. (2008). On estimating the fracture probability of nuclear graphite components, *Journal of Nuclear Materials* 381: 185–198.

Noel, N. N. & Robert L. B. (2010). Overview of statistical models of fracture for nonirradiated nuclear-graphite components, *Nuclear Engineering and Design* 240: 1-29.

Ovri, J. E. O. (2000). A parametric study of the biaxial strength test for brittle materials. *Materials Chemistry and Physics* 66: 1–5.

Pagniano, R.P., Seghi, R.R., Rosential, S.F., Wang, R., Katsube, N. (2005). The effect of a layer of resin luting agent on the biaxial flexure strength of two all-ceramic systems. *The Journal of Prosthetic Dentistry*: 459-466.

Seldin, E. J., (1966). Stress-Strain properties of Polycrystalline graphite in tension and compression at room temperature. *Carbon* 4: 177-191.