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DOE Deep Burn Program

The Characterization of Grade PCEA Recycle Graphite Pilot Scale Billets

Tim Burchell and Pete Pappano

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Summary

Here we report the physical properties of a series specimens machined from pilot scale (~ 152 mm diameter x ~305 mm length) grade PCEA recycle billets manufactured by GrafTech. The pilot scale billets were processed with increasing amounts of (unirradiated) graphite (from 20% to 100%) introduced to the formulation with the goal of determining if large fractions of recycle graphite have a deleterious effect on properties. The properties determined include Bulk Density, Electrical Resistivity, Elastic (Young's) Modulus, and Coefficient of Thermal Expansion. Although property variations were observed to be correlated with the recycle fraction, the magnitude of the variations was noted to be small.

1. Introduction

The High Temperature Reactor (HTR) variant of the Deep Burn reactor will be either a prismatic or pebble bed reactor. Independent of the type of reactor ultimately chosen, or the operating temperature of that reactor, large portions of the graphite moderator/structure will require relocation or removal as it approaches its useful life. This irradiated graphite, including graphite materials associated with fuel elements, represents a significant volume of waste, with associated economic, environmental, and long-term disposal implications. In order to mitigate the impact of this potentially enormous volume of activated waste, strategies for managing irradiated nuclear graphite for the HTR/Deep Burn program are underway. The most obvious strategy is to maximize the residence time of graphite in the core by shuffling into lower flux areas rather than removal and replacement of components. The second option is to directly recycle nuclear graphite by crushing and reforming the nuclear graphite after it has reached its useful lifetime.

The ultimate scope of commercial graphite recycle is challenging as it will require the large scale processing (crushing, pressing, chemical purification and machining) of large blocks of irradiated graphite. Such and endeavor will certainly require a significant infrastructure investment similar to that of fuel processing. Rather than addressing the breadth of such an effort, the current Deep Burn effort related to graphite recycle is limited to whether the reformation of graphite is technically viable. To address this, three areas of work have been undertaken:

1) Because working with non-irradiated graphite is significantly less difficult and expensive than working with irradiated materials, the viability of recycling non-irradiated graphite was carried out. GrafTech International (GTI) who is under contract to the Deep Burn program carried out a parametric study, with parameters of grinding, mixing, forming, and heat treatment. Properties were then measured by ORNL and the effectiveness of the produced product evaluated. The results of characterization performed at ORNL are reported here.

2) As the process of irradiation fundamentally alters the structure of the graphite crystal, and this structure is dependent on the annealing which will occur during the normal heating process associated with the graphite forming process, a series of experiments was carried out to determine the annealing kinetics of irradiated graphite in the annealing range appropriate to the recycle process.

3) A set of nuclear graphite materials, similar to that studied by GrafTech International, but that had been irradiated in the High Flux Isotope Reactor, were recycled. A process line for grinding, mixing, forming, and heat treating radioactive graphite was qualified and implemented. Recycled graphite was made with increasing percentages of ground (un-irradiated) graphite, and properties were measured for each specimen. The process was then carried out on the irradiated, radioactive graphite.

Nuclear grade graphite used as a moderator should preferably exhibit certain key properties in order to minimize structural damage during irradiation. A nuclear grade graphite generally has a small coke particle size (<1.6 mm), an isotropic coke feedstock, relatively high strength, and low impurity levels. The process of nuclear grade graphite manufacture is shown in the process flow diagram in Figure 1.

The starting raw materials for making graphite are petroleum coke or pitch coke and coal tar pitch. The coke and pitch are mixed together, usually at an elevated temperature, and then formed into a shape via extrusion, compression molding, isostatic pressing, or vibratory molding. The forming method affects the microstructure of the graphite, and to some extend its behavior under irradiation. Nuclear-grade graphite typically requires an isotropic microstructure, so isostatic molding is preferred. However, the costs associated with using this forming method to produce a graphite block large enough to be used in a reactor make iso-molding prohibitive (unless they offer improved lifetime). High quality, isotropic graphite have been made via extrusion and vibro-molding, so long as special attention was paid to coke properties (coke isotropy).

Following forming, the “green” graphite body is baked in order to carbonize the pitch binder. Nuclear-grade graphite requires a high density and strength so the baked artifact is normally re-impregnated with pitch in order to densify the piece. Pitch impregnation, or “PI,” can be repeated numerous times in order to increase density and strength. Nuclear graphite available today is generally impregnated three times.

The impregnated graphite is heat treated to >2500°C in order to graphitize the carbon structure. Following graphitization, the material is machined to size and then is ready for use. The process for making nuclear-grade graphite takes approximately 6-9 months.

The recycle graphite process, on an industrial scale, would be identical to the flow diagram shown in Figure 1, except that the coke filler would be replaced with particles of ground irradiated graphite.

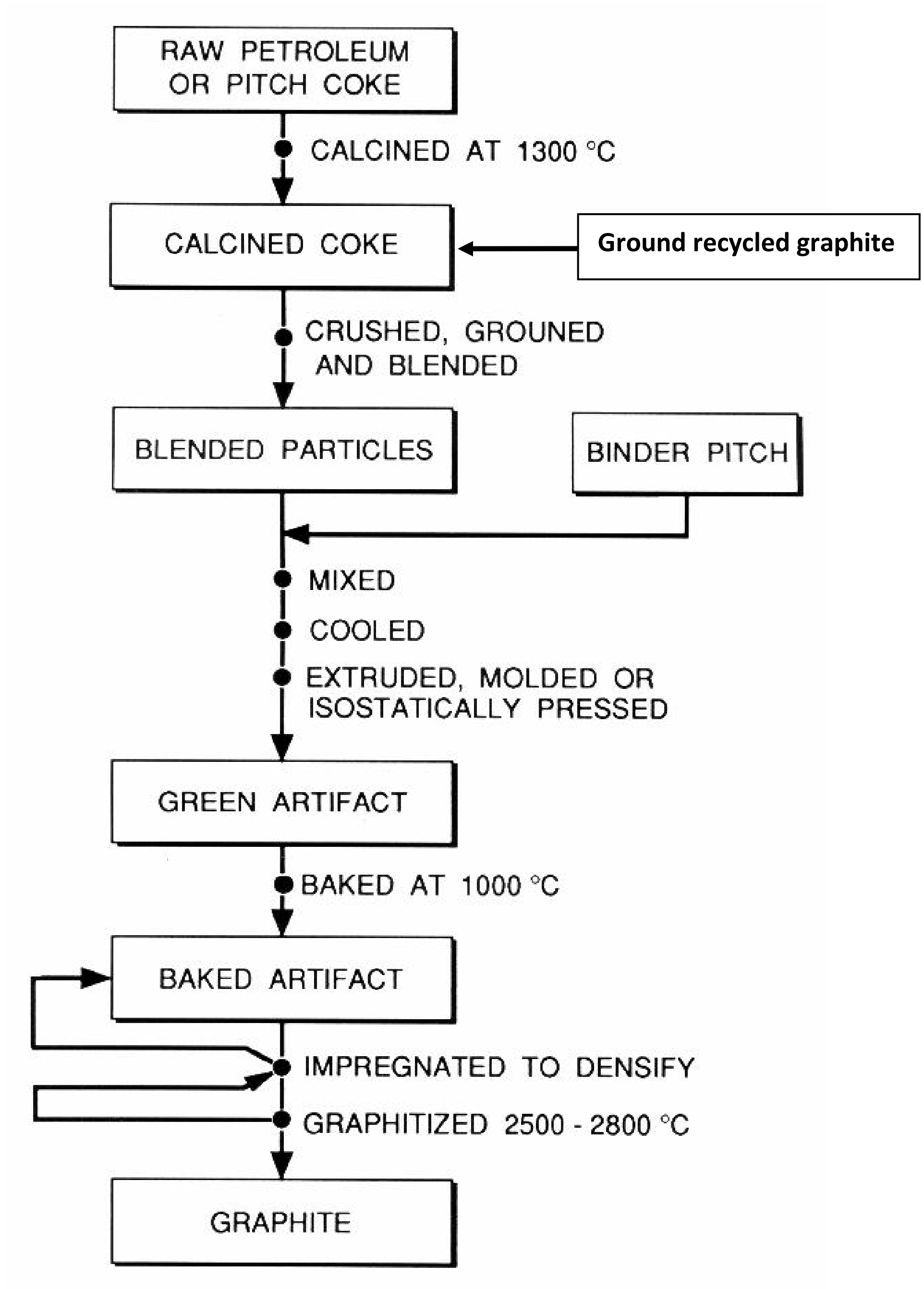

Figure 1. Process flow diagram for production of graphite

The pilot scale billets were processed with increasing amounts of (unirradiated) graphite (from 20% to 100%) introduced to the formulation with the goal of determining if large fractions of recycle graphite have deleterious effect on properties. Ultimately, selected specimens will provide irradiated raw stock for the study of irradiated graphite recycle.

2. Experimental

2.1. Materials

GrafTech International (GTI) was contracted to perform graphite recycle of their nuclear grade graphite, PCEA. PCEA is an extruded grade of graphite that is pitch impregnated three times.

A block of PCEA graphite was obtained and ground into a set particle size. The exact particle size and other process parameters were not disclosed, as they are considered proprietary to GTI. The ground PCEA graphite particles were designated as “RG” particles for tracking purposes. In total, 27 graphite billets were fabricated for the initial phase of this study. Nine (9) mix formulations were used and three (3) billets were made per mix formulation. Table 1 shows the percentage of RG that was used per mix formulation and the properties that were measured for each billet. Figure 2 shows an image of nine (9) billets, one from each mix formulation.

In addition to the “virgin” nature of the recycled graphite starting material, and the scale of the graphite products being formed, the other significant difference in the GTI work as compared the ORNL effort previously reported [1] is the fact that it is a 3 PI material. Thus, the GTI materials are appreciably denser due to the second and third impregnation.

Table 1 shows that the density of the recycle graphite (as reported by GTI) was relatively unaffected by the percentage of RG particles. The densities of all of the billets (except the second in Mix #1) are all over 1.8 g/cc, which is typical of nuclear graphite. The lowest density (1.789 g/cc) was actually found in the set of billets with the least amount of recycle material, which was 20%. The density of the final recycle graphite does not appear to be negatively impacted by having 100% recycle material. No virgin material, such as coke or synthetic graphite, appears necessary when making the recycled graphite from non-irradiated stock.

Table 1. Mass, volume, and densities for GTI recycle PCEA graphite

	Billet Identification No.	Wt. (gm)	Avg.Diam. (in)	Avg.Lgth. (in)	Density (g/cc)	Vol. (cc)
	092001-2	**10013.6**	5.845	12.448	**1.829**	5474
Mix 1 (20% RG as particles)	**092003-1**	**9048.6**	5.681	12.179	**1.789**	5058
Mix 1 (20% RG as particles)	**092003-2**	**9044.9**	5.721	11.909	**1.803**	5017
Mix 1 (20% RG as particles)	**092003-3**	**9269.3**	5.708	12.249	**1.805**	5137
Mix 1 (20% RG as particles)	**092003-4**	**9338.5**	5.727	12.209	**1.812**	5153
Mix 1 (20% RG as particles)	**092003-5**	**9356.8**	5.715	12.274	**1.814**	5159
Mix 2 (27.3% RG as particles)	**092004-2**	**9502.6**	5.794	12.190	**1.804**	5267
Mix 2 (27.3% RG as particles)	**092004-3**	**9467.7**	5.783	12.142	**1.812**	5226
Mix 2 (27.3% RG as particles)	**092004-4**	**9592.9**	5.780	12.294	**1.815**	5287
Mix 2 (27.3% RG as particles)	**092004-5**	**9548.3**	5.796	12.203	**1.810**	5276
Mix 3 (34.8% RG as particles)	**092005-3**	**10638.1**	5.827	13.491	**1.804**	5896
Mix 3 (34.8% RG as particles)	**092005-4**	**10498.8**	5.804	13.334	**1.816**	5781
Mix 3 (34.8% RG as particles)	**092005-5**	**9812.3**	5.807	12.467	**1.814**	5410
Mix 4 (42.4% RG as particles)	**092006-1**	**10720.7**	5.789	13.755	**1.807**	5933
Mix 4 (42.4% RG as particles)	**092006-2**	**10594.1**	5.828	13.241	**1.830**	5788
Mix 4 (42.4% RG as particles)	**092006-3**	**10411.3**	5.792	13.170	**1.831**	5685
Mix 4 (42.4% RG as particles)	**092006-4**	**10675.4**	5.801	13.407	**1.839**	5806
Mix 4 (42.4% RG as particles)	**092006-5**	**10431.8**	5.798	13.092	**1.842**	5664
Mix 5 (50% RG as particles)	**092007-1**	**9803.4**	5.803	12.339	**1.833**	5347
Mix 5 (50% RG as particles)	**092007-2**	**9800.3**	5.825	12.175	**1.843**	5317
Mix 5 (50% RG as particles)	**092007-3**	**9884.7**	5.814	12.325	**1.843**	5363
Mix 5 (50% RG as particles)	**092007-4**	**9900.1**	5.831	12.270	**1.844**	5369
Mix 5 (50% RG as particles)	**092007-5**	**9716.6**	5.775	12.371	**1.830**	5309
Mix 6 (42.4% RG part and flour)	**092008-2**	**9805.5**	5.846	12.271	**1.817**	5397
Mix 6 (42.4% RG part and flour)	**092008-3**	**9634.3**	5.818	12.172	**1.817**	5302
Mix 6 (42.4% RG part and flour)	**092008-4**	**9903.2**	5.870	12.310	**1.814**	5460
Mix 6 (42.4% RG part and flour)	**092008-5**	**9844.9**	5.828	12.373	**1.820**	5408
Mix 7 (65%RG part and flour)	**092009-1**	**9592.4**	5.759	12.295	**1.828**	5248
Mix 7 (65%RG part and flour)	**092009-2**	**9633.1**	5.776	12.176	**1.843**	5228
Mix 7 (65%RG part and flour)	**092009-4**	**9586.0**	5.747	12.226	**1.844**	5198
Mix 7 (65%RG part and flour)	**092009-5**	**9529.1**	5.727	12.226	**1.846**	5161
Mix 8 (72.7% RG part and flour)	**092010-1**	**9735.9**	5.813	12.222	**1.832**	5316
Mix 8 (72.7% RG part and flour)	**092010-3**	**10111.6**	5.896	12.363	**1.828**	5530
Mix 8 (72.7% RG part and flour)	**092010-4**	**10047.4**	5.874	12.367	**1.830**	5491
Mix 8 (72.7% RG part and flour)	**092010-5**	**9734.0**	5.855	12.060	**1.829**	5321
Mix 9 (100% RG part and flour)	**092011-1**	**9902.2**	5.832	12.397	**1.825**	5427
Mix 9 (100% RG part and flour)	**092011-2**	**10356.4**	5.948	12.507	**1.819**	5695
Mix 9 (100% RG part and flour)	**092011-3**	**10356.1**	5.986	12.349	**1.818**	5695
Mix 9 (100% RG part and flour)	**092011-4**	**10101.7**	5.902	12.380	**1.820**	5551
Mix 9 (100% RG part and flour)	**092011-5**	**10257.7**	5.930	12.461	**1.819**	5638

Figure 2 Image of recycle PCEA billets formed by GTI under Subcontract to the Deep Burn program; one is shown from each mix formulation

The details of the billets shipped to Oak Ridge are given in Table 1. The recycle graphite fraction was introduced into the green mix in two forms, particles or a particle/flour mix. The billets were samples to provide 15 specimens (6x6x50 mm) in the with-grain (WG) and 15 specimens in the against-grain (AG) orientation. The density values reported in Table 2 are the means of the 15 specimens in each of the specimen orientations. Also shown are the billet densities reported by GrafTech.

2.2. Experimental Methods

2.2.1. Bulk Density

The bulk density was determined by mensuration using calibrated measuring equipment in accordance with ASTM Standard Test Method C559 [2].

2.2.2. Elastic Modulus

The elastic modulus was determined using the fundamental frequency method in accordance with ASTM method C 747 - 93 (Reapproved 2005) [3] using a GrindoSonic Mk5, S/N 0620843. Figure X shows the experimental apparatus. The elastic modulus, E (Pa) is given by:

$$E = CMf^{2}$$

where C is a dimensionless constant that depends upon the specimen size and geometry and Poisson's ratio, M is the specimens mass (kg), and *f* is the fundamental frequency of flexural mode vibration (Hz).

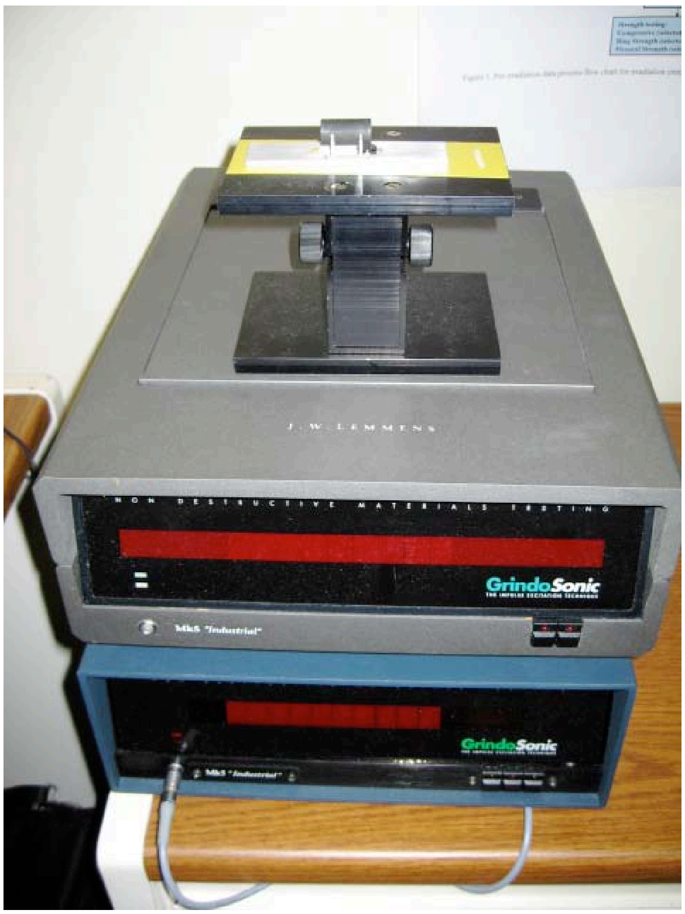

Figure 3. GindoSonic Mk5 fundamental frequency modulus system

2.2.3. Electrical Resistivity

Electrical resistivity was determined in accordance with ASTM Standard C 611 – 98 (reapproved 2005), "Standard Test Method for Electrical Resistivity of Manufactured Carbon and Graphite Articles at Room Temperature" [4]. The test setup employed a Keithley 2400 Source Meter (current supply), and a Keithley 2182 Nanovoltmeter. A pair of probes (knife edges) for applying the electrical current through the specimen and another pair of probes for measuring the potential was mounted in an insulating, Plexiglas block. The magnitude of the current was low, and the time that the current was allowed to flow through the specimen was kept short such that the temperature rise in the specimen was negligible, and consequently the resistance of the specimen was not changed. Multiple resistivity measurements were made on each specimen to determine an average value.

The electrical resistivity, ρ, is calculated from:

$$\rho = \frac{R \cdot A}{L} \quad (\Omega.\text{m}) \tag{1}$$

where: A = cross-sectional area, m^2

L = gauge length, m

R = resistance, Ω

and is given by R = (V_x/I) where V_x is the mean voltage (V) and I is the current (A).

2.2.4. Coefficient of Thermal Expansion (CTE)

CTE testing was performed using a dilatometer supplied by Anter Inc in accordance with ASTM E 228 [5]. The samples were heated from 100 to 900°C at a rate of 5°C/minute with a 30 minutes dwell time at each increment of 100°C after which the expansion was recorded. Periodic calibration checks were performed by running a certified Crystallox standard also supplied by Anter Inc. A typical calibration run result is shown in Figure 4.

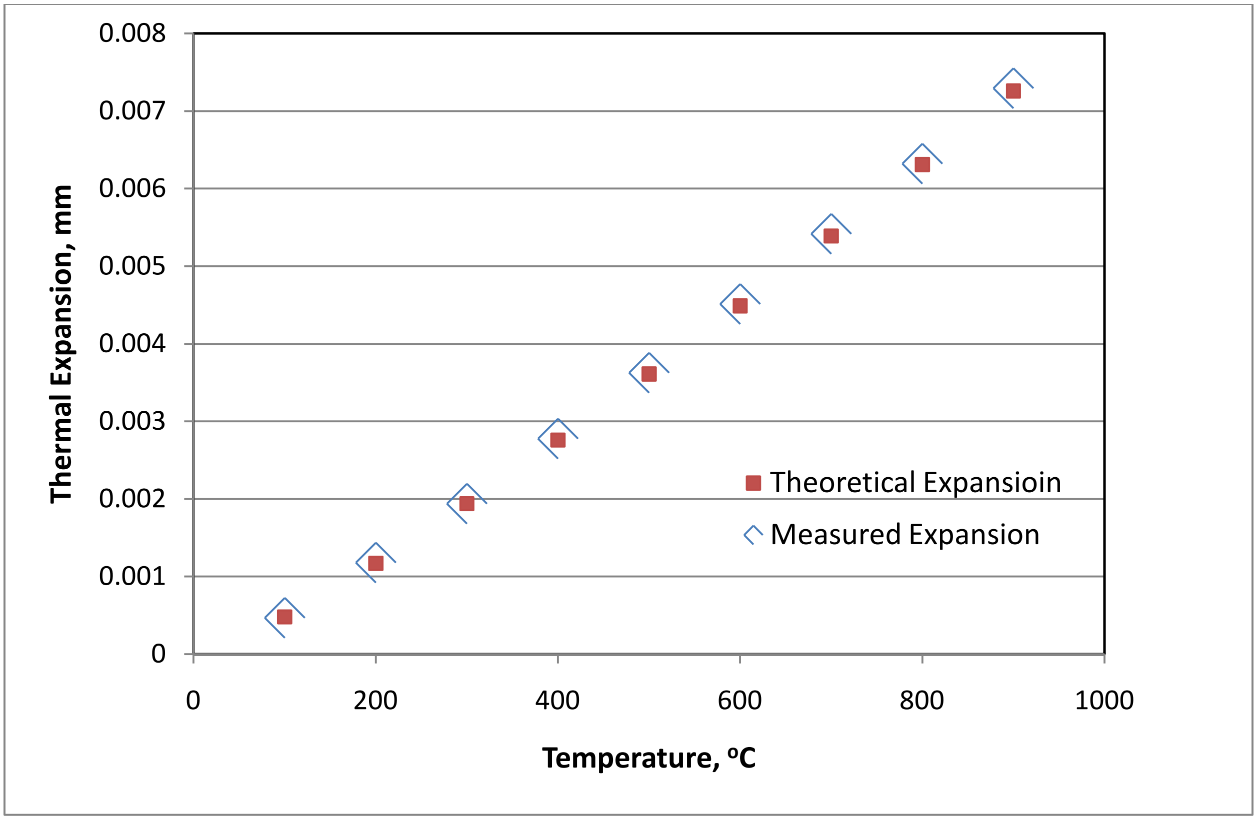

Figure 4. Dilatometer calibration run performed August 18th 2010 using a certified Crystallox standard.

3. Results and Discussion

3.1. Bulk Density

The details of the billets shipped to Oak Ridge are given in Table 2. The recycle graphite fraction was introduced into the green mix in two forms, particles or a particle/flour mix. The billets were sampled to provide 15 specimens (6x6x50 mm) in the with-grain (WG) and 15 specimens in the against-grain (AG) orientation. The density values reported in Table 1 are the means of the 15 specimens in each of the specimen orientations. Also shown are the whole billet densities reported by GrafTech.

Table 2. GrafTech pilot scale billets being characterized at ORNL

Billet Code	Specimen Orientation	Number of specimens	Recycle Fraction %	Mean Density g/cc	GrafTech Billet Density g/cc	Recycle Fraction Condition
03-4	AG	15	20	1.828	1.812	particles
03-4	WG	15	20	1.829	1.812	particles
04-3	AG	15	27.3	1.813	1.812	particles
04-3	WG	15	27.3	1.822	1.812	particles
05-4	AG	15	34.8	1.822	1.816	particles
05-4	WG	15	34.8	1.827	1.816	particles
06-3	AG	15	42.4	1.831	1.831	particles
06-3	WG	15	42.4	1.834	1.831	particles
07-3	AG	15	50	1.838	1.843	particles
07-3	WG	15	50	1.846	1.843	particles
08-3	AG	15	42.5	1.816	1.817	particles &
08-3	WG	15	42.5	1.812	1.817	particles &
09-2	AG	15	65	1.846	1.843	particles &
09-2	WG	15	65	1.839	1.843	particles &
10-5	AG	15	72.7	1.819	1.829	particles &
10-5	WG	15	72.7	1.833	1.829	particles &
11-4	AG	15	100	1.815	1.82	particles &
11-4	WG	15	100	1.818	1.82	particles &

The density in the WG orientated specimens appear to be consistently greater than in the AG (Figs. 4 and 5), and reflects the preferred orientation of the extruded billet texture. The range of densities is similar for both graphite's (flour/particles and particles) although the graphite with flour/particles in its mix appears to exhibit a maximum density at ~60% recycle fraction. A maximum is not seen in the density of the graphite with particles only, rather, the density increases when the recycle fraction increases above ~25% reaching ~1.84 g/cm^3 at 50% recycle fraction. It should be noted that the overall variation in density of the graphite billets were relatively small, ranging from 1.812 to1.846 g/cm^3, a variation of less than 2%, in agreement with the GTI whole billet data.

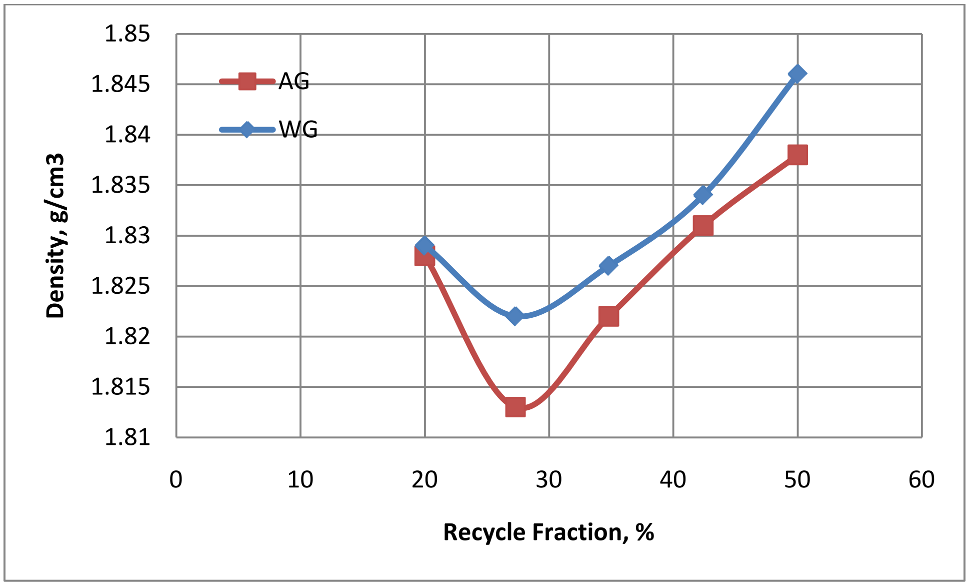

Figure 5. The variation of bulk density with recycle fraction for the GrafTech billets made with recycle graphite as particles in the green mix

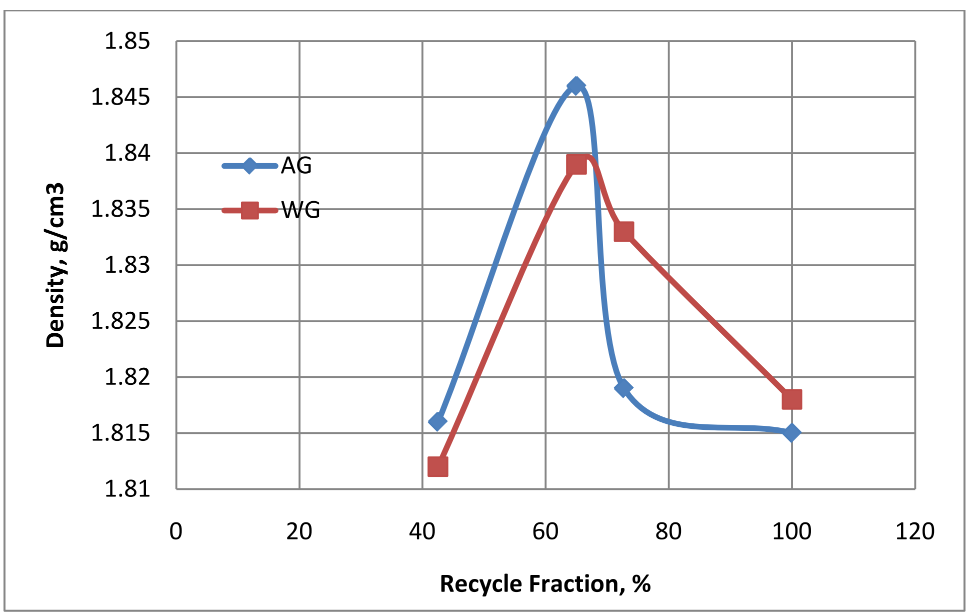

Figure 6. The variation of bulk density with recycle fraction for the GrafTech billets made with recycle graphite as a particle/flour mix in the green mix

3.2. Elastic Modulus

Figures 7 and 8 report the mean Dynamic Young's Modulus values obtained from the specimen sets. Again, property anisotropy is observed, the WG value being consistently greater than the AG value, although the observed differences were generally less than 10% (typical for near-isotropic graphite). There appears to be no systematic variation of modulus with recycle fraction.

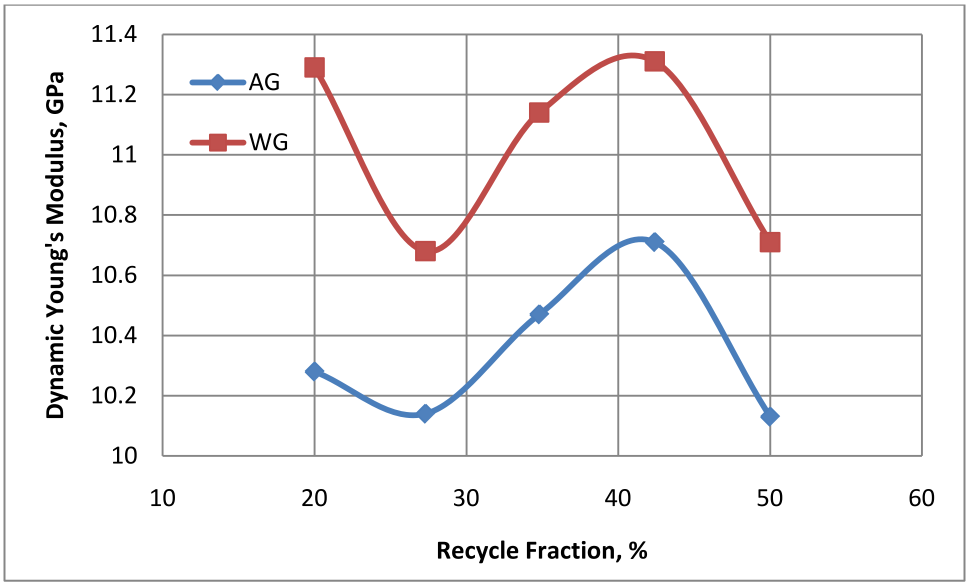

Figure 7. Variation of Mean dynamic Young's modulus with recycle fraction for the GrafTech billets made with recycle graphite as particles in the green mix

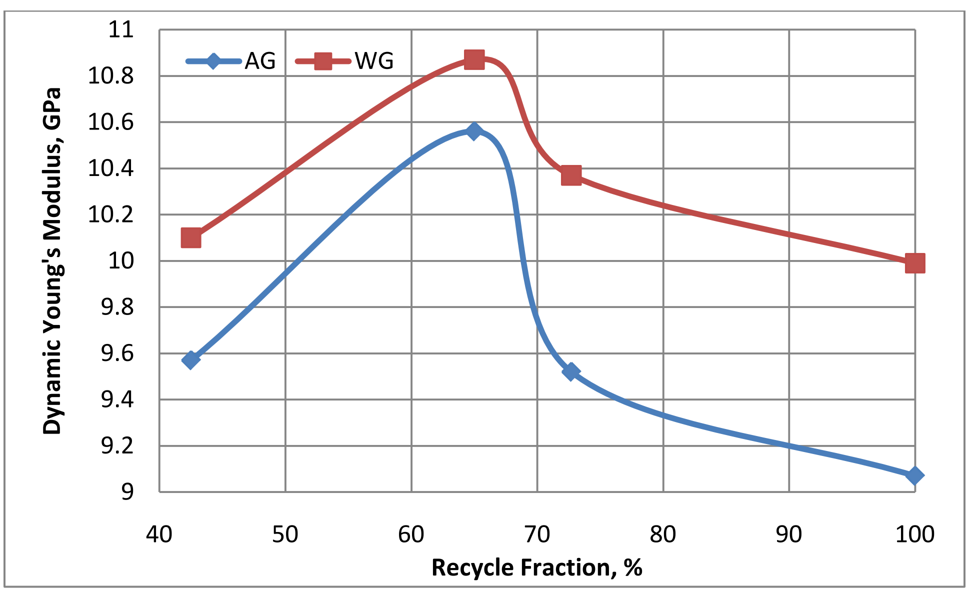

Figure 8. Variation of Mean dynamic Young's modulus with recycle fraction for the GrafTech billets made with recycle graphite as a particles/flour mix in the green mix

3.3. Electrical Resistivity

The measured values of electrical resistivity for each of the specimens, and the mean and standard deviation of the billet/orientation group, are reported in Table 3 (particles only) and Table 4 (particles and flour). The data are summarized in Figs. 9 and 10.

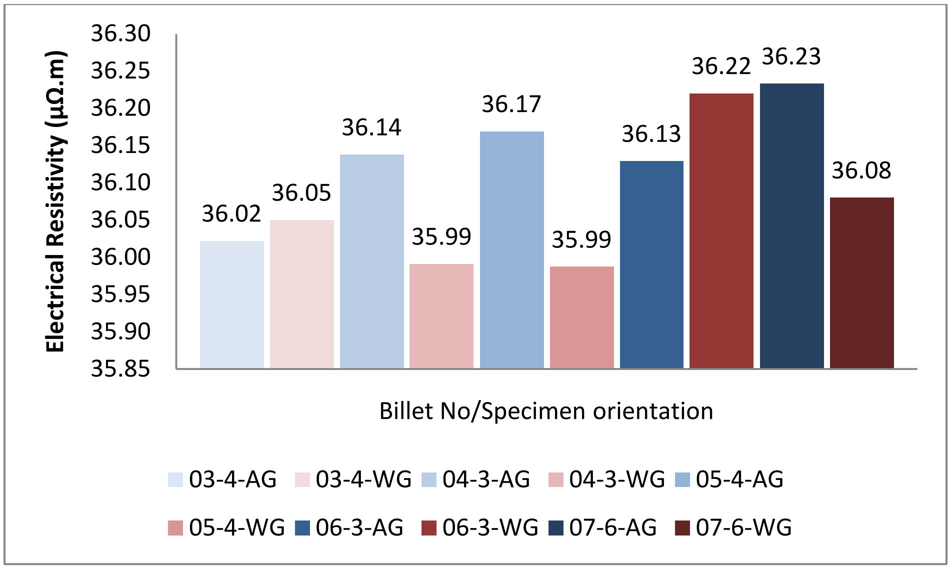

Figure 9. Summary of mean Electrical Resistivity data organized by % of recycle particles and specimen orientation

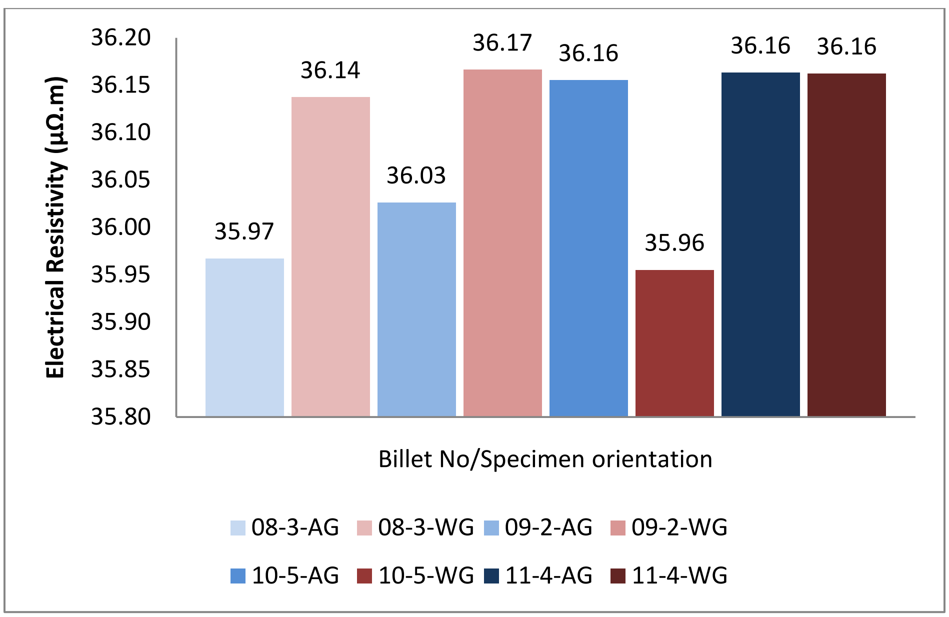

Figure 10. Summary of mean Electrical Resistivity data organized by % of recycle particles/flour and specimen orientation

Table 3. Electrical resistivity for each specimen, and the mean and standard deviation of the billet/orientation group (particles only)

Deep Burrn PCEA Recycle Graphite(Particles only)																			
Billet 03-4-AG		**Billet 03-4-WG**		**Billet 04-3-AG**		**Billet 04-3-WG**		**Billet 05-4-AG**		**Billet 05-4-WG**		**Billet 06-3-AG**		**Billet 06-3-WG**		**Billet 07-6-AG**		**Billet 07-3-WG**	
Specimen Number	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)	**Specimen Number**	Resistivity (μΩ. m)
03-4-AG-1	35.96	03-4-WG-1	35.99	04-3-AG-1	36.19	04-3-WG-1	35.97	05-4-AG-1	36.13	05-4-WG-1	36.09	06-3-AG-1	36.08	06-3-WG-1	36.09	07-3-AG-1	36.27	07-3-WG-1	36.18
03-4-AG-2	36.15	03-4-WG-2	36.05	04-3-AG-2	36.17	04-3-WG-2	35.94	05-4-AG-2	36.19	05-4-WG-2	36.00	06-3-AG-2	36.13	06-3-WG-2	36.32	07-3-AG-2	36.30	07-3-WG-2	35.96
03-4-AG-3	36.00	03-4-WG-3	35.99	04-3-AG-3	36.06	04-3-WG-3	36.06	05-4-AG-3	36.10	05-4-WG-3	36.00	06-3-AG-3	36.15	06-3-WG-3	36.10	07-3-AG-3	36.20	07-3-WG-3	36.01
03-4-AG-4	36.03	03-4-WG-4	36.08	04-3-AG-4	36.12	04-3-WG-4	35.96	05-4-AG-4	36.21	05-4-WG-4	35.97	06-3-AG-4	36.12	06-3-WG-4	36.29	07-3-AG-4	36.27	07-3-WG-4	36.17
03-4-AG-5	36.09	03-4-WG-5	36.17	04-3-AG-5	36.09	04-3-WG-5	36.05	05-4-AG-5	36.27	05-4-WG-5	35.97	06-3-AG-5	36.16	06-3-WG-5	36.25	07-3-AG-5	36.26	07-3-WG-5	36.12
03-4-AG-6	36.06	03-4-WG-6	36.08	04-3-AG-6	36.07	04-3-WG-6	35.88	05-4-AG-6	36.11	05-4-WG-6	36.05	06-3-AG-6	36.26	06-3-WG-6	36.21	07-3-AG-6	36.34	07-3-WG-6	36.14
03-4-AG-7	36.00	03-4-WG-7	35.97	04-3-AG-7	36.06	04-3-WG-7	36.06	05-4-AG-7	36.06	05-4-WG-7	35.94	06-3-AG-7	36.07	06-3-WG-7	36.22	07-3-AG-7	36.30	07-3-WG-7	36.10
03-4-AG-8	35.97	03-4-WG-8	36.11	04-3-AG-8	36.22	04-3-WG-8	35.82	05-4-AG-8	36.22	05-4-WG-8	35.94	06-3-AG-8	36.33	06-3-WG-8	36.23	07-3-AG-8	36.21	07-3-WG-8	36.14
03-4-AG-9	36.02	03-4-WG-9	35.94	04-3-AG-9	36.14	04-3-WG-9	36.00	05-4-AG-9	36.13	05-4-WG-9	36.08	06-3-AG-9	36.11	06-3-WG-9	36.29	07-3-AG-9	36.11	07-3-WG-9	36.09
03-4-AG-10	36.03	03-4-WG-10	36.03	04-3-AG-10	36.24	04-3-WG-10	36.12	05-4-AG-10	36.08	05-4-WG-10	36.00	06-3-AG-10	36.11	06-3-WG-10	36.28	07-3-AG-10	36.23	07-3-WG-10	36.04
03-4-AG-11	35.97	03-4-WG-11	36.05	04-3-AG-11	36.14	04-3-WG-11	35.98	05-4-AG-11	36.16	05-4-WG-11	36.03	06-3-AG-11	36.08	06-3-WG-11	36.23	07-3-AG-11	36.23	07-3-WG-11	36.01
03-4-AG-12	36.09	03-4-WG-12	36.00	04-3-AG-12	36.20	04-3-WG-12	36.04	05-4-AG-12	36.19	05-4-WG-12	35.97	06-3-AG-12	36.05	06-3-WG-12	36.11	07-3-AG-12	36.11	07-3-WG-12	36.18
03-4-AG-13	35.96	03-4-WG-13	36.17	04-3-AG-13	36.16	04-3-WG-13	36.03	05-4-AG-13	36.23	05-4-WG-13	35.90	06-3-AG-13	36.05	06-3-WG-13	36.29	07-3-AG-13	36.35	07-3-WG-13	35.97
03-4-AG-14	35.99	03-4-WG-14	36.08	04-3-AG-14	36.14	04-3-WG-14	36.00	05-4-AG-14	36.24	05-4-WG-14	35.94	06-3-AG-14	36.13	06-3-WG-14	36.16	07-3-AG-14	36.20	07-3-WG-14	36.02
03-4-AG-15	36.03	03-4-WG-15	36.09	04-3-AG-15	36.05	04-3-WG-15	35.96	05-4-AG-15	36.22	05-4-WG-15	35.94	06-3-AG-15	36.12	06-3-WG-15	36.22	07-3-AG-15	36.11	07-3-WG-15	36.08
Mean	36.02	Mean	36.05	Mean	36.14	Mean	35.99	Mean	36.17	Mean	35.99	Mean	36.13	Mean	36.22	Mean	36.23	Mean	36.08
St. Dev.	0.056	St. Dev.	0.067	St. Dev.	0.061	St. Dev.	0.076	St. Dev.	0.065	St. Dev.	0.055	St. Dev.	0.076	St. Dev.	0.074	St. Dev.	0.078	St. Dev.	0.074

Table 4. Electrical resistivity for each specimen, and the mean and standard deviation of the billet/orientation group (particles & flour)

Deep Burrn PCEA Recycle Graphite (Particles & Flour)															
Billet 08-3-AG		**Billet 08-3-WG**		**Billet 09-2-AG**		**Billet 09-2-WG**		**Billet 10-5-AG**		**Billet 10-5-WG**		**Billet 11-4-AG**		**Billet 11-4-WG**	
Specimen Number	Resistivity (μΩ.m)	**Specimen Number**	Resistivity (μΩ.m)	**Specimen Number**	Resistivity (μΩ.m)	**Specimen Number**	Resistivity (μΩ.m)	**Specimen Number**	Resistivity (μΩ.m)	**Specimen Number**	Resistivity (μΩ.m)	**Specimen Number**	Resistivity (μΩ.m)	**Specimen Number**	Resistivity (μΩ.m)
08-3-AG-1	35.84	08-3-WG-1	36.22	09-2-AG-1	36.03	09-2-WG-1	36.16	10-5-AG-1	36.10	10-5-WG-1	35.99	11-4-AG-1	36.21	11-4-WG-1	36.12
08-3-AG-2	36.00	08-3-WG-2	36.21	09-2-AG-2	36.06	09-2-WG-2	36.15	10-5-AG-2	36.19	10-5-WG-2	35.85	11-4-AG-2	36.30	11-4-WG-2	36.12
08-3-AG-3	35.93	08-3-WG-3	36.13	09-2-AG-3	36.08	09-2-WG-3	36.15	10-5-AG-3	36.27	10-5-WG-3	35.98	11-4-AG-3	36.05	11-4-WG-3	36.26
08-3-AG-4	35.90	08-3-WG-4	36.14	09-2-AG-4	36.17	09-2-WG-4	36.20	10-5-AG-4	36.03	10-5-WG-4	35.87	11-4-AG-4	36.02	11-4-WG-4	36.27
08-3-AG-5	36.09	08-3-WG-5	36.14	09-2-AG-5	36.08	09-2-WG-5	36.08	10-5-AG-5	36.22	10-5-WG-5	35.94	11-4-AG-5	36.00	11-4-WG-5	36.30
08-3-AG-6	35.99	08-3-WG-6	36.14	09-2-AG-6	35.87	09-2-WG-6	36.05	10-5-AG-6	36.20	10-5-WG-6	35.91	11-4-AG-6	36.02	11-4-WG-6	36.15
08-3-AG-7	36.08	08-3-WG-7	36.17	09-2-AG-7	35.97	09-2-WG-7	36.15	10-5-AG-7	36.08	10-5-WG-7	35.90	11-4-AG-7	36.20	11-4-WG-7	36.12
08-3-AG-8	35.97	08-3-WG-8	36.19	09-2-AG-8	36.04	09-2-WG-8	36.21	10-5-AG-8	36.07	10-5-WG-8	36.01	11-4-AG-8	36.25	11-4-WG-8	36.17
08-3-AG-9	35.87	08-3-WG-9	36.14	09-2-AG-9	36.06	09-2-WG-9	36.23	10-5-AG-9	36.20	10-5-WG-9	35.90	11-4-AG-9	36.25	11-4-WG-9	36.14
08-3-AG-10	35.89	08-3-WG-10	36.07	09-2-AG-10	36.01	09-2-WG-10	36.26	10-5-AG-10	36.25	10-5-WG-10	36.04	11-4-AG-10	36.21	11-4-WG-10	36.08
08-3-AG-11	36.03	08-3-WG-11	36.06	09-2-AG-11	36.05	09-2-WG-11	36.10	10-5-AG-11	36.07	10-5-WG-11	35.97	11-4-AG-11	36.05	11-4-WG-11	36.16
08-3-AG-12	35.92	08-3-WG-12	36.12	09-2-AG-12	36.01	09-2-WG-12	36.05	10-5-AG-12	36.16	10-5-WG-12	36.00	11-4-AG-12	36.19	11-4-WG-12	36.09
08-3-AG-13	35.91	08-3-WG-13	36.04	09-2-AG-13	35.98	09-2-WG-13	36.19	10-5-AG-13	36.21	10-5-WG-13	35.94	11-4-AG-13	36.25	11-4-WG-13	36.04
08-3-AG-14	36.07	08-3-WG-14	36.19	09-2-AG-14	36.00	09-2-WG-14	36.23	10-5-AG-14	36.15	10-5-WG-14	36.03	11-4-AG-14	36.26	11-4-WG-14	36.26
08-3-AG-15	36.03	08-3-WG-15	36.09	09-2-AG-15	35.98	09-2-WG-15	36.27	10-5-AG-15	36.13	10-5-WG-15	36.00	11-4-AG-15	36.18	11-4-WG-15	36.16
Mean	35.97	Mean	36.14	Mean	36.03	Mean	36.17	Mean	36.16	Mean	35.96	Mean	36.16	Mean	36.16
St. Dev.	0.081	St. Dev.	0.055	St. Dev.	0.066	St. Dev.	0.071	St. Dev.	0.072	St. Dev.	0.061	St. Dev.	0.102	St. Dev.	0.078

There appears to be no systematic variation of electrical resistivity with recycle fraction. The observed variations were small, being less than ±1 standard deviation. Typically for an extruded product the WG resistivity is less that the AG resistivity. This was not uniformly observed in our data sets. However, grade PCEA is an isotropic grade and thus the variation with grain orientation would be expected to be small and may be less than the experimental error (standard deviation).

3.4. Coefficient of Thermal Expansion

The thermal expansion behavior and average CTE for specimens tested to date are shown in Figs. 11-30. Only a limited number of specimens have been tested to date because of the duration of the test cycle (~18 hours) and limited availability of the dilatometer. Additional funding will be required to complete a sufficient number of CTE measurements on each billet/orientation.

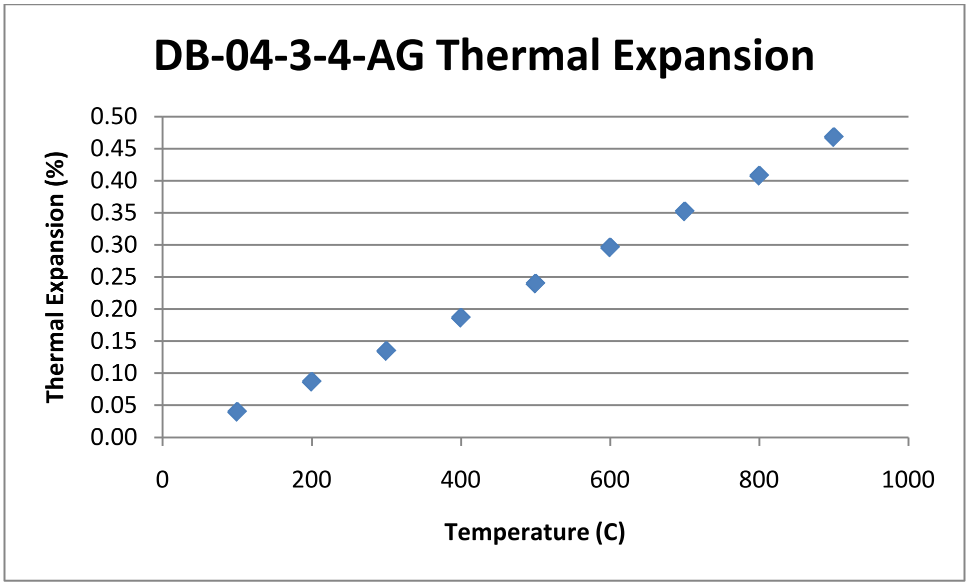

Figure 11. Thermal expansion behavior of PCEA recycle graphite specimen 04-3-4 (AG) with 27.3% of recycle particles

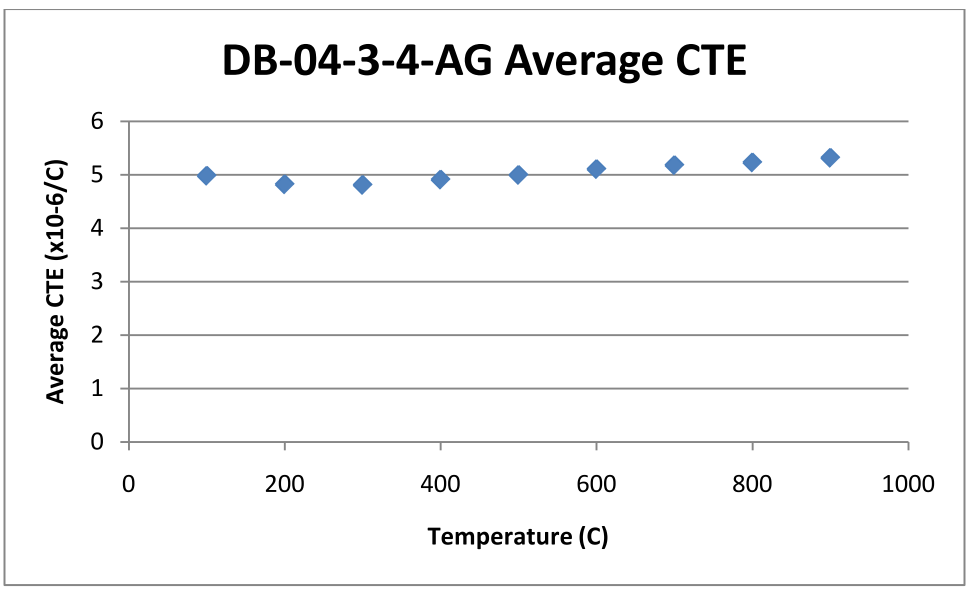

Figure 12. Average CTE of PCEA recycle graphite specimen 04-3-4 (AG) with 27.3% of recycle particles

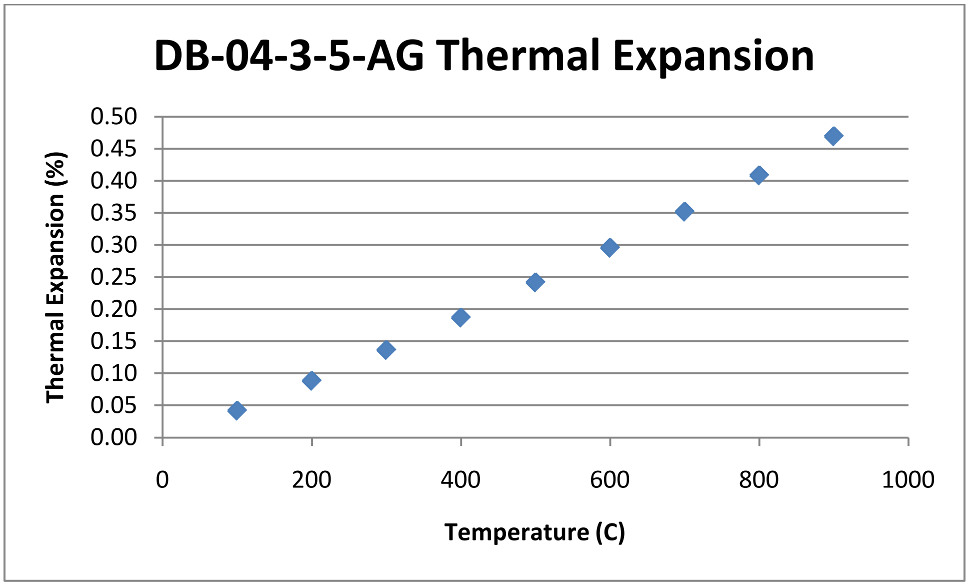

Figure 13. Thermal expansion behavior of PCEA recycle graphite specimen 04-3-5 (AG) with 27.3% of recycle particles

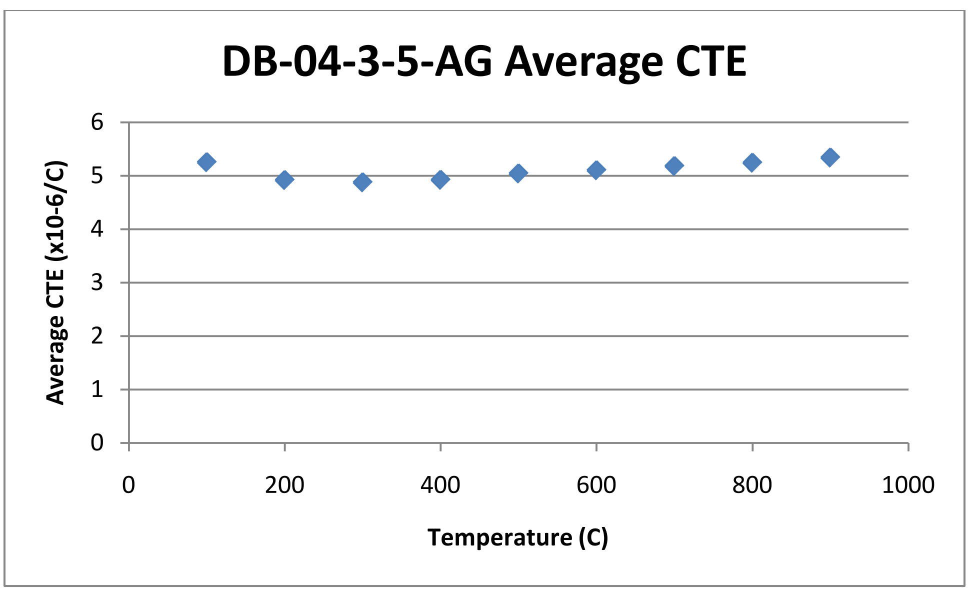

Figure 14. Average CTE of PCEA recycle graphite specimen 04-3-5 (AG) with 27.3% of recycle particles

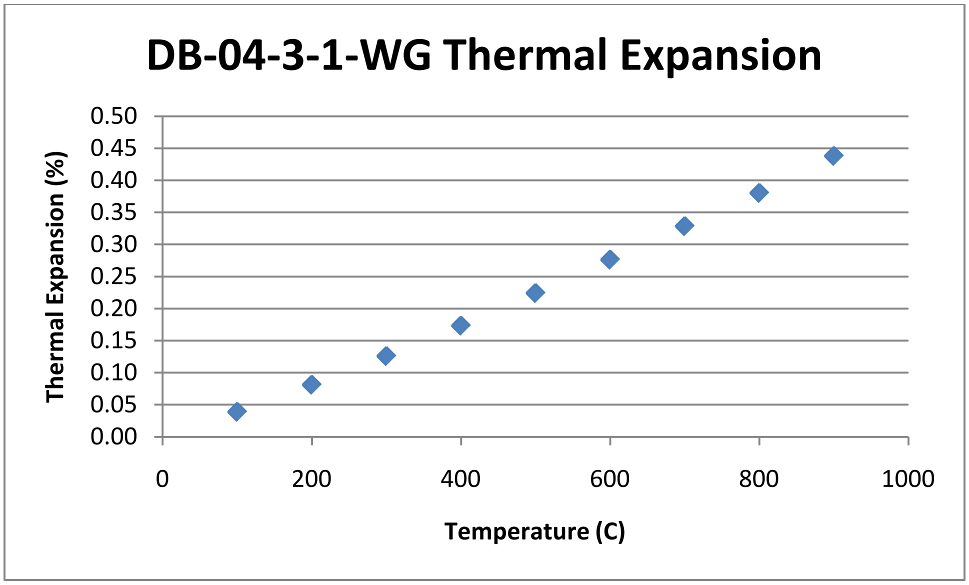

Figure 15. Thermal expansion behavior of PCEA recycle graphite specimen 04-3-1 (WG) with 27.3% of recycle particles

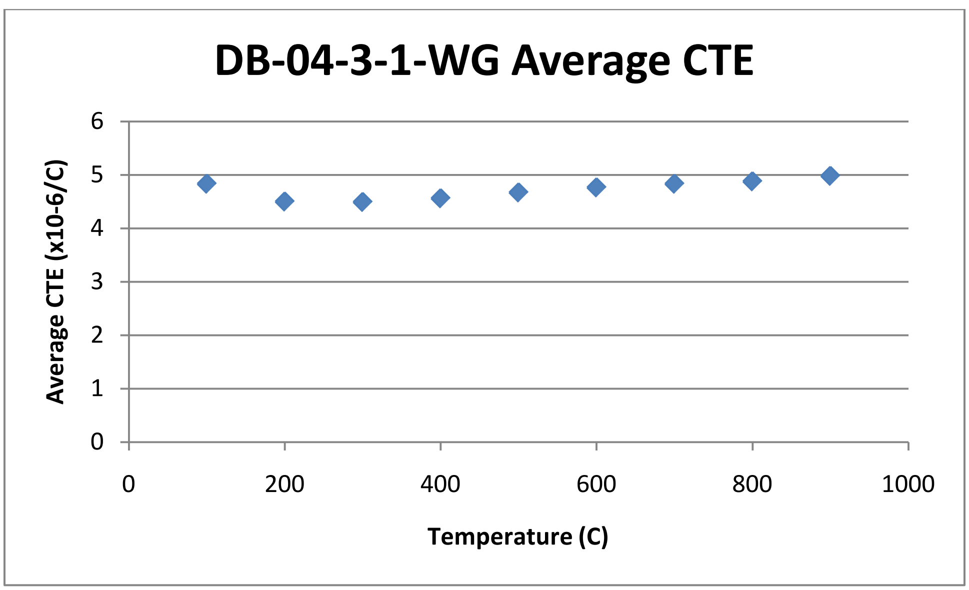

Figure 16. Average CTE of PCEA recycle graphite specimen 04-3-1 (AG) with 27.3% of recycle particles

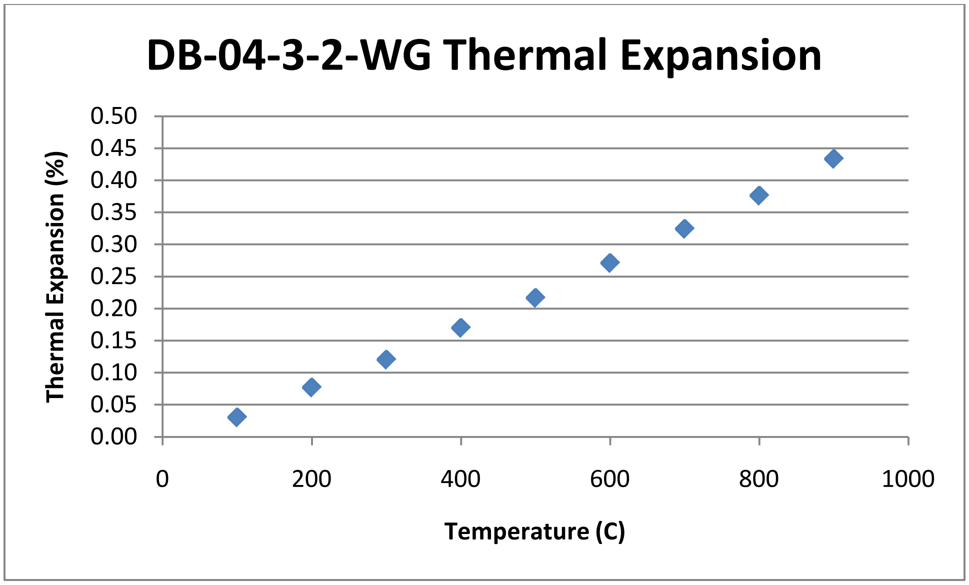

Figure 17. Thermal expansion behavior of PCEA recycle graphite specimen 04-3-2 (WG) with 27.3% or recycle particles

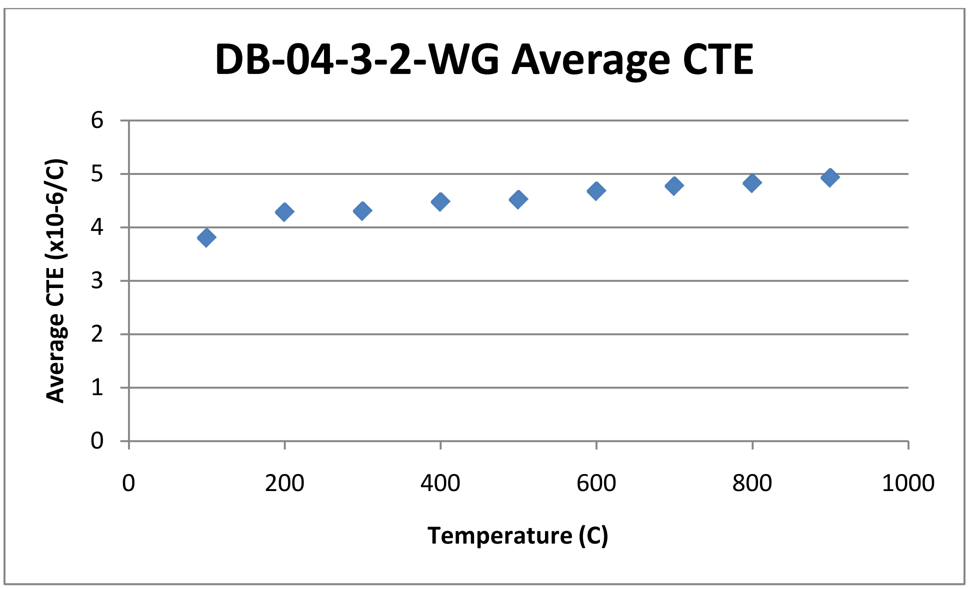

Figure 18. Average CTE of PCEA recycle graphite specimen 04-3-2 (WG) with 27.3% of recycle particles

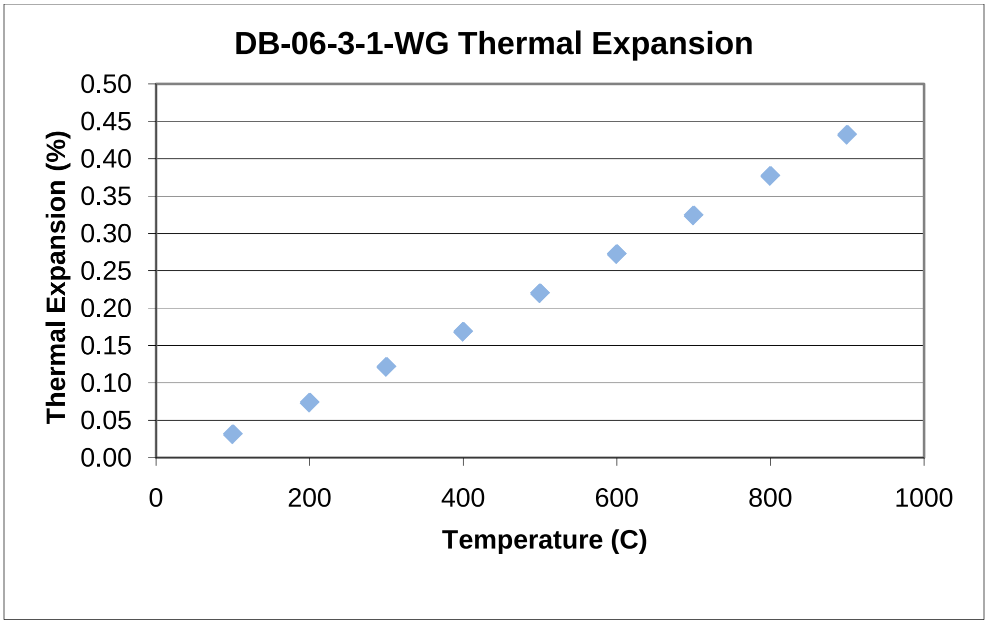

Figure 19. Thermal expansion behavior of PCEA recycle graphite specimen 06-3-1 (WG) with 42.4% of recycle particles

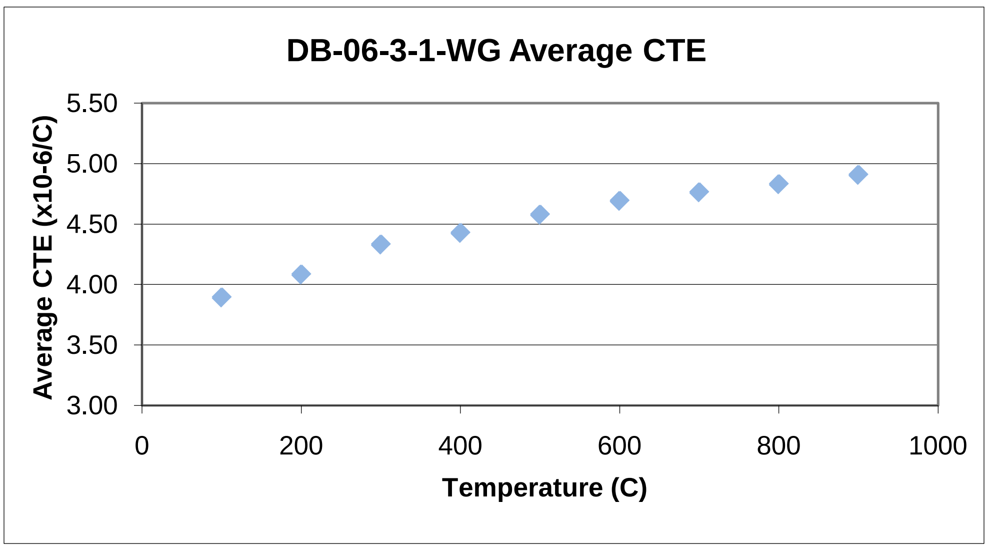

Figure 20. Average CTE of PCEA recycle graphite specimen 06-3-1 (WG) with 42.4% of recycle particles

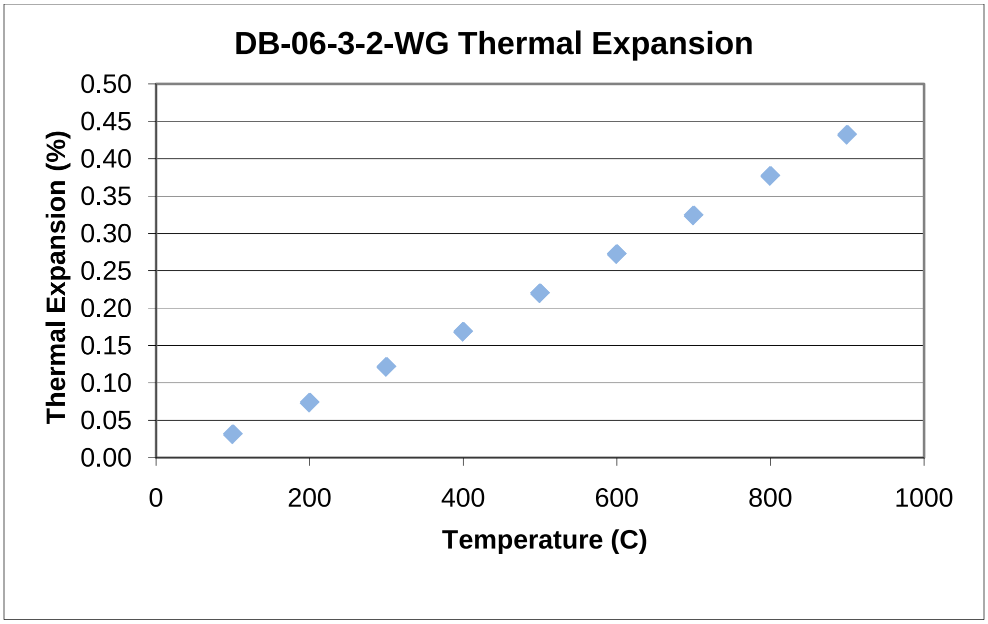

Figure 21. Thermal expansion behavior of PCEA recycle graphite specimen 06-3-2 (WG) with 42.4% of recycle particles

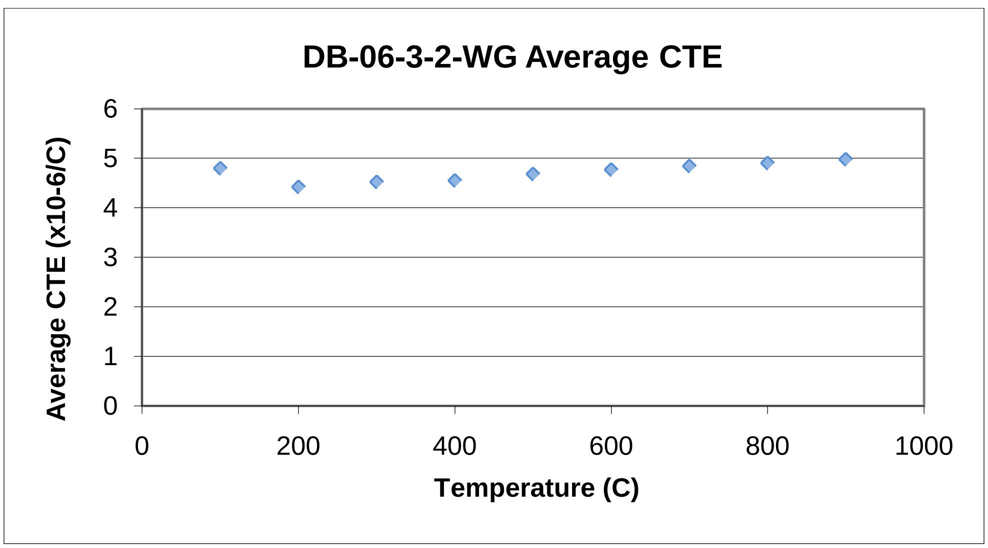

Figure 22. Average CTE of PCEA recycle graphite specimen 06-3-2 (WG) with 42.4% of recycle particles

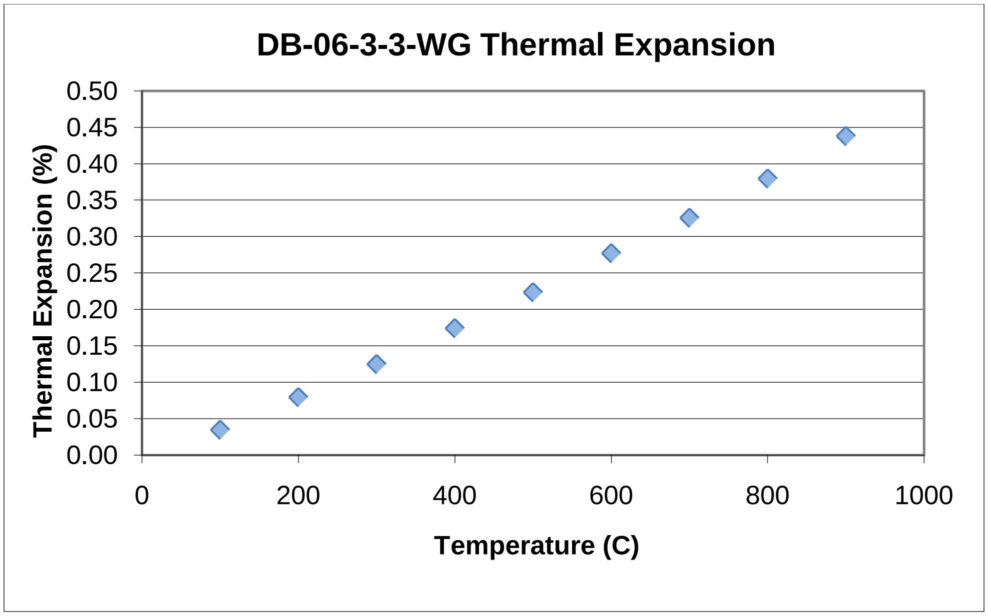

Figure 23. Thermal expansion behavior of PCEA recycle graphite specimen 06-3-3 (WG) with 42.4% of recycle particles

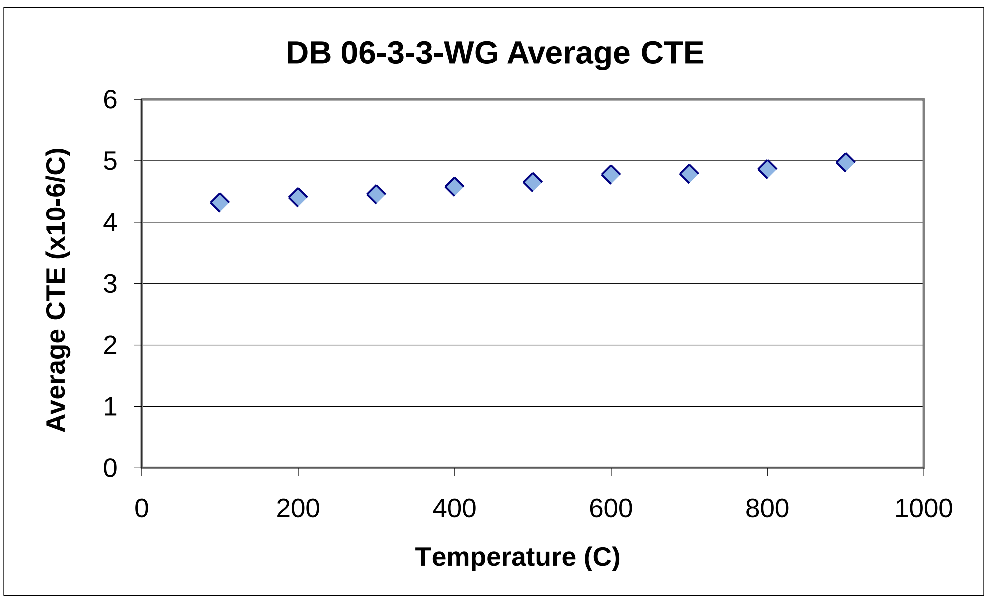

Figure 24. Average CTE of PCEA recycle graphite specimen 06-3-3 (WG) with 42.4% of recycle particles

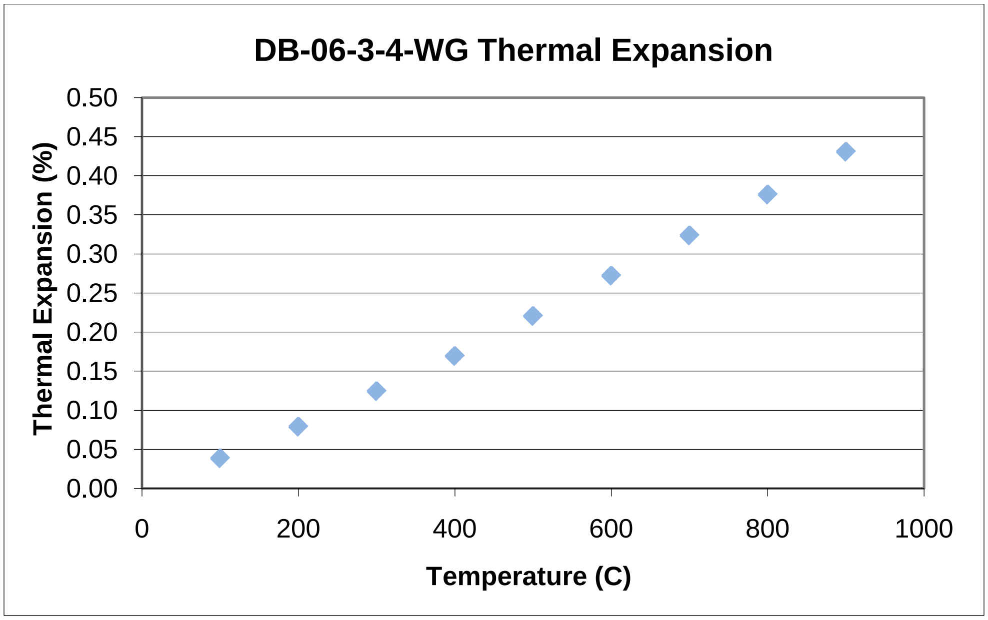

Figure 25. Thermal expansion behavior of PCEA recycle graphite specimen 06-3-4 (WG) with 42.4% of recycle particles

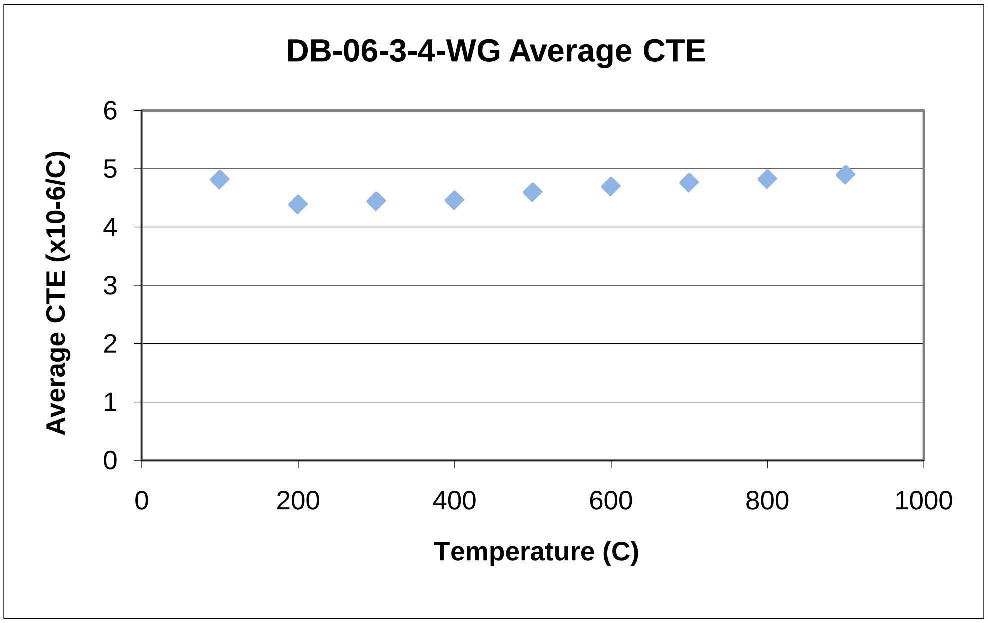

Figure 26. Average CTE of PCEA recycle graphite specimen 06-3-4 (WG) with 42.4% of recycle particles

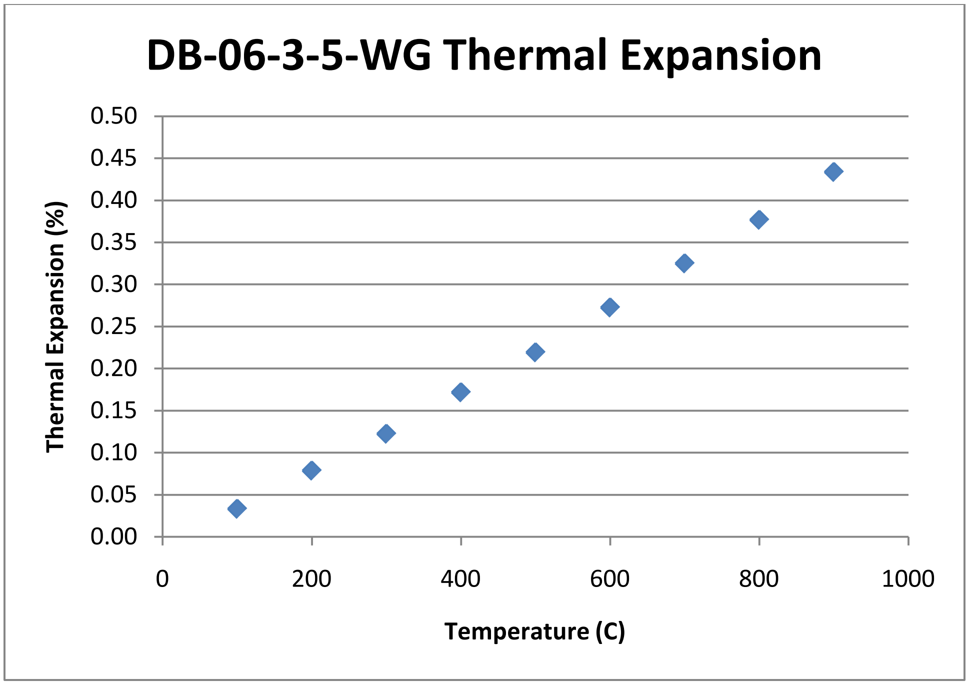

Figure 27. Thermal expansion behavior of PCEA recycle graphite specimen 06-3-5 (WG) with 42.4% of recycle particles

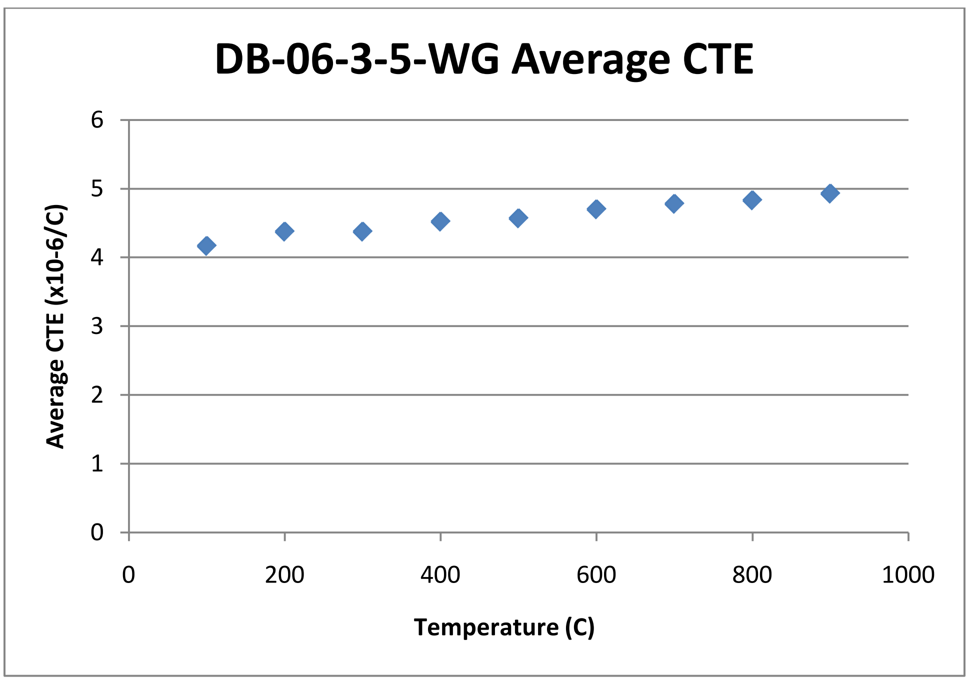

Figure 28. Average CTE of PCEA recycle graphite specimen 06-3-5 (WG) with 42.4% of recycle particles

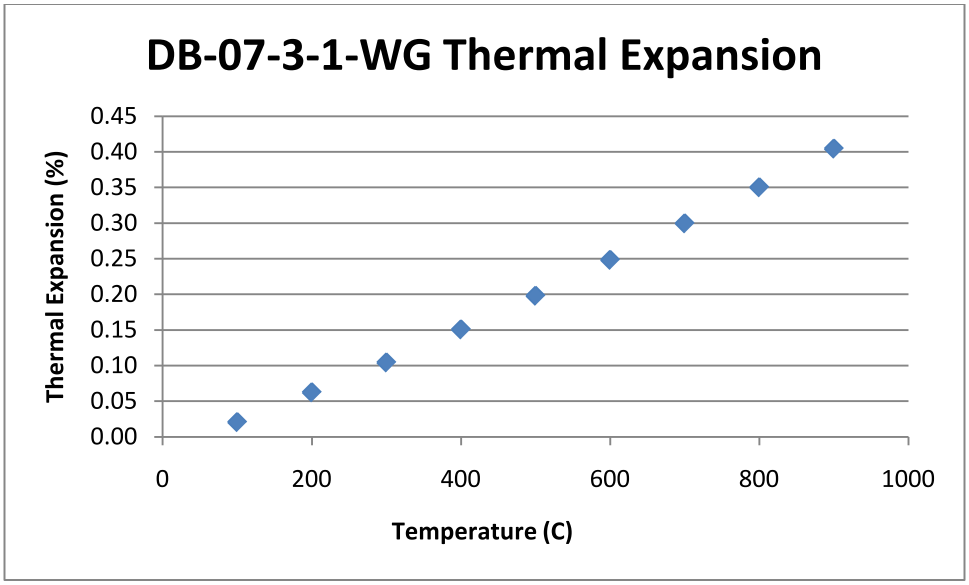

Figure 29. Thermal expansion behavior of PCEA recycle graphite specimen 07-3-1 (WG) with 50% of recycle particles

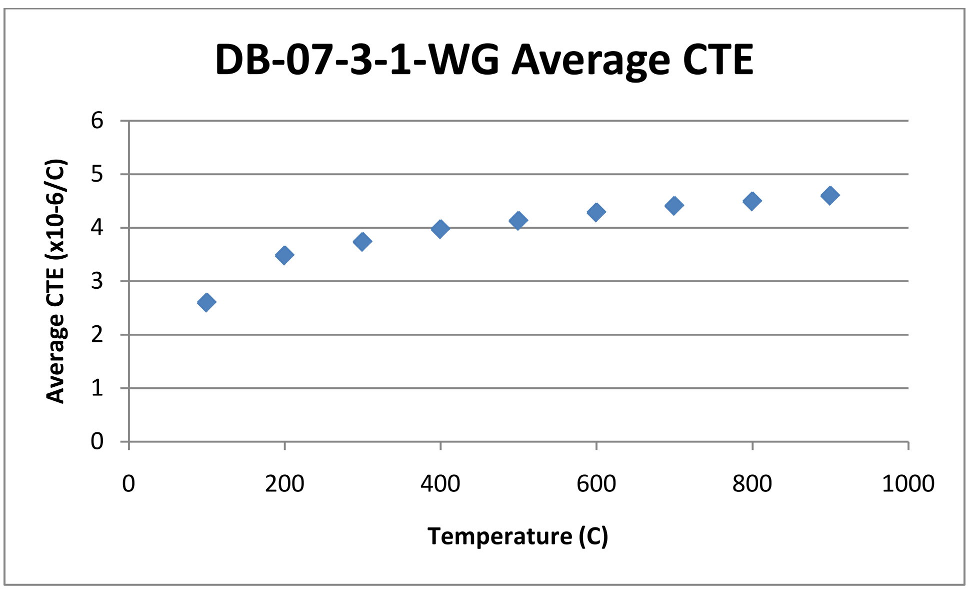

Figure 30. Average CTE of PCEA recycle graphite specimen 07-3-1 (WG) with 50% of recycle particles

All of the specimens tested thus far have exhibited the expected thermal expansion behavior with an increasing expansion coefficient with increasing temperature. The ASTM specification [6] for nuclear graphite required the average CTE (RT to 500°C) in the WG direction to be between 3.5 and 5.5 $°C^{-1}$. All specimens tested to date, regardless of recycle fraction, have met this requirement. Figure 31 compares the average CTE of PCEA recycle graphite billet 04-3 (27.3% recycle fraction) in the WG and AG directions. The AG CTE is larger than the WG CTE as expected. The ASTM Specification [6] requires the CTE (RT-500°C) ratio (AG/WG) to be between 1.0 and 1.10 for isotropic extruded nuclear graphite. The data for billet 03-4 shown in Figure 31 gives the CTE (RT-500°C) as 1.09 indicating the recycle PCEA graphite would qualify as an "isotropic" grade.

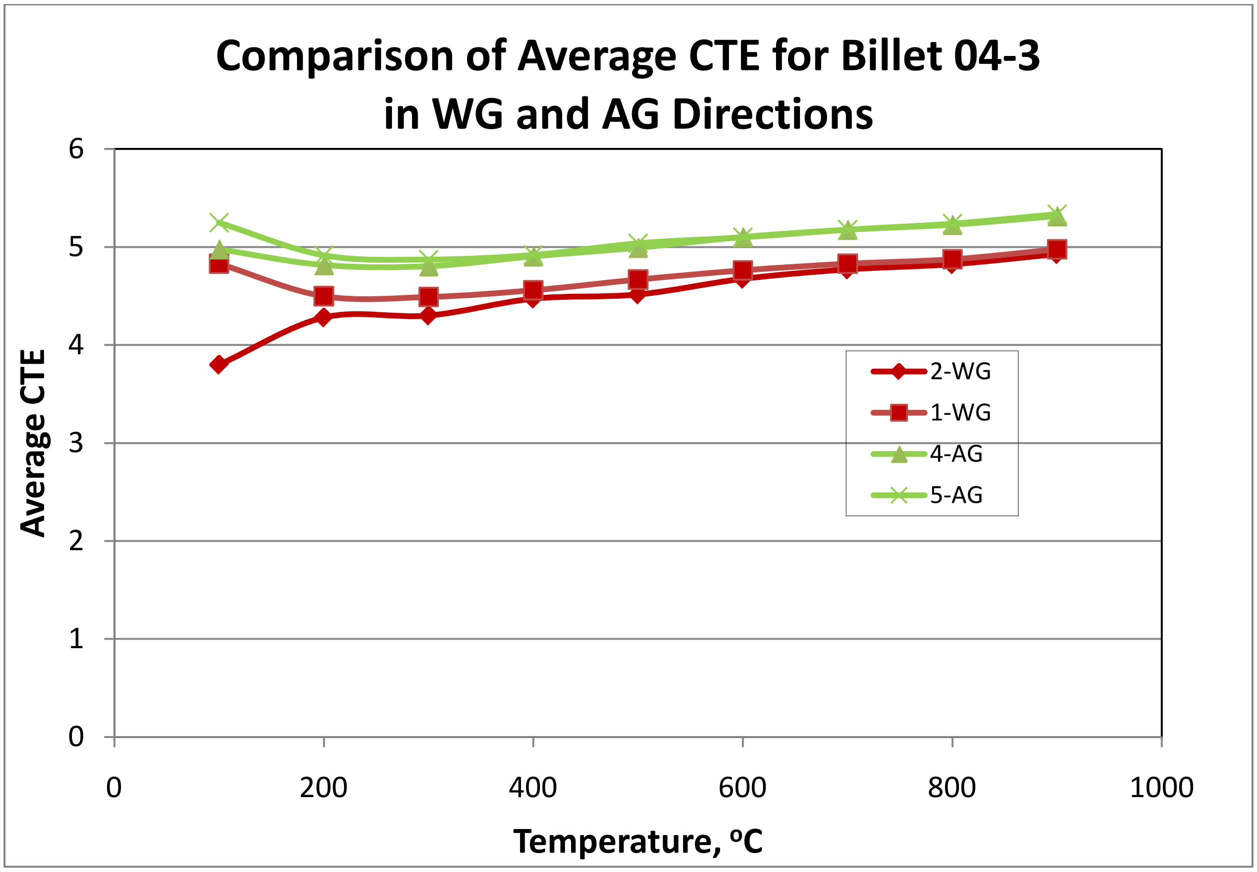

Figure 31. A comparison of the average CTE of PCEA recycle graphite billet 04-3 (27.3% recycle fraction) in the WG and AG directions

Figure 32 shows a comparison of the average CTE of PCEA recycle graphite billet 04-3 (27.3% recycle fraction), billet 06-3 (42.4% recycle fraction) and billet 07-3 (50% recycle fraction) in the WG direction. The limited data set for CTE suggests that increasing the recycle fraction causes a slight reduction in CTE. Further work is needed to confirm this.

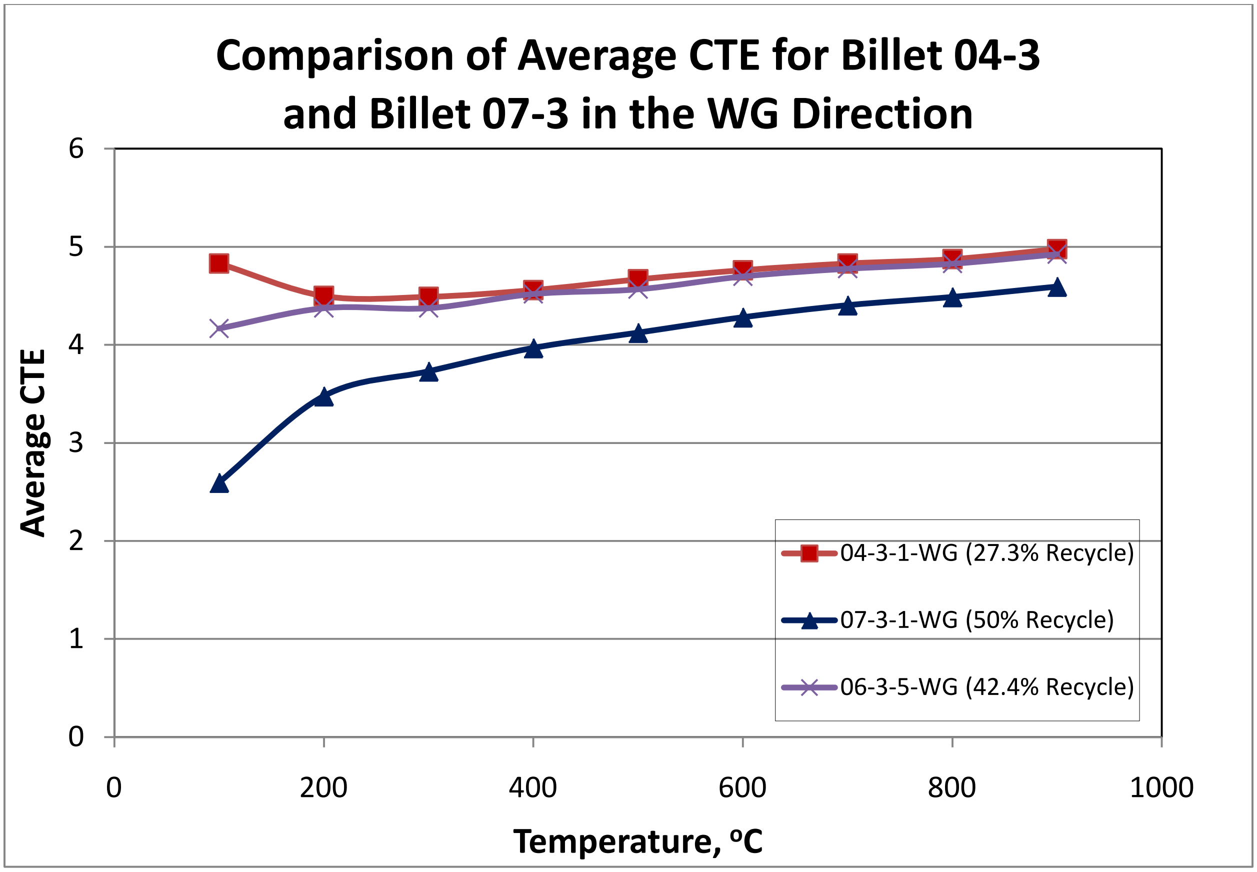

Figure 32. A comparison of the average CTE of PCEA recycle graphite billets 04-3 (27.3% recycle fraction), billet 06-3 (42.4% recycle) and billet 07-3 (50% recycle fraction) in the WG direction

4. Conclusions

A series of recycle PCEA graphite pilot scale billets were manufactured by GTI under contract with ORNL. Thirty specimens were machined from each billet (15 with grain and 15 against-grain specimens). Each specimen has been measured to determine bulk density, electrical resistivity, and dynamic Young's modulus. Coefficient of Thermal Expansion has been determined on selected specimens. The experimental data suggests that adding increasing amounts of recycle fraction has little or no effect on the billet properties. There is some evidence of CTE being reduced by increasing recycle fractions, but the reduction is small. The characterization performed here suggests that increasing the recycle fraction of graphite has no deleterious effect on the properties of the graphite. Thus it appears as if the coke filler could be completely replaced with ground recycle graphite.

Further work is needed to complete this study. Additional CTE measurements are needed to fully characterize the recycle billets. Moreover, a fraction of the specimens from each billet require flexural testing to determine their strength. Our ultimate goal is to irradiated the recycle graphite, determined the effects of recycle fraction on irradiation behavior, and then recycle the irradiated graphite into "new" recycle material   This material should then be re-irradiated to determine if graphitization returns the graphite to its original condition and irradiation behavior.

5. References

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3. ASTM C747, Standard Test Method for Moduli of Elasticity and Fundamental Frequencies of Carbon and Graphite Materials by Sonic Resonance, American Society for Testing And Materials, West Conshohocken, PA 19428
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6. ASTM D7219, Standard Specification for Isotropic and Near-isotropic Nuclear Graphites, American Society for Testing And Materials, West Conshohocken, PA 19428

6. Acknowledgments

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