The Graphite and Carbon Studies Group   1962-1972

In April 1962 our work on Stored Energy was complete. The final report had been written and it was time to move on. The division head had decided to transfer the Stored Energy equipment to the Reactor Division, so that they could continue to monitor the state of the BEPO reactor, I had expected to go with it, but John Simmons wanted me to stay with him and undertake a new project. I must admit that after working on the same subject for 10 years, I was quite ready for a change.

The name of the group also changed at this time, I suppose to reflect a broader outlook in this field. However, the main object of our work would still be the study of radiation damage in reactor Graphite.

My new task was to be the measurement of the Lattice Parameters of Graphite using X-Ray Diffraction. Put in simpler terms: measuring the spacing between the atoms in the Graphite crystal. I had no experience in this field, so I was a little daunted by the prospect of learning a completely new technology. When at college, we had briefly touched on the subject of X-Ray diffraction, but all I could remember was the Bragg formula for the diffraction of X-Rays from a system of parallel layers of atoms was: $2d\sin\theta = n\lambda$ We also discussed simple cubic structures of atoms, and that was it – the limit of my knowledge!

Fortunately John Simmons had found an expert in the field of X-Ray diffraction – it was George Bacon in the General Physics division. I made an appointment to see him. Fortunately he was only a short distance away in Building 153. George had already done some work on the structure of reactor graphite in the 1950's. He gave me a copy of his work, and also a copy of the work that Rosalind Franklin had done in this field, while working at a university in Paris. [Franklin was better known for her early work that led to the discovery of DNA] He also suggested that, as I was new to this field of science, it would be a good idea to obtain a copy of the book: 'X Ray Diffraction Procedures' by Klug and Alexander – a bible on the subject.

I spent two days with George while he explained the structure of reactor graphite. For a start it is polycrystalline and is extremely anisotropic. A single crystal consists of layers of atoms in a hexagonal structure, held together by strong covalent bonds. The forces between the layers are of the weak Van der Waals type. This structure means that the material has a slippery feel, and is often used as a lubricant. The bulk polycrystalline material is made up of a more or less random collection of small single crystals, but it is made anisotropic by the manufacturing process, which is either pressing or extrusion.

Within the single crystal there are two different ways in which the layers can be stacked. These are described as hexagonal: ABAB which is the predominant form, and a small fraction of rhombohedral: ABCABC. As there are three equivalent directions in the basal plane, the planes are best described by the Bravais-Miller system. i.e. hkil. However as i = -(h+k), it is easier to write it as the Miller notation: hk.l the dot standing in for the missing third index.

What I would be measuring was the 'a' spacing, the distance between the atoms in the basal plane; i.e. the distance between the flats of the hexagon, and 'c', twice the distance between the planes for a hexagonal system; i.e. the distance ABA.

All this was a bit overwhelming, so I was glad when George moved on to how these parameters could be measured. He took me on a tour of the various pieces of equipment that were available in his department. These included a Diffractometer, which automatically scanned the sample and displayed the results on a 'Strip Chart' recorder, a flat plate film camera for studying single crystals, and various Debye-Scherrer powder cameras.

As we were dealing with a polycrystalline material, the best way, and the most accurate, was to use a Debye-Scherrer powder camera. In this the sample is placed at the centre of a cylindrical camera, and the diffraction pattern recorded on a strip of photographic film wrapped around the cylinder. If the diffraction angle of a line is θ, the X-ray beam is deflected by 2θ. In order to avoid the problem of locating the zero position, in the centre of the exit X-ray beam, it was normal practice to measure the angular distance between the same line on each side of the camera, which would be equal to 4θ.

One popular powder camera had a diameter of 11.46 cm. This may seem an odd figure, but it meant that one degree on the films cylindrical surface was equal to one mm. Also, The film could be in one piece, so that a direct measurement of the angle 4θ could quickly be made using a millimetre ruler. It was however necessary to punch a hole in the film where the X-ray beam leaves the camera. This simple system was very useful in analysis, and identification of unknown materials. The pattern of lines could be compared with a published index.

However George suggested that, as we needed to have very accurate measurements, the most suitable system was the 19 cm. Debye-Scherrer camera. This was more complicated; because of the large size, it needed two strips of film, one on each side of the camera. Note that one strip of film would be almost 60 cm. long and difficult to process. This additional complexity did mean that the calibration of the system was very different. A knife edge, on either side of the exit tube cast a shadow on the film. A similar knife edge was located on each side of the entry tube. Each knife edge pair was a single piece of metal with a hole through the centre for the X-ray beam. Calibration of the system only required the diameter of the camera, and the linear distance between each pair of knife edges. Both these could easily be measured using a micrometer. Of course that did mean extra complexity in using two films; it was now necessary to measure the position of the lines from the knife edge.

I must admit that the extra complexity of the 19 cm. camera was a bit daunting. However George assured me that the extra work was well worth the effort to get the most accurate results. He also warned me that the use of film required great care. It was not a good idea to punch holes in it, and it did not like temperature extremes.

George then introduced me to his assistant Stewart Wilson, who would show me how to prepare the samples, how to use the camera, how to process the film, and how to make the measurements. I spent the next week with Stewart and learnt a lot of new skills.

Following this I was a bit overwhelmed, and uncertain how I should proceed. I was therefore surprised to get a phone call from George Bacon, asking to see me. As I went to his office, I was wondering what he wanted to say. He told me that he was leaving Harwell to take the post of Professor of Physics at Sheffield University. Because of this, his staff and all their equipment would be transferred to other groups. He then asked me if I would like to take over the X-ray machine and the 19 cm. camera, that I had been using. It seemed that it was all old equipment, that no one else wanted. He said that he would also throw in the film measuring machine as well. It was an offer I could not refuse. I did not bother to ask John Simmons, I just gratefully accepted

the offer. The only problem was that he wanted all the equipment removed from building 153, before the end of the month.

As the X-ray machine was so big and heavy, I had to call in the 'Heavy Gang' to transport it to building S37. I personally made several trips to carry the more delicate items. I was very worried that something may be damaged. I put all this in the second big lab. which was now half empty. So now I had a General Electric X-ray generator, complete with 40 Kv. Power supply, a Cambridge 19 cm. Debye-Scherrer camera and a splendid film measuring machine made by Cambridge Instruments to an N.P.L. design.

I now had to persuade John Simmons that I needed a special room for the X-ray machine, which also had to be 'Dark Room' for processing the film. There was no choice; John agreed to build two new rooms at the far end of the empty lab. It fitted in well, because we also needed another office, to be shared by the two new Scientific Officers, Peter Thrower and Peter Goggin. It was agreed that I would retain my office in room 13A. The film measuring machine could be conveniently located in the temperature controlled room, which was used for the dimensional change measurements.

All this, of course, would take some time to set up, so it was convenient that I was just about to get married and take some time off.

Getting Married

So far I have said very little about my personal life. Not that it was insignificant, but just because I have found it difficult to weave it into the general story of my working life. More details can be found in a later chapter on 'Family History'.

I met Alison Donovan at the Reading Mountaineering Club Christmas dinner in December 1960. An area of common ground was quickly established; she was just about to go skiing at Saas Fee in Switzerland, and I had spent three weeks there last summer climbing the mountains. This made the conversation flow very well. I soon found that she was studying Dairy Science at Reading University. She was not only beautiful but very clever. I was most impressed, and quickly decided that this was the girl that could change my life. She did indeed change my life. We were engaged in the summer of 1961, but because Alison wished to complete her degree, the wedding was delayed until she had taken her 'finals' in 1962.

We were married at Holy Trinity Church in Sutton Coldfield on the 25th August 1962. Our honeymoon was spent touring around Europe in my bright red TR2 spots car. We were camping most of the time, but stayed in a hotel when we needed a bath and a bit of luxury. Our only objective was to visit Rome. We travelled through France, Switzerland, Italy and Austria. Visiting Pisa, Rome, Siena, Florence and Venice. We also spent some time in the mountains of Austria and Switzerland. In all we travelled about 2,600 miles. After almost three weeks we returned to our new home 'Solina' in the village of Cold Ash, near Newbury.

Back To Work

When I returned to work, I found that the new X-ray room had been built and was just waiting for the plumbing etc. to be installed. I had to wait another two weeks before it was completed and decorated. Meanwhile I had the time to order things that I would need: X-ray film, developing chemicals and the little extras that I would need to prepare the samples. Stuart Wilson had used a

standard developing tank to process the film. I did not like this as the film was constrained in a tight spiral. I decided to use a developing dish, where the film could remain flat. The disadvantage of this was that the dish was not the right shape for two narrow strips of film, so it wasted a lot of developer.

When I eventually moved the X-ray machine into the room, I found that, as the room was quite small, I had to position this so that the X-ray beam was facing directly at the office next door. As the wall was only a thin partition, I had to fix a thin lead shield to the wall to protect the boys next door. Of course there was a beam stopper on the camera exit tube, but as this had to be removed when inserting the sample, it was possible that it could be left off by accident.

Calibrating and Testing the System

I was now ready for a test. George had suggested that I measure a sample of Diamond powder. This would give me some experience of using the system, and also enable me to calibrate the camera. He had given me a small bottle of Diamond powder, and a supply of sample tubes. The tubes were ½ mm. bore, sealed at one end, and the other end a funnel for loading the sample powder. The tubes were very thin glass, which produced no X-ray pattern. With the tube fixed in the sample holder, I then had to make sure it was perpendicular and centred in the camera. This was elegantly done by placing a light source in the X-ray entry tube, and a small telescope in the exit tube. On rotating the sample holder it was possible to check and correct the sample position. [Of course it was necessary to ensure that the machine was switched off first!]

After a short X-ray exposure I developed the films, and was delighted to see a large number of sharp lines. I waited until the next day, to ensure that the films were completely dry, then started the measurements.

Using unfiltered Copper radiation with a wavelength of 1.54 Å. I was able to measure thirteen lines. A small correction was made for vertical divergence; then a linear extrapolation made against diffracting angle θ to $\theta = 90°$, using the method of Nelson and Riley, to allow for absorption. This gave a value for the 'a' spacing of Diamond of 3.5667 Å, in good agreement with the accepted value. [I was surprised to find that Diamond, which is a very strong material, where the four co-valent bonds of the carbon atom, are in a tetragonal form, actually had a unit cell that was cubic!]

Preparing the Graphite samples

Stuart Wilson had given me some instruction on how to prepare the samples for the X-Ray camera. The graphite was first powdered, then mixed with a small amount of Gum Tragacanth, and a little water to make a paste. This was then rolled between two microscope slides, until it was ½ mm. in diameter. When dry it was cut to the correct length and installed in the camera specimen holder. This required a certain amount of skill to get the right diameter. I later used a small press to extrude the mixture though a die of the correct diameter. The Gum used gave no significant addition to the X-Ray pattern. I found that the best way to powder the graphite was to drill a hole in the bulk material, using a small hand tool called a Pin vice. This meant that the sample could be returned for further use.

Measurements on Unirradiated Graphite

In unirradiated grade 'A' graphite, only ten lines were available.

The 00.2 α&β, 00.4 α&β and the 00.6 α giving the 'c' spacing
The 10.0 α&β, 11.0 α&β and the 12.0 α giving the 'a' spacing

Lines of a general index: hk.l were not used.

The same extrapolation method was used But because of the small number of lines, and the low angles at which they occur, no claim can be made for the absolute accuracy. A mean value for three different blocks of grade 'A' graphite was:

$c = 6.7198 \pm 0.0006$ Å   and $a = 2.4618 \pm 0.0006$

Lines of a general index: hk.l   originating from pyramidal planes, are less easy to deal with. However they can be used by a method of successive approximations. It does require as a starting point the ratio of a/c, which is already known from the other lines. A calculation can then be made from the line data, giving new values for a & c, and hence a new ratio. The process is then repeated until the final value of the ratio is equal to (or close to) the input value. Of course it does require that the calculation should converge. However this did work and give good results, but was an exceeding tedious iterative calculation. This was very suitable for a computer calculation, and this gave me a good excuse to write a program to do the whole calculation. [However, Bacon has shown that these lines can only give a and c for an ordered group of layers, hence these values would be different in graphites which contain a significant amount of disordered material.]

Programming in Fortran

I was already familiar with the Fortran programming language, from my work on Stored energy. However I had not written a program on my own; I had always had some help from Dennis Rimmer in the Theoretical Physics division. Without thinking much about it, I plunged straight into writing the program. Of course I did make a number of blunders. The worst, and most conspicuous error was when I forgot about the 'Carriage Control Character'. This was the first character of a printed line, which determines what the printer carriage feed does before printing the rest of the line. It can advance one or two lines, or to a new page. Blank gives one line, '0' two lines, and '1' goes to first line of the next page. As I often started the printed line with 'hkl', and h was often '0' or '1', I got into a right mess with lots of blank pages. I was ticked off by the computer manager, and given a lesson on how to write Format statements. It seemed that this was a fairly common error.

As we now had an IBM 360 computer at Harwell, it was much easier to submit jobs, and I often got the results back the same day. The program was a great success and saved a lot of time in tedious calculations.

Elliot Automation

During this time I had been requested to advise on a project to provide a simple standard method of measuring Stored Energy. Elliot Automation had been given a contract, to design a new system that could be used by all the nuclear power stations. I went twice to London to visit the company, first to agree what was required, and later to see what they proposed. I did not approve of what was suggested. Our apparatus used a closed loop control system, with a linear DC power amplifier. Elliot were proposing a control system using SCR's (Silicon Controlled Rectifiers), These work of AC mains, and control by switching off part of the AC cycle. This would produce

a lot of high frequency noise and may effect the sensitive temperature measurement system, and cause instability. After that I heard nothing more about their proposed system, and never knew whether it was ever built.

Measurements on Irradiated Graphite

The lines became fainter and more diffuse with increasing neutron dose. At an irradiation temperature of 650 °C all the lines were still measurable, so the same extrapolation method could be used. However, at 200 °C only the 00.2 and 10.0 were measurable, even at modest doses. When only a single line was available, the spacing was not given, only the change relative to the unirradiated material; $\Delta c/c$ and $\Delta a/a$. In this way the systematic errors were minimised.

An unwanted move

In less than a year into my new task, we were told that we would have to move to another building. This was because, unknown to us, the plot of land that enclosed our building (S37), was the site for the new Ceramics building. We did not have any choice. It was ironic that we were to move to Building 153, which is where the X-Ray equipment was located before I moved it to S37! I was not pleased. George Bacon had now departed to Sheffield University, and his group dispersed, so 153 was now completely empty. This came at a bad time for me as it coincided with birth of our son.

Our first child is born

Alison became pregnant soon after we returned from our honeymoon. She suffered from a lot of 'Morning Sickness', but it all turned to joy when our son, Andrew Robert was born on the 3rd July 1963. He was born at home, in the room that we had dedicated for the nursery, and I was present at the birth. Following this, it was a lot of work for both of us, but a wonderful time.

Moving to 153

As 153 was completely empty, we had a long discussion about how the space should be allocated. I got the whole upper floor wing on the north side. One very large lab. in which I had just one work bench, a very nice office on the north east corner, and adjacent, a small lab. with sink and all plumbing already fitted. The latter made a good space for the X-Ray machine.

It took me several weeks to move all my equipment to the new building, and to get things set up. I took this opportunity to improve the way in which I developed the films. I had constructed a water tank which was controlled to be at 20 degC. This had the facility of loading the new developing tubes, which allowed the film, in a carrier, to remain flat. It also pumped water to the X-ray camera to keep it cool. I was very pleased with the new setup.

The film measuring machine was again installed in the temperature controlled room, which was used for the dimensional change measurements. I also stored the films in this room.

The move had interrupted all our measurements, so we had a large backlog to catch up. I think that, in the end we lost about 3 months. The big bonus was that my new office was very pleasant, and had an attractive outlook, which was a big change from the dismal view from my shared office in S37.

Continuing the Measurements

It did not take long to catch up with the backlog. However, for me, the results were a little disappointing! It soon became obvious, that the changes in the lattice parameters, bore very little relation to the dimensional changes in the bulk material. The 'c' spacing increased, and the 'a' spacing decreased, but soon saturated. The bulk material however continued to change; Expanding in the direction perpendicular to the direction of extrusion, and contracting in the direction parallel to extrusion.

For irradiation temperatures of 650 degC and above, the changes were very small or insignificant. Only at 200 degC did the results show a feature in the 'c' spacing change that saturated at two different levels. Subsequent annealing of the samples, up to 1840 degC, showed that the lattice parameters returned to their pre irradiation values, but the dimensional changes in the bulk material did not! This leads to an interesting conclusion about the nature of the damage.

The preliminary results of these investigations were published, in a joint paper, in the Carbon Journal in 1964. Peter Goggin included the photographs of single crystals, Alan Perks the bulk dimensional changes, and I provided the X-Ray measurements. Bill Reynolds, our deputy group leader, stitched the whole thing together.

Peter Thrower was studying the radiation damage using an 'Electron Microscope'.

A list of publications will be given at the end of this chapter.

Our Second child is Born

Our beautiful daughter Debbie Mary was born on 16th April 1965. She also was born at home, in our house Solina. This was a great joy and made our family complete. However, it did mean a lot of work for Alison.

At this time, just before Debbie was born, we had decided that our house was too small. We needed more bedrooms. I planned the changes that we wanted, but we did hire an architect to make sure it was a workable design. As we lived in a bungalow, we could expand upwards. A new double garage was built, and integrated, on the east side, and the old garage converted into a dining room and a utility room. To install the staircase, we extended the hall into the small third bedroom, which had been the nursery. Upstairs we built our new bedroom, complete with en suite, above the new garage. Two more bedrooms were built in the roof space, along with a large storeroom at the west end.

The building work started in June 1965. As soon as they had built the foundations of the new garage, they wanted to get inside the house. We had arranged for Aly and the two children to move to Glanyrodyn, in Wales, for the duration of the building work. I would stay at Solina, and visit the family at the weekends. Glanyrodyn was a holiday house owned by Alison's mother. It was situated in the tiny village of Tresaith in west Wales, not far from Cardigan. It was a delightful house, overlooking a magnificent beach.

Alison arranged a stream of visitors, so it was not so lonely being there with just the children. However, it was hard work for her. I managed to visit almost every weekend; it was a long trek, over 400 miles for just two days. However, being at home, during my working week, meant that

I could keep a close eye on the progress of the building work, and was also on hand to answer any queries.

At first the work progressed quite rapidly, but when the main structural work was done, it seemed to slow to a crawl. The finishing work seemed to take forever, some weeks little progress was made. I did complain, but it made very little difference.

When it got to October and it began to get cold, Alison wanted to come back, but the house was in no fit state for this to happen. I looked for some where to rent locally, but it seemed difficult to find the right place at a reasonable price. Fortunately a friend advised me to try the Housing department at Harwell. I must admit that I did not know it existed. They turned out to be very helpful, and offered me a 'Prefab' on the Harwell site at a modest rent. The next weekend I moved the family from Wales to our new accommodation. The 'Prefab' turned out to be a great success, much better than I expected. The 'Prefab' was located in Wayland Cresent, part of a small estate on the southern edge of the Harwell site. It had a kitchen, living room, bathroom, and two bedrooms. It was very convenient, only a short walk for me to get to my work, and there was a grocery shop on site.

It did have one disadvantage; it was very poorly insulated. It was unfortunate that it was a very cold autumn with temperatures well below zero. The only heating was a coke stove in the living room. We had to supplement this with electric heaters brought from Solina.

As Christmas approached, Alison was desperate to return to Solina. However, the house was still not finished! One week before Christmas we turned the decorators out, collected all the pots of paint etc. and put them in a cupboard. We moved back into our house just in time to enjoy a very happy Christmas with our new family. The decorators returned in the new year to complete the job.

[The 'Prefab' was an 'Aircon', made of aluminium, and built in aircraft factories following WW2. It was a very clever design, made in two halves. One contained the kitchen, living room and bathroom, the other the hall and two bedrooms. If the reports are to be believed, they produced one every 12 minutes! It could be installed on a prepared concrete base in one day!]

Cold Neutron Scattering

In the midst of all this action, John Simmons asked me to help David Martin who wished to study radiation damage using Cold Neutron Scattering. A facility was available on the Dido high flux reactor, which was run by Peter Egelstaff. This had small tank of liquid hydrogen at the centre, which was the source of the cold neutrons. Suitably filtered, this provided a beam of low energy neutrons, with an equivalent wavelength of 7.5 Å. These neutrons would not cause any damage, and would not produce any Bragg reflections. However, they could be used to study defects in the carbon structure.

I set up the experiment on the Dido reactor. This was a rotating sample holder, holding three things: An irradiated graphite sample, an unirradiated sample of the same graphite, and a piece of Vanadium. Using a timer, this put each sample in the neutron beam for 30 minutes at a time, and the scattered neutrons recorded on a bank of BF3 neutron counters. The experiment was run for two days, and the results recorded on tape. Following this the tape was analysed to show the scattering pattern for each sample. Vanadium is an isotropic scatterer, and was used to calibrate the system.

Further runs were made after annealing at temperatures up to 1100 degC. Also with samples which had been irradiated at temperatures up to 450 degC.

David Martin interpreted these results as being produced by a group of 4 +/- 2 interstitial atoms. However, it appeared that the size of the group did not significantly change, either with irradiation temperature, or the state of the anneal. I was very sceptical, but David managed to publish two papers on this subject.

Back to normal

The work for David Martin did interrupt my routine measurements of lattice parameters. After the delay, I continued measuring samples irradiated in various reactors. These included Dido and Pluto at Harwell, the DFR fast reactor at Dounreay, and even some Nuclear Power stations.

Another Move !!!!!

In 1967 the head of the metallurgy division decided that our group was too far removed from the main building. As there was now room to spare, he wanted us to move into building 393. I was very cross; it seemed such a stupid idea, and it would waste yet more time and money. John Simmons, Alan Perks and the main part of the group were housed in good offices and labs. in the central corridor. Bill Reynolds was moved to another department; not sure why.

Peter Goggin and I were given a very unsuitable area, previously known as the 'Snake Pit'. It was two very large rooms that were about four feet lower than the rest of the building. It had no offices or small rooms! This meant that we had to wait until this was modified to add offices, a dark room, and a room for the X-Ray machine.

At this time Peter was given a new job to study and test 'Carbon Fibres'. He was glad of the extra space, as he had to install testing machines to measure the strength of the fibres.

While waiting for the building work to complete, I amused myself by writing a satirical piece called 'Raving Buildings', a parody of the childish game of 'Musical Chairs'. Most people thought it was very funny; I only wish that I had the nerve to send a copy to the division head.

Back to work

It was several months before I was able to continue my measurements. I had a huge backlog of samples. I also wanted to try and measure the crystallite size for the unirradiated graphite, and also for the high temperature irradiated samples. This meant that I had to measure the line profile, and my machine had no facility to do this. I decided to modify the film measuring machine in order to do this, but I still had to carry on with the routine measurements.

A little bit of Automation

First on the list was to replace the eyepiece with a photometer, the output of which was connected to a digital voltmeter. I then managed to find someone in the engineering division to advise me on fitting a screw to the machine to move the carriage. I got good advice, and the job was given to an external precision engineering firm. The result was excellent and worked well. The screw was turned by a 'stepper' motor, which was driven by a standard '2000' series unit. The 2000 series

was a Harwell collection of modular units, which could be fitted into a 19" rack. The steps were counted on another module, and the output from the counter and the digital voltmeter, were scanned out to a paper tape punch. The only punch available was part of a 'Teletype'. This was a teleprinter with an attached punch and reader. This proved to be more useful than I expected. The whole system worked well, and I could either just scan one line, or the whole film. The paper tape, together with a Fortran program was processed on the Harwell IBM 360 computer. I was very pleased with this new system.

HUW - Harwell Users Workshop

At this time a new computer facility became available. It allowed realtime access to the IBM 360. It consisted of a 'Mini' computer (a Dec PDP 11 – I think) which provided an interface to the IBM. To this could be attached a number of Teletype terminals. The Mini computer acted as a line concentrator. At first only 40 terminals were available; most taken up by the Theoretical Physics division. Later, with another Mini computer, they added another 100 lines. Only one line was allocated to our division, I managed to get this line, but had to share with another user. With this I could write a Fortran program, compile it, run it and have the results printed on the terminal. For a simple job, this could be accomplished in only a few minutes.

Crystallite Size

This measurement could only be carried out on unirradiated samples, as the radiation damage caused line broadening. It was necessary to measure the width of the line after removing the effect of instrumental broadening. This was done by a method due to Stokes, which used Fourier Analysis. This sounds complicated, but it was easy to write a Fortran program to do the calculation.

An extruded PGA graphite gave the values $L_c = 400$ Å and $L_a = 700$ Å

A near Isotropic graphite $L_c = 170$ Å and $L_a = 365$ Å

The End is Nigh

In 1969 the group was shrinking. Several people had left, and were not replaced. Peter Goggin had moved out to building 401, to provide a materials testing service. He was still testing Carbon Fibre batches, and composites, but he now had responsibility for several large testing machines. At the end of the year Peter Thrower left to take the position of Professor of Physics at Pen State University in the USA. I was now left alone in the 'Snake Pit' and was worried that there would be pressure to make me move again! I had already made it clear that I would not move the X-Ray machine again!

In 1970 I was still measuring the constant stream of samples from various reactors. However, I was very concerned that, although my data had contributed to several joint papers in the Carbon Journal, I had nothing that I could call my own. I resolved that it was time for me to correct this dismal state of affairs.

I had already tried, and failed, to explain the 'steps' in the lattice parameter changes in the irradiated samples at 250 degC, so I needed something completely new.

The Relationship of the Dimensional changes of Polycrystalline Graphite to the corresponding changes in constituent Crystals

I started with a simple geometric theory of how the crystals were stacked. The result depended on the size and shape of the crystals. My first attempt gave a shape that, although not out of line, was a bit unrealistic. I later modified this theory with a crystal shape that was ellipsoidal, with the ratio La/Lc = 2 (approx). It all worked well, and I was able to calculate the relation of thermal expansion and dimensional change, between the two states.

A simple explanation of Neutron Radiation Damage

When Graphite is bombarded with neutrons, some carbon atoms are displaced from the lattice, and end up stuck between the planes. This causes stress by forcing the planes apart. This stress gives rise to the Stored Energy we see when the temperature of radiation is low, say less than 100 degC. Above this temperature, the interstitial atoms become mobile, which would lead to the formation of groups, and in the extreme case, create a new plane. The holes, or vacancies, left in the planes are less mobile, but when there a several close together, it is thought that the strong bonds in the plane would cause it to collapse. Again in the extreme case, this would lead to a reduction in the diameter of the crystal.

Putting this model into the theory, it is possible to calculate the dimensional changes of the bulk polycrystalline material as it changes with neutron dose. The results show very similar features to the high dose irradiation's at 400-600 degC.

Conflict

I was very pleased with the theory, but when I showed to John Simmons I got a very hostile reaction! I had a very good relationship with John, he was very clever, and had been my mentor for almost twenty years. We did not always agree, but when we did he would be patient and explain in detail why I was wrong. He was nearly always right!

This time however, he just seemed angry and did not wish to discuss the theory. I was quite shocked and left his office feeling completely squashed. I did keep pushing, and eventually he agreed that I could publish it as an internal AERE report, but he would not recommend publishing it in the Carbon Journal.

I feel I got a 'black mark' for all my efforts. I already had one because I had refused to carry on Peter Throwers work on electron microscopy. I did not think that I was suited to interpreting the data on photographs. All my experience was in measurement.

Serendipity

I still had contact with Peter Goggin. Although he was now in a different building, we still met frequently to discuss our progress, or lack of it! I was very interested in the work he was doing on Carbon fibres, and how this was applied to making composites. I was not familiar with the structure of the Carbon fibre, so driven by curiosity I begged a sample and popped it into my X-Ray camera. I was not sure how to mount the sample, so I put several fibres into a glass specimen tube. The result was a very weak picture, and I could not interpret what I had found. I had been told that the structure was a chain of C=C bonds. I tried various methods of mounting the fibres, but had little success. Finally, as I was using a powder camera, I decided to cut the

fibre into very short lengths, and ground it in a mortar. I was well aware that this was a bit brutal! The result was quite startling. The lines on the film were well defined, but at first I could not recognise the pattern. However, when I compared it with a sample film of Graphite, it soon became obvious that it was 'graphitic'? but with a difference. The HK.0 lines were all present, but there was no sign of the 00.L lines, or the pyramidal lines. I could only think to interpret this in one way. The sample appeared to contain well ordered hexagonal nets of carbon atoms, but there was no coherence between adjacent layers! It seemed to be an impossible structure, and I was at a loss to explain what I was observing. After much thought, I came to the conclusion that the layers must be wrapped into a cylindrical spiral. This would retain the structure of the individual layers, but would prevent any correspondence between the layers. The adjacent layers of atoms would drift in and out of phase, so destroying any coherence.

I took this evidence to John Simmons, who for once seemed to take an interest in my wild ideas. He was just about to go to a meeting about Carbon fibres, so he asked me to go with him, and explain what I had found. Unprepared, and faced with a lot of senior physicists, I struggled to put this into words. When I had finished, there was a long pause, and then I was bombarded with a lot of aggressive questions, many of which I could not answer. I got the awful feeling that most of the people at the meeting, did not believe what I was saying, or even what I had done. There were many questions about the origin of the fibre, and how I had prepared the sample. The overall consensus was, that I had 'butchered' the sample, and it was no longer Carbon fibre! So, much to my amazement, they were not interested!

After the meeting John said that this was an oddity, and not my problem. He told me to drop it and get on with other work. I did get back to my routine measurements, but I could not get this out of my mind.

At the meeting, it was pointed out that although there would be no exact correspondence between the layers of the spiral, there would be some evidence of the layer structure. I was just wondering how to explain that, when I visualised the hexagonal net being wrapped into a ball! However, I could not see how this could be possible.

I then remembered, that in my theory of the polycrystalline graphite structure, the calculated density was slightly lower than the measured density. This had pointed to the possibility that there was some non-graphitic carbon, trapped in the space between the crystallites.

When reactor graphite is manufactured, coke is ground to a powder, mixed with pitch, and heated in an oven. It is then 'graphitised' in a furnace at 3000 degC. It was a puzzle to me, that some of the carbon resisted being converted to the graphite structure. It could not be impurities, as the final material had to very pure, to ensure that it had a very low cross section to neutrons.

If it were possible to wrap the hexagonal net into a ball, so that it had no external bonds, then this would satisfy both cases! The 'ball' would still show the properties of the hexagonal net, but would not show any layer structure. It would also be invisible in the normal graphite X-Ray picture.

I took a piece of 'triangular' graph paper, and drew on it two hexagons joined along one edge. The external angle, where the hexagons met was obviously 120 deg. However, by folding the paper along the line joining the two hexagons, it was obvious that the external angle was reduced. If the angle was reduced to 108 deg. It would be possible to fit a pentagon into the gap. Reducing the angle to 90 deg. would fit a square, and with a further reduction to 60 deg. the gap would

accommodate an equilateral triangle. It was also obvious that the angle could not exceed 120 deg. so the it would be impossible to fit a heptagon, or any greater polygon into the hexagonal net.

This elementary geometric observation quite excited me, as it was clear that the introduction of pentagonal holes in the hexagonal net would cause some curvature. I next drew on the graph paper a ring of six hexagons, surrounding a central hexagon. I cut this out and removed one hexagon from the ring, and also cut out the central hexagon. I then joined the loose ends of the ring with a piece of cellotape. This was very encouraging, as I had five hexagons surrounding a pentagonal hole. The structure was curved like a small bowl. At this point I was not certain that by continuing the construction, it would form a closed surface. It looked as if the total size would be quite large, so I temporarily abandoned this to look at a smaller model.

I then repeated the construction, but now wrapping three hexagons around a triangular hole. Adding just one more hexagon completed a polyhedron! It had 4 hexagonal faces and 4 triangular holes. If each hexagon was a carbon ring, it would contain 12 atoms. By now I had gained the confidence to cut out the next figure in one piece, This was quickly and easily fixed with tape to form a 14 sided polyhedron. This had 8 hexagonal faces, 6 square holes, and of course could contain 24 atoms.

Going back to the model with pentagonal holes, I tried to visualise the total size; It was not easy! I suddenly remembered that I had seen this before in a maths book. I was sure that I had the book at home, but could not wait and made a quick dash to the library. After a short search, I found the book. It was 'Mathematical Recreations and Essays' by W.W. Rouse Ball. This contained a chapter on Polyhedra, and much information on Eulers rules. It also contained a plate showing a complete set of polyhedra. The three models that I was trying to construct were all there. I rushed back to my office, and competed the one with pentagonal holes. It had 20 hexagonal faces, and 12 pentagonal holes. If the hexagons were carbon rings, the structure would contain 60 atoms.

I now had three models, C12, C24 and C60. They certainly looked pretty, but I had grave doubts about the angles to which the bonds were stretched. I had no idea whether this was possible. However, one thing was immediately obvious. In all three models each hexagon was joined to three others in a symmetric pattern, so all the carbon rings could retain their Kekulé structure of alternate single and double bonds. As all the bonds are satisfied, this structure would be extremely stable. Of the three polyhedra, C60 seemed to be the most probable. Certainly it was the most beautiful.

I went home that night feeling very pleased with myself, but was soon overwhelmed by doubts. How could I justify this extraordinary structure? There was so little evidence! I resolved to say nothing until I had the chance to give the matter more thought.

The next morning I sat looking at the models for some time, almost mesmerised by the perfect symmetry. I had other work to do, but I decided to make just one more model. The C60 model was a bit of a mess, mainly because, as I was building it, I was not sure where I was going until it was finished. Now I could cut larger arrays of hexagons in one piece, so that there were fewer parts to assemble. I was just half way through this construction when John Simmons walked through the door.

He sat in the chair opposite my desk, and we chatted for some minutes before he noticed the models on my desk. I just knew that he would be unable to resist asking me what I was doing. I was not ready for this, but I had no choice. I launched into a rather hesitant explanation. John

remained remarkably quiet, and I began to wonder whether he was listening. When I had finished, he looked very sad, and launched into a very scathing analysis of what I had done. When I tried to defend my proposal, he did not even appear to hear what I was saying. I did not take in all that he said, most of it washed over my head, but I did get the gist of it. It seemed that he could not accept the possibility, that a new form of carbon had gone unobserved for so long. The host of famous people who had studied carbon, from Bragg and Bernel, down to George Bacon and Rosalind Franklin; how could they have missed such a structure. Without any further discussion he left my office, leaving the words 'fanciful nonsense' ringing in my ears. That seemed to be his final word, we never talked about it again, despite my many attempts to engage his interest.

I was left completely squashed and demoralised. I sat and brooded for a long time, wondering what to do next. The easy option was to complete the C60 model that I had started. This I found to be a very satisfying and calming experience. It was also an act of defiance! I then placed the three models in a prominent position on my desk, and went home.

As the days went by, I described these new structures to many colleagues who wandered into my office. Some, I think, did not understand the significance of these models, but most, I am sure, thought I was crazy.

[In 1985, a letter to the journal 'Nature' from H.W. Kroto et al of Rice university in the USA, described experiments which vaporised carbon with a laser beam. This produced stable clusters of 60 carbon atoms. Their speculative suggestion for this structure, was a polyhedron with 20 hexagonal, and 12 pentagonal faces. (Sounds familiar) At that time they had no proof!

However, it was later verified that this model was correct. Subsequently, C60 was found in many different forms of Carbon.

In 1999, Peter Thrower, who by then was Professor of Physics at Pen. State University in the USA, and the editor of the Carbon Journal, acknowledged my work on C60 in an editorial.]

Back to the Routine

I soon forgot about C60, and the humiliation that I had received. It was time to get back to the routine measurements, that I had neglected for far too long. However, my relationship with John Simmons was never quite the same again.

In the early 1970's Harwell was running down the 'Nuclear' work, and pushing into the new ground of 'diversification'. Our Group was no exception, with many members leaving, or transferred to other work. Indeed, at the end of 1972, my work was terminated, and I was moved to another group. As I had no sympathy for the objectives of this new work, I decided to leave Harwell in 1973.

References

Dimensional Changes in the Irradiated Graphite Lattice

P. R. Goggin, R. W. Henson, A. J. Perks and W.N.Reynolds
Carbon 1964 Vol 1, pp. 189-200 .

The Scattering of Long Wavelength Neutrons by Defects in Neutron Irradiated Graphite

D. G. Martin and R. W. Henson
Philosophical Magazine Vol 9, No. 100, p. 659   April 1964

The Effect of Irradiation Temperature on the Scattering of Long Wavelength Neutrons by Defects in Neutron Irradiated Graphite

D. G. Martin and R. W. Henson   Carbon 1967 Vol. 5, pp. 313-314

Lattice Parameter Changes in Irradiated Graphite

R. W. Henson and W. N. Reynolds   Carbon 1965 Vol. 3, pp. 277-287

Lattice Parameter and Dimensional Changes in Graphite Irradiated between 300 and 1350 °C

R. W. Henson, A. J. Perks and J. H. W. Simmons   Carbon 1968 Vol. 6, pp. 789-806

A Simple Theory of the Thermal Expansion and Dimensional Changes of Polycrystalline Graphite

R. W. Henson AERE - R 6652 1970

C60 - Buckminsterfullerene

H. W. Kroto, J. R. Heath, S. C. O'Brian, R. F. Curl and R. E. Smalley

Nature Vol. 318 14th November 1985

Editorial   Dr. P. A. Thrower   Carbon Vol. 37 No. 11, pp. 1677-1678 1999

Radiation Damage in Graphite   J. H. W. Simmons   Pergamon Press 1965

Physical Properties of Graphite   W. N. Reynolds   Elsevier 1968