# A Safety Assessment of the Use of Graphite in Nuclear Reactors Licensed by the U.S. NRC

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### ABSTRACT

This report reviews existing literature and knowledge on graphite burning and on stored energy accumulation and releases in order to assess what role, if any, a stored energy release can have in initiating or contributing to hypothetical graphite burning scenarios in research reactors. It also addresses the question of graphite ignition and self-sustained combustion in the event of a loss-of-coolant accident (LOCA).

The conditions necessary to initiate and maintain graphite burning are summarized and discussed. From analyses of existing information it is concluded that only stored energy accumulations and releases below the burning temperature (650°C) are pertinent. After reviewing the existing knowledge on stored energy it is possible to show that stored energy releases do not occur spontaneously, and that the maximum stored energy that can be released from any reactor containing graphite is a very small fraction of the energy produced during the first few minutes of a burning incident.

The Windscale and Chernobyl accidents are summarized and reviewed. It is shown that there is no evidence from the Chernobyl event that stored energy releases played a role either initiating or contributing to this accident. An improperly controlled process of annealing the graphite at Windscale with nuclear heat resulted in damage to the fuel elements that initiated fuel burning which resulted in a graphite fire. Stored energy releases did not initiate or contribute to this accident either.

The conclusions from these analyses are that the potential to initiate or maintain a graphite burning incident is essentially independent of the stored energy in the graphite, and depends on other factors that are unique for each research reactor and for Fort St. Vrain. In order to have self-sustained rapid graphite oxidation in any of these reactors, certain necessary conditions of geometry, temperature, oxygen supply, reaction product removal, and a favorable heat balance must be maintained. There is no new evidence associated with either the Windscale Accident or the Chernobyl Accident that indicates a credible potential for a graphite burning accident in any of the reactors considered in this review.

Research reactors as used herein means research, test, and training reactors.

### INTRODUCTION

On September 3, 1986 the NRC published in the Federal Register [51FR3134, 1986] a notice of receipt of a petition for rule making filed by The Committee to Bridge The Gap to consider the subject of graphite fires in U.S. research nuclear reactors. Under contract with the NRC staff, Brookhaven National Laboratory staff with past experience in safety evaluation of graphite burning and stored energy releases initiated a reevaluation of graphite burning and stored energy information. The objective of this evaluation was to develop an analysis of the potential role of stored energy releases in initiating or contributing to graphite burning scenarios, as well as an analyses of graphite ignition and self-sustained combustion in the event of a LOCA accident.

The 1986 accident at Chernobyl motivated studies describing the causes for the accident. As a result of this new information, BNL has undertaken a reevaluation of the Windscale Accident, graphite burning studies, and stored energy information that might be relevant to hypothetical graphite burning scenarios in nuclear reactors.

Prior to a detailed analysis of the Windscale Accident, the British mistakenly assumed that the accident might have been initiated by a stored energy release that took place during the anneal of the reactor. Subsequent work by both the team at Brookhaven National Laboratory and the British showed that this was not true, and that the accident was triggered by an uranium fire. In the Prime Minister's report to Parliament, [Penney, 1957], the following statement was made,

"...the most likely cause of the accident was the combined effect of the rapid (nuclear) heating and the high temperature reached by the fuel elements in the lower front part of the pile. In all probability, one or more end caps of the cans of fuel elements were pushed off, and uranium exposed."

As a result of the extensive full scale work carried out at BNL, a great deal of detailed information was developed on the factors affecting both the burning of graphite and the stored energy releases that occurred during anneals [Schweitzer, 1962c; Kosiba, 1953].

### 2. GRAPHITE BURNING

For reasons that are well understood, graphite is considerably more difficult to burn than is coal, coke, or charcoal. Graphite has a much higher thermal conductivity than have coals, cokes or charcoals, making it easier to dissipate the heat produced by the burning and consequently making it more difficult to keep the graphite hot. Concomitantly, coals, cokes and charcoals develop a porous white ash on the burning surfaces which greatly reduces radiation heat losses while simultaneously allowing air to reach the carbon surfaces and maintain the burning. In addition, coals, cokes and charcoals are heavily loaded with impurities which catalyze the oxidation processes. Nuclear graphite is one of the purest substances produced in massive quantities.

The literature on the oxidation of graphite under a very wide range of conditions is extensive. Effects of temperature, radiation, impurities, porosity, etc., have been studied in great detail for many different types of graphites and carbons [Nightingale, 1962]. This information served as a foundation for the full scale detailed studies on graphite burning accidents in air-cooled reactors initiated and completed at Brookhaven National Laboratory [Schweitzer, 1962a-f]. After British experimenters at Harwell confirmed the results obtained at BNL [Lewis, 1963] there appeared to be no new conclusions from additional work in this field. The aspects of the work pertinent to evaluating the potential for graphite burning accidents are described here in some detail.

Burning, as used here, is defined as self-sustained combustion of graphite. Combustion is defined as rapid oxidation of graphite at high temperatures. Self-sustained combustion produces enough heat to maintain the reacting species at a fixed temperature or is sufficient to increase the temperature under actual conditions where heat can be lost by conduction, convection, and radiation. In the case where the temperature of the reaction increases, the temperature will continue to rise until the rate of heat loss is just equal to the rate of heat production. Sustained combustion is distinguished from self-sustained combustion when, in the first case, the combustion is sustained by a heat source other than the graphite oxygen reactions (e.g., decay heat from reactor fuel).

Early attempts to model the events at Windscale [Robinson, 1961; Nairn, 1961] were followed by the BNL work described here.

Some 50 experiments on graphite burning and oxidation were carried out in 10-foot long graphite channels at temperatures from 600°C to above 800°C. To obtain a lower bound on the minimum temperature at which burning could occur, the experiments were specifically designed to minimize heat losses from radiation, conduction, and convection.

The objectives of the full scale channel experiments were to determine under what conditions burning might initiate in the Brookhaven Graphite Research Reactor (BGRR) and how it could be controlled if it did start. Channels 10-feet long were machined from the standard 4 in. x 4 in. blocks of AGOT graphite used in the original construction. The internal diameter of the BGRR channel was 2.63 inches. Experiments were also carried out on channel diameters of one to three inches on 10-foot long test channels in order to obtain generic information. The full length of the channels was heated by a temperature controlled furnace and was insulated from conductive heat losses. At intervals along the length there were penetrations in the furnace through which thermocouples used to read the temperature of the graphite and air were introduced, and from which air and air combustion products were sampled. A preheater at the inlet of the graphite channel was used to adjust the air to the desired temperature. The volume of air was controlled and monitored by flow meters to allow flow measurements in both laminar and turbulent flow conditions.

<sup>2.</sup> Trade name for nuclear graphite used in the BGRR.

In a typical experimental run the graphite was first heated to a preselected temperature. The external heaters were kept on to minimize heat losses by conduction and radiation. The temperature changes along the graphite channel were then measured for each flow rate as a function of time with the heaters kept on. It was observed that below  $675^{\circ}\text{C}$  it was not possible to obtain temperature rises along the channel if the heat transfer coefficient (h) was greater than  $10^{-4}$  cal/cm-sec-°C. Below  $650^{\circ}\text{C}$  it was not possible to get large temperature rises along the channel with  $30^{\circ}\text{C}$  inlet air temperatures at any flow rate. For h values lower than  $10^{-4}$  cal/cm-sec-°C maximum temperature rises were  $0-50^{\circ}\text{C}$  and remained essentially constant for long periods of time (five hours). For h values greater than  $10^{-4}$  cal/cm-sec-°C the full length of the channel was cooled rapidly.

There were two chemical reactions occurring along channels. At low temperatures the reaction C +  $0_2$  to form  $CO_2$  predominated. As the temperature increased along the channel CO formed either directly at the surface of the channel or by the reaction  $CO_2$  + C. At temperatures above 700°C, CO reacts in the gaseous phase to form  $CO_2$  with accompaniment of a visible flame. It was observed that the unstable conditions which were accompanied by large and rapid increases in temperature involved the gas phase reaction  $CO + O_2$  and occurred only for h values below  $10^{-4}$  cal/cm-sec-°C below 750°C. Temperature rises associated with the formation of  $CO_2$  from C +  $O_2$  were smaller than those due to  $CO + O_2$  and decreased with time. They too occurred at h values below  $10^{-4}$  cal/cm-sec-°C.

In a channel which was held above 650°C there was an entrance region running some distance down the channel which was always cooled. A position was reached where the heat lost to the flowing gas and the heat lost by radial conduction through the graphite was exactly equal to the heat generated by the oxidation of the graphite and of the CO. This position remained essentially constant with time. Beyond this point rapid oxidation of graphite occurred with the accompaniment of a flame (due to the CO-O gas phase reaction). Under conditions of burning, the phenomena were essentially independent of the bulk graphite chemical reactivity. Rate controlling reactions during burning were determined by surface mass transport of reactants and products.

The experiments were used to develop an equation which expressed the length of channel that can be cooled as a function of temperature, flow rate (heat transfer coefficient), diameter and reactivity of the graphite. It was found that the maximum temperature at which thermal equilibrium (between heat generated by graphite oxidation and heat removed by the air stream) will occur in a channel can be predicted from the heat transfer coefficient, the energy of activation and a single value of the graphite reactivity at any temperature. Above this maximum temperature the total length of channel is unstable and graphite will burn. The studies show that the bounding conditions needed to initiate burning are:

- 1. Graphite must be heated to at least 650°C.
- 2. This temperature must be maintained either by the heat of combustion or some outside energy source.

- 3. There must be an adequate supply of oxidant (air or oxygen).
- 4. The gaseous source of oxidant must flow at a rate capable of removing gaseous reaction products without excessive cooling of the graphite surface.
- 5. In the case of a channel cooled by air these conditions can be met. However, where such a configuration is not built into the structure it is necessary for a geometry to develop to maintain an adequate flow of oxidant and removal of the combustion products from the reacting surface. Otherwise, the reaction ceases.

To illustrate how difficult it is to "burn" graphite the following was excerpted from a report by Woodruff and Bogert [Reich, 1986]<sup>3</sup>. These tests were carried out in a search for methods for extending the useful life of the N-Reactor. (The following is quoted directly from text of the report.):

"Dry Burning Test: Three pieces of graphite were weighed and stacked together as indicated in Figure 1. Grafoil and carbon felt were placed under and around the blocks. This wrapping material was used as thermal insulation to hold heat in the blocks, and as a buffer to prevent catalysis by contact with the stainless steel tank used to contain the test. Thermocouples were placed at 5 locations in the blocks to monitor temperatures through the test. Two oxyacetylene torches delivering a combined heat output of approximately 2.7 x 10<sup>5</sup> BTU/Hr. through rosebud nozzles were positioned about 2 inches above the graphite. Oxygen flow rates to the torches were

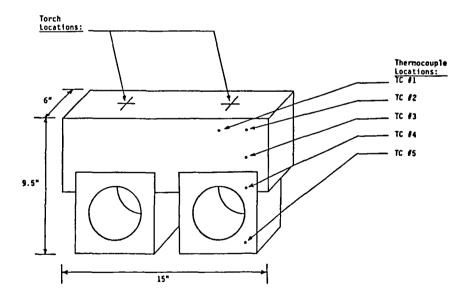


Figure 1. Graphite burn configuration.

<sup>3.</sup> The receipt of this report from Mr. W. Quapp of United Nuclear Corporation, Inc. is gratefully acknowledged.

adjusted to produce nearly neutral flames. Still photographs and a video tape were made to visually record the test.

"Five minutes after ignition, the surface of the top block in regions directly below the torches was glowing yellow-white at an estimated temperature of 1832°F (1000°C).

"Twenty-five minutes after ignition, the lower blocks were also red over their entire surface. Block temperatures continued to rise at rates of a few degrees centigrade per minute until fuel to the torch over the thermocouples was shut off 57 minutes into the test. The peak recorded temperature for thermocouple #1 was 2300°F (1260°C). Other temperatures appear in Table 1. Using an optical pyrometer, the blocks maximum surface temperature was estimated to be approximately 3000°F (1650°C) directly under the torches.

TABLE 1: PEAK TEMPERATURE DATA

Thermocouple		Dry Test			
TC	#1	2300°F	(1260°C)		
TC	#2	2140°F	(1170°C)		
TC	#3	1890°F	(1030°C)		
TC	#4	1615°F	( 880°C)		
TC	<b>#</b> 5		( 825°C)		

### FUEL AND BLOCK WEIGHT DATA

Acetylene Consumed:	13.0 1ь	(2.69 x	10 <sup>5</sup> BTU)
Oxygen Consumed:			20.0 1ь
Total Block Weight Lo	ss:		1.314 1ь _
BTU/lb Weight Loss:			$2.05 \times 10^5$

"With the acetylene to one torch shut off, oxygen was being blown onto the hot block at a rate of approximately 0.16 pounds per minute (1.9 cfm). The oxygen alone could not sustain a reaction with the graphite and the region below the nozzle cooled quickly. Sixty-five minutes after starting the test, both torches were removed, and the blocks were allowed to cool. When cool, the blocks were reweighed to determine weight loss.

"In the dry burn test, small craters were formed directly beneath each of the two torches. They are approximately 2 inches in diameter and their bottoms average 3/8 inch below the original graphite level. These craters account for only a small portion of the total weight loss. The remainder of the weight loss is the result of oxidation on the blocks surfaces that were exposed to air.

"In the interface areas where one block rested on top of or beside another, there are no visible signs of oxidation.

### "DISCUSSION:

There is a common perception taken from our experiences with coal and charcoal that when a mass of these fuels achieves a glowing red condition a self-sustaining combustion is underway. Transferring this perception to graphite has led to repeated references to "burning" graphite when in fact a self-sustaining reaction was not in progress. The test sequences described in these tests demonstrate how difficult it can be to achieve conditions for self-sustained combustion of graphite."

### STORED ENERGY

### 3.1 Summary

A review was made of existing literature and knowledge on stored energy accumulation and releases in order to assess what role, if any, a stored energy release can have in initiating or contributing to hypothetical graphite burning scenarios in research reactors.

From analyses of existing information it is concluded that only stored energy accumulations and releases below the burning temperature (650°C) are pertinent. A review of existing information on stored energy has shown that stored energy releases do not occur spontaneously but are initiated by mechanisms that raise the graphite temperature above the irradiation temperature. Moreover, the maximum releasable graphite stored energy that could be produced by combustion from any reactor containing graphite is a very small fraction of the energy produced if graphite burning were to occur.

Conclusions from these analyses are that the potential to initiate or maintain a graphite burning incident is essentially independent of the stored energy in the graphite.

### 3.2 Wigner Energy -- Its Generation and Buildup

From the earliest days of the Manhattan Project, E. P. Wigner [Wigner, 1946] recognized that if graphite was used as a moderator in nuclear reactors used to produce plutonium, "the collision of neutrons with the atoms of any substance placed into the pile (reactor) will cause displacement of these atoms. ... The matter has great scientific interest because pile irradiations should permit the artificial formation of displacements in definite numbers and a study of the effect of these on thermal and electrical conductivity, tensile strength, ductility, etc. as demanded by theory."

The theoretical prediction has been amplified by the work of F. Seitz [Seitz, 1958], the experimental work of Burton [Burton, 1956] and many others. One of the many observed effects of neutron bombardment of graphite in slowing down the fast neutrons produced in fission to thermal energies is the production of large numbers of displaced carbon atoms and vacancies. Many of these displaced atoms of carbon come to rest in between the planes which constitute the structure of the graphite. The rest of the displaced atoms may

either wander back to their equivalent positions in the lattice, or to crystal boundaries. This introduction of new atoms between the planes increases the spacing between the original planes. This can be measured by the increase in the dimensions of the C-axis. This change in C-axis dimensions is reflected by a change in the gross dimensions of the graphite specimen. Distortion of the lattice results in an increased energy of the overall system. This increase in lattice energy is called the Wigner energy or stored energy.

It was recognized that these two effects, dimensional changes and Wigner energy, might prove to be troublesome in the operation of graphite moderated reactors. The total stored energy of the graphite increases with neutron exposure and is a function of the temperature of the exposure, and the energy distribution of the neutrons. The stored energy that can be released is spread over a range of temperatures. It has been shown that when graphite irradiated at moderate temperatures (less than 100°C) is heated above the irradiation temperature some of the stored energy is released as heat when the temperature of the test specimen is raised some 50-100°C above the irradiation temperature. Increases in exposure to fast neutrons increases the total energy stored. Eventually the stored energy which is releasable up to a temperature of 700°C saturates even though the total stored energy can continue to accumulate with increasing exposure. Total stored energy can be determined by combustion of the sample. Stored energy releases also can be measured by differential thermal analysis where the difference in behavior of an unirradiated specimen and an irradiated specimen are compared in a calorimeter by increasing the temperature in a pre-determined manner.

Broad experimental programs were undertaken during the Manhattan Project. This work was followed by basic and applied programs in the late forties and fifties. Much of this early work was presented at the first Geneva Conference on The Peaceful Uses of Atomic Energy held in Geneva in 1954 [Woods, 1956]. By the early fifties it was known that large dimensional expansions take place in reactor graphite structures and that stored energy accumulated. The British decided to control the stored energy of the Windscale reactor by heating up the graphite moderator (annealing). This process was carried out at regular intervals. The Brookhaven graphite gas cooled research reactor (BGRR) was annealed to reduce the dimensional changes (growth) caused by irradiation and to release the stored energy. Prior to carrying out this work considerable experimental work was carried out to determine the rate of growth and the rate of buildup of stored energy as a function of irradiation exposure and temperature of exposure.

A large body of complex literature exists on the accumulation of stored energy at different irradiation temperatures and fast neutron exposures. Much of this work is not pertinent to the problem of how much stored energy can be released below a given temperature. In this report we have analyzed existing information in order to identify the factors needed to determine the quantity of stored energy that can be released below the bounding temperature (650°C) needed to initiate graphite burning.

The energy required to raise graphite from some initial temperature  $T_0$  to some higher temperature,  $T_0$ , is the enthalpy, which is calculated from the integral of the specific heat at constant pressure over the temperature interval of interest [Schick, 1966]. Consider a starting temperature of 30°C, and a final temperature of 650°C, the minimum temperature required for graphite to burn. The energy required to go from 30°C to 650°C is 202 calories per gram. Energies required to reach 650°C from various starting temperatures are shown below:

Starting Temperature (C)	Final Temperature (C)	Enthalpy (cal/g)		
30	650	202		
50	650	195		
150	650	175		
200	650	160		

Observed stored energy accumulation is non-linear, and depends upon irradiation temperatures, levels of exposures to fast neutron fluxes, neutron energy spectra, spatial distribution of the flux, properties of specific graphites, geometries of individual reactors, etc.

At low temperatures and at low exposures, the displaced carbon atoms move into interstitial positions [Kircher, 1964; Schweitzer, 1962a], and the resulting forces between these displaced atoms and planes in the lattice force the lattice apart, leading to expansions that are initially linear with fast neutron exposure. As neutron irradiation continues, the number of simple defects increases until they begin interacting and result in the formation of larger complexes [Schweitzer, 1964b]. Similarly, initial stored energy increases are linear with neutron irradiation, until a dose is eventually reached at which the stored energy tends to saturate.

Figure 2a shows that a sample exposed for 5000 MWd/AT<sup>4</sup> at 30°C has a total stored energy of 620 cal/g, but only 275 cal/g is released in annealing temperatures up to 800°C [Davidson, 1959, in Nightingale, 1962]. Similar results for other exposures and annealing temperatures up to 400°C are shown in Figure 2b [Kinchin, 1956].

Results of calorimetric and heating experiments show that stored energy will not be released until the annealing temperature exceeds the irradiation temperature by some specific amount. This threshold temperature increase has been reported between 50°C to 100°C above irradiation temperatures [Kircher, 1964; Cottrell, 1958; Woods, 1956].

<sup>4.</sup> Units of neutron dosage are reported in different units by different authors. For this report we generally use the conversion one megawatt-day per adjacent ton [MWd/AT] = 3.9 x 10<sup>17</sup> thermal neutrons per square centimeter [nvt(th)]. For data from Kinchin [Kinchin, 1956] and Bridge [Bridge, 1962], we use 1 MWd/AT = 5.56 x 10<sup>17</sup> nvt(th). For these data we were unable to obtain conversion factors for fast neutron flux.

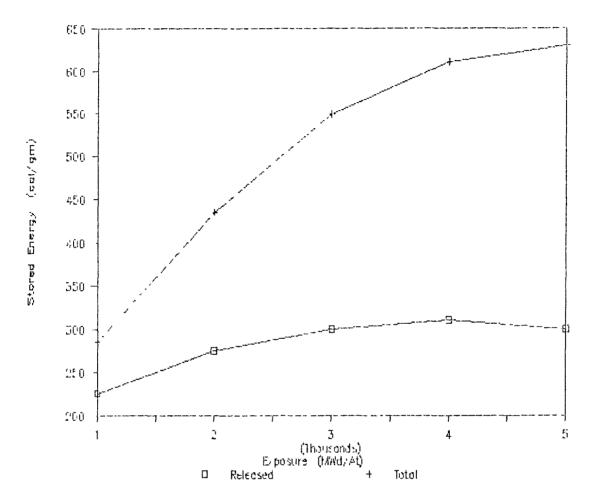


Figure 2a. Total vs released stored energy [Nightingale, 1958],  $T_{irradiation} = 30\,^{\circ}\text{C}$ ,  $T_{anneal} = 800\,^{\circ}\text{C}$ .

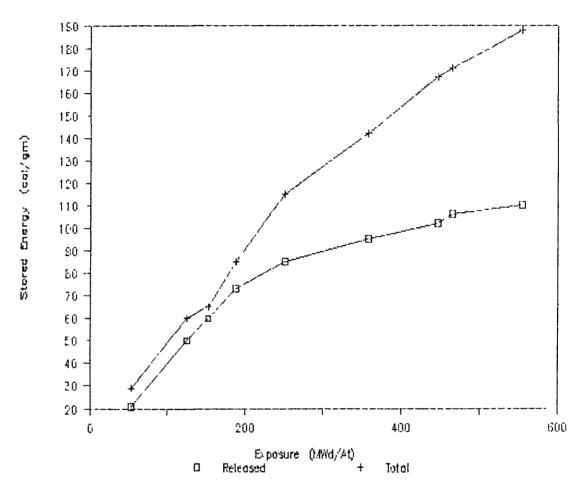


Figure 2b. Total vs released stored energy [Kinchin, 1956].

Tirradiation = 30°C, Tanneal = 400°C.

At irradiation temperatures above about  $150\,^{\circ}\text{C}$  the rate of accumulation of total stored energy is very low [Bridge, 1962; Neubert, 1957; Nightingale, 1958, 1962]. At about  $30\,^{\circ}\text{C}$  and at low total exposures, the total stored energy increases with exposure at a near linear rate of about  $40\,^{\pm}$  10 cal/g per  $100\,^{\circ}\text{MWd/AT}$ . As the exposure continues, the rate of accumulation of total stored energy decreases, and the stored energy that can be released below the minimum bounding temperature to initiate graphite burning (i.e.  $650\,^{\circ}\text{C}$ ) saturates and then appears to decrease. An upper bound on the stored energy that can be released to  $700\,^{\circ}\text{C}$  can be found from existing data. Figure 3 shows this as about  $120\,^{\circ}\text{cal/g}$  for an irradiation in the temperature range of  $35-70\,^{\circ}\text{C}$  at an exposure of  $930\,^{\circ}\text{MWd/AT}$  (equivalent to about  $3.6\,^{\circ}\text{x}$   $10^{20}\,^{\circ}\text{nvt}$  (thermal) [Neubert, 1957]. (This is about  $1/60\,^{\circ}$  the heat of combustion of graphite.)

### 3.3 Stored Energy Releases

A great deal of evidence exists demonstrating that stored energy is released through a series of complex and interactive thermally activated processes. Release of stored energy is generically attributed to the recombination of various interstitial defects with vacancies, or the annealing of the interstitials to edge atoms or other voids in the graphite crystal. Removal of interstitials species from between the graphite planes reduces the stored energy, lattice parameter increases, and other forms of radiation damage.

Existing views of irradiation changes in graphite support the claim that irradiation produces different defects that thermally anneal with different activation energies (i.e. different energies are required to initiate the releases). The type of defects and their respective quantities depend upon the magnitude of the irradiation, the temperature of the irradiation, and whether or not the graphite was subjected to anneals between irradiations. In the latter cases [Schweitzer, 1964a, 1964b] data show that defects interact with each other and that changes that occur during such anneals are very different from the changes observed after a single irradiation.

At any given temperature the stored energy that can be released with time can result from several different processes whose rates decrease as the defects anneal. No evidence exists that stored energy releases are spontaneous. The observation that a  $50-100\,^{\circ}\text{C}$  increase above the irradiation temperature is required to observe finite release rates is consistent with the exponential changes in release rates with reciprocal temperature associated with thermally activated processes.

From our review of the literature on Wigner energy we have compiled data on releaseable stored energy at various combinations of exposures, and irradiation and annealing temperatures and have plotted this information in Figure 4 and Figure 5. In both figures a curve is shown of the amount of energy required for a sample of carbon to go from 100°C to the particular temperature of interest (i.e., the enthalpy between 100°C and some temperature T). Also shown are curves entitled "envelope of releases," which simply delineate an upper bound on stored energy releases found in the technical literature. Data above the enthalpy curve indicate a region where a sample in an adiabatic environment would heat up to the upper intersection of the enthalpy curve and the envelope of releases. Figure 4 shows that the maximum releasable stored

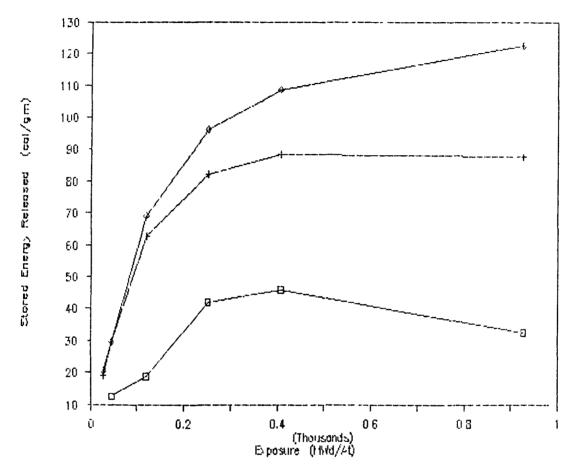


Figure 3. Stored energy released [Neubert, 1957],

Tirradiation = 30-70°C.

 $\Box$  T<sub>anneal</sub> = 250°C + T<sub>anneal</sub> = 500°C  $\triangle$  T<sub>anneal</sub> = 700°C

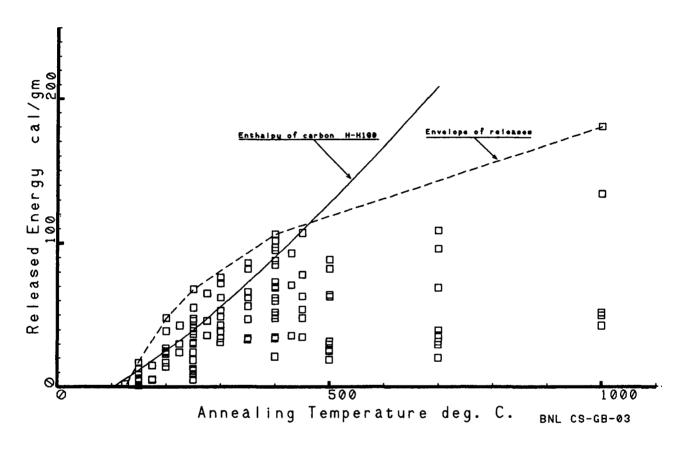


Figure 4. Cumulative energy release; exposure of 500 MWd/AT or less.

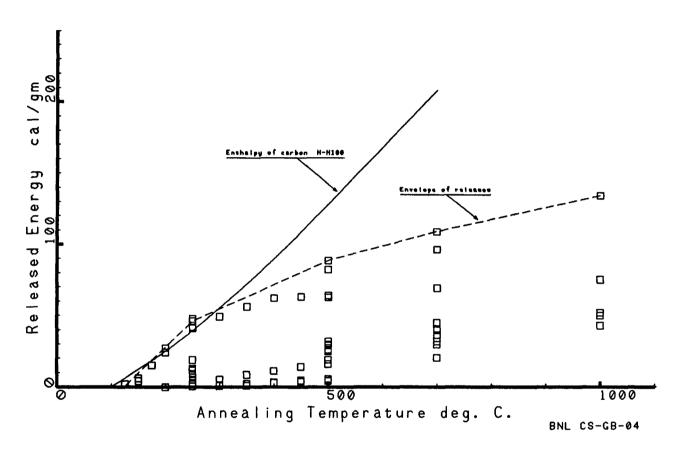


Figure 5. Cumulative energy release; irradiations at 70°C and above.

energy in irradiations below 500 MWd/AT (irrespective of irradiation temperatures) is sufficient to raise the carbon temperature from  $100^{\circ}$ C to about  $450^{\circ}$ C. Figure 5 illustrates the amount of releasable stored energy at exposures in the range 16-5700 MWd/AT [equivalent to about 9 x  $10^{18}$  - 3.2 x  $10^{21}$  nvt (thermal)] and irradiation temperatures greater than  $70^{\circ}$ C. Figure 5 indicates that irradiations at  $70^{\circ}$ C or above (irrespective of exposures) have resulted in temperature rises from  $100^{\circ}$ C to no more than about  $300^{\circ}$ C.

### 3.4 Calculational Approaches

Buildup of stored energy in graphite is a result of the formation of a large number of ill-defined defects each of which can be associated with a stored energy release of unknown specific magnitude, unknown activation energy and unknown temperature range. Since the sum total of these defects determines the accumulated stored energy and since this in turn depends upon the level of the irradiation, the temperature of the irradiation, and the history of irradiations and anneals, BNL does not believe that any of the calculational approaches involved in the past UCLA license renewal hearings can be defended. Other calculational approaches such as the bounding method used by Spinrad [Spinrad, 1986] rely heavily on a number of empirical correlations which involve appreciable uncertainties. These include determining the fraction of energy transferred to carbon atoms by neutron moderation that goes into atomic displacement energy. This must be combined with the fraction of stored energy that self-anneals at various irradiation temperatures. Aside from the direct dependence of this method on measurements showing a great deal of uncertainty, these models cannot account for the non-linear buildup of stored energy, the saturation effects, the temperature dependence of releases, the exposure dependence of releases and the complex consequences of irradiations combined with several anneals.

After review and analyses of existing information on estimating stored energy pertinent to graphite burning scenarios, we believe the approach proposed in this report is consistent with existing data and is acceptable for safety assessments. Total stored energy accumulation has no overall correlation with the stored energy that can be released at temperatures below 650°C. The stored energy that can be released below this temperature saturates at a value that can be bounded from existing knowledge. The dependence of the saturation value of the stored energy released on irradiation temperature can also be bounded from existing data. This approach allows for safety analyses irrespective of the uncertainties in total exposure and total accumulated stored energy.

We emphasize again, that the adiabatic assumption that all the released stored energy goes into heating the graphite is bounding but unrealistic. Under adiabatic conditions where the decay heat is transferred from the nuclear fuel to the graphite, steady increases in the graphite temperature could occur that are much larger than those due to the hypothetical single spike from the release of stored energy.

Because heating graphite to at least 650°C is necessary but not sufficient to initiate burning, the conclusion of these analyses is that the potential to initiate or maintain a graphite burning incident is essentially independent of the stored energy in the graphite.

### 4. THE CHERNOBYL ACCIDENT

BNL has examined recent studies analyzing the Chernobyl accident to determine if any additional information on graphite burning has been developed. The accident summary described here has been taken from Kouts [Kouts, 1986]:

On April 25-26, 1986, "The accident took place during an experiment conducted at the start of a normal reactor shutdown scheduled for routine maintenance. The operating staff had prepared to do what they considered to be a test of some electrical control equipment that was meant to serve a safety purpose."

The objective of the experiment was to see whether the coastdown of the turbine of the nuclear reactor system would supply power long enough to allow for start-up of the standby diesels. The test required that the reactor power had to be reduced to a level (700 MW[th]) just above the value which was known to be low enough to become unstable. In approaching this level, a series of unfortunate operations were carried out in which many safety systems were intentionally by-passed for unknown reasons. In one of these operations, the power level began to decrease rapidly, and fell to an estimated 30 MW(th) before the operator could halt the drop by control rod motion. After the operator had stopped the rapid drop, he managed to achieve some measure of control at 200 MW(th). At this point, the number of control rods in the reactor were far less than regulations permitted.

Further manipulation of the cooling and feed-water systems resulted in other problems eventually leading to a rapid power surge estimated at 300,000 MW(th). Six violations of safety requirements, eventually resulted in a steam explosion that blew off the top of the reactor. The explosion disintegrated the fuel elements, fragmented the graphite, and exposed the graphite and fuel to air. The force of the steam explosion blew pieces of the core and fuel through the roof of the reactor building. A second explosion lifted the cover plate shearing the fuel channels releasing primary system steam pressure to the exterior. Falling hot projectiles ignited asphalt roofing materials causing extensive fires.

Graphite burned for many days supported by asphalt fires and decay heat from the buried fuel. Soviet teams tried to put out the fires by dropping massive amounts of materials from helicopters. The attempts were not successful presumably because the dropped material insulated the hot debris. Eventually liquid nitrogen was used to cool and inert the burning debris. No evidence exists that stored energy in graphite played any role in this accident.

# 5. "ACCIDENT AT WINDSCALE NO. 1 PILE ON 10th OF OCTOBER, 1957"5

Windscale Pile No. 1, was a graphite moderated, air cooled reactor, fueled by natural uranium metal encased in sealed aluminum cans to prevent the uranium from reacting with the components of the air and to contain the gaseous and solid fission products produced in fission. In 1952, the Wigner (stored) energy was found to be releasing on a shutdown of this reactor because the graphite temperature rose above its normal operating temperature when the forced cooling was reduced on reactor shutdown.

To avoid a recurrence of such an incident the Windscale piles were therefore regularly heated above their normal operating temperature to bring about a controlled release of the Wigner energy. The accident developed during the course of one of these controlled releases on October 7th, the day of the start of the Wigner release. Nuclear heating was used, but with cooling essentially shut down to increase the temperature of the graphite above its normal operating temperature. In this instance the first nuclear heating was thought to have inadequately heated enough of the core graphite. To bring about a more uniform temperature throughout the graphite structure the reactor was "pulsed again" but according to the investigators of the accident the rate of increase of nuclear energy input was too rapid, and caused the uranium cladding to break and expose uranium to air. Uranium is an extremely reactive metal. It reacts readily with oxygen, nitrogen, and hydrogen with the release of a large amount of heat. There is also the possibility that the initiating event in this accident may have been the failure of some aluminum clad magnesium lithium cartridges which were in the reactor at the time.

The operator of the reactor was not aware of the cladding failure due to an inadequate number of thermocouples and inadequate radioactive sensing devices at the outlet of the cooling channels. Radioactivity sensing was done at a point some distance from the channel. Since the anneal procedure required allowing the heat to be conducted through the graphite structure by maintaining the cooling shutdown for a day or longer the failed slugs heated adjacent ones and they too failed. Finally after a couple of days during which the temperatures of portions of the reactor were noted to be rising, efforts were made to cool the reactor by admitting air. These efforts failed to cool the hot sections of the reactor. On October 10th a plug in the charging wall of the reactor was removed. The uranium cartridges in the four channels which could be viewed were at red heat. Water was finally used to cool down the reactor after other efforts failed.

There is no evidence that stored energy releases initiated or played a significant role in the evolution of the Windscale accident.

<sup>5.</sup> Title of a report presented to Parliament by the Prime Minister by command of Her Majesty, November 1957. Other sources on this accident -- "Final Report of the [Alexander Fleck] Committee Appointed by the Prime Minister to Make a Technical Evaluation of Information Relating to the Design and Operation of the Windscale Piles and to Review the Factors Involved in the Controlled Release of Wigner Energy." Presented to Parliament by the Prime Minister by command of Her Majesty, July 1958.

### 6. U.S. RESEARCH REACTORS

# 6.1 Criteria for Stored Energy in Graphite

Analyses of existing information indicate that the conditions associated with the initiation and maintenance of graphite burning scenarios are essentially independent of the stored energy in the graphite, irrespective of its value.

As shown in Section 3, if the irradiation temperature of the graphite was 70°C or above, the maximum stored energy releasable below 650°C for any level of irradiation cannot raise the graphite temperature to the minimum value which would be required for initiating a self-sustained burning reaction. For graphite irradiation temperatures below 70°C total exposures of about 500 MWd/AT (3.5 x  $10^{19}$  nvt) are required to continue to heat the graphite from about 100°C to 650°C if an external heat source can raise the graphite from its ambient temperature to 100°C. We have assumed that if the stored energy in the graphite cannot be bounded, any process that heats the graphite to 100°C should be treated as if it heats the graphite to at least 650°C.

The analyses and conclusions on stored energy releases and graphite burning conditions described above provide a meaningful method of categorizing nuclear reactors with respect to stored energy releases below 650°C (the threshold temperature for graphite burning) as follows:

- (1) Any reactor containing graphite in which the lowest irradiation is 70°C or higher, can be excluded from stored energy safety concerns.
- (2) Any reactor in which the graphite is irradiated at temperatures below 70 °C but has received a total fast neutron exposure that is much less than 500 MWd/AT (3.5 x  $10^{19}$  nvt) can be excluded from stored energy safety concerns.
- (3) Those reactors which have graphite that has received more than about 500 MWd/AT (3.5 x 10<sup>19</sup> nvt) of fast neutron irradiation below 70°C without thermal anneals or subsequent reirradiation at higher temperatures would require detailed heat transfer analyses to determine if the graphite were capable of reaching 650°C following an event that raised its ambient temperature to about 100°C. It is important to recognize that even under conditions that allow the graphite to reach 650°C or above, this is not sufficient to initiate burning.

In order to separate reactors into these categories, it is necessary to determine only the total fast neutron exposure reached by graphites irradiated at temperatures below 70°C.

<sup>6.</sup> Estimated fast neutron fluence was converted to MWd/AT using the conversion factor: 7 x 10<sup>16</sup> nvt = 1 MWd/AT.

# 6.2 Stored Energy in Graphite

The significance of stored energy for U.S. research reactors under NRC's licensing authority was assessed in light of criteria in Section 6.1. The information used in the assessment was obtained from Safety Analysis Reports (SAR's) and other readily available data representing the main types of these reactors. The objective of the assessment was to determine if stored energy releases can initiate or significantly contribute to the evolution of graphite burning accidents, and if graphite would play a role in previously reviewed potential accident scenarios.

For the purpose of overall screening of the research reactors, rough estimates of the graphite exposure were made. Only operating research reactors containing graphite and licensed to operate at powers greater than 100 W were included in the survey.

For TRIGA reactors GA Technologies publication GA-4361 [West, 1963] was used to derive a maximum neutron fast flux (above 0.1 MeV) in the side reflector. In addition, an analysis performed by GA Technologies [GA Technologies, 1987] shows, for three out of the four locations where graphite is found in the reactor (i.e., graphite reflectors in the top and bottom of the fuel elements and in the radial graphite reflector) that stored energy would not be sufficient to raise the graphite temperature to 650°C. The reason for this is that these locations satisfy, in essence, either criterion 1 or 2 in Section The dummy elements, which are not in every TRIGA reactor, were found to have enough stored energy such that the graphite could reach 650°C if the temperature of the graphite is elevated to at least 120°C. However, no normal or abnormal operation would produce an initiation temperature of 120°C. Even if this temperature were reached, water cooling of the aluminum clad surrounding the graphite would preserve the integrity of the clad and prevent exposure of the graphite. Additional discussion on the significance of stored energy in TRIGA reactors is found in Section 6.3.

The remaining research reactors were reviewed to assess their stored energy accumulation. These reactors are listed in Table 2. Values of fast flux at the graphite were obtained from the licensees. Where licensee data were not available, peak fast neutron flux data for the reactor core compiled by the American Nuclear Society [Burn, 1983] were used, keeping in mind that the neutron flux that could be expected at a graphite reflector located close to the core would be about a factor of 2 to 10 lower. In the case of MTR reactors, the published data on power and fast flux in the ANS compilation were correlated, removing an outlier, to arrive at a flux-to-power conversion factor.

The total neutron exposure in some reactors was available from the licensees in terms of MWd of operation. In those few cases where these data were not directly available they were estimated based on data of first full power operation and reported equivalent days of full power operation for 1983.

From the survey (see Table 2) it appears that four reactors (General Electric, North Carolina State University, University of Lowell, and University of Virginia) have stored energy greater than 500 MWd. However, the

presence of stored energy above the 500 MWd threshold in parts of the reactor graphite is not by itself taken as a safety concern, as discussed in greater detail in the preceding sections of this report and in Section 6.3.

Table 2. Stored energy calculations in graphite for non-TRIGA
Research Reactors

Reactor Identifier	Туре	Power kW	Year	Duty h/yr	Total MWd	Fast Flux n/cmsq/s	Dose nvt	MWd/AT	Irradiated Temperature °C
General Electric Co.	Spec.	1.00E+02			100.0 <sup>†</sup>	5.00E+11	4.3E+19	617	
Westinghouse Electric	Spec.	1.00E+01			-	3.00E+11	-	-	
N. Carolina State U.	Pulstar	1.00E+03			403.0	1.30E+12	4.5E+19	647	
Georgia Inst. Tech.	MTR D20	5.00E+03			708.0	5.00E+10	6.1E+17	9	
M.I.T.	MTR D20	5.00E+03						*	160.00
National Bureau Stds.	MTR D20	2.00E+04			52013.0	2.00E+09	4.5E+17	6	
Cintichem	MTR	5.00E+03	1961	7800	42250.0	2.80E+08	2.0E+17	3	
Ohio State U.	MTR	1.00E+01	1961	200	2.2	2.60E+11	4.9E+18	70	
Purdue U.	MTR	1.00E+01			_		-	~	
Rhode Island	MTR	2.00E+03						*	148.00
U. Lowell	MTR	1.00E+03			140.0	5.00E+12	6.0E+19	864	
U. Missouri (Columbia)	MTR	1.00E+04						*	100.00
U. Missouri (Rolla)	MTR	2.00E+02	1962	62	12.9	4.86E+12	2.7E+19	387	
U. Virginia	MTR	2.00E+03			1702.0	3.50E+12	2.6E+20	3676	
Worcester Poly.	MTR	1.00E+01	1960	100	-		-	_	
Iowa State U.	Argonaut	1.00E+01			-		~	-	
U. Florida	Argonaut	1.00E+02	1959	213	24.9	1.30E+11	2.8E+18	40	
U. Washington	Argonaut	1.00E+02	1967	100	8.3	1.30E+11	9.4E+17	13	

### NOTES:

Year - Year of initial operation at (at least) one half of full power.

Duty - Number of hours of operation per year, reported for 1983.

Total - Total MW days of operation to date.

Fast Flux - Peak fast neutron flux in the core or graphite reflector.

Dose - Product of years of operation, duty, and fast flux. Represents maximum possible dose to any graphite. MWd/AT - Equivalent dose in MWd/AT. Factor 7el6 nvt = 1 MWd/AT.

Irradiated Temperature - Normal maximum operating temperature of exposed graphite.

- - Not significant because of low power.

<sup>† -</sup> The graphite in the General Electric Co. reactor was annealed in 1976 when the reactor fuel container was replaced for a leak in the weld area. Total MWd since that anneal is 44 MWd.

<sup>\* -</sup> Since irradiated temperature is above 70°C stored energy was not estimated.

# 6.3 Graphite Burning

Research reactors which use graphite in or near their cores and are licensed to operate at power levels greater than 100 watts (thermal) were categorized with respect to:

- 1. Quantity and location of graphite in and near the core,
- 2. Geometry,
- Accident conditions considered by the NRC staff in the licensing bases of the reactors.
- 4. Fast neutron flux,
- 5. Normal operating sequence, and
- 6. Graphite irradiation temperatures.

Although present information indicates a great deal of variation in fast flux, operating sequences and graphite temperatures for reactors within a given type, our analyses of existing information shows that these factors are not significant to those factors related to graphite burning. In scenarios that postulate graphite burning, the quantity of graphite that can burn is an important factor in determining the consequences of burning. However, the credibility associated with a postulated burning accident depends upon the existence of all of the conditions necessary for graphite burning, including the capability to heat the graphite to temperatures above 650°C and maintaining this temperature in the presence of much cooler flowing air. In any given reactor, this not only depends upon the original geometry, but also upon the geometry resulting from the accident that allowed the graphite to heat up in the presence of air.

In assessing the potential for graphite burning in the research reactors licensed by NRC, consideration has been given to conditions during normal operation and conditions that may exist following a LOCA. The LOCA was selected as having conditions most likely to result in high temperatures in the fuel and graphite and, therefore, most likely to release the graphite stored energy and to result in conditions with the potential for graphite burning.

All TRIGA reactors operate in water pools. Since graphite does not burn under water, all accidents in which the core and graphite reflector remain submerged will not be subject to graphite burning. GA Technologies [GA Technologies, 1987] has estimated in a response submitted to the NRC on January 28, 1987 that aluminum clad graphite in dummy elements could, under loss of coolant conditions for some of the reactors, reach 770°C and result in melting of the cladding. GA Technologies claims that the hot graphite at 770°C cannot burn because the specific requirements for graphite burning cannot be met since the graphite radiates its energy rapidly and quickly cools to the ambient air temperature. Our assessment of this claim is based on the experiments discussed in Section 2. That is, radiant heat losses to the cooler

surrounding structures coupled with convective cooling by the cooler air surrounding the graphite could cool the graphite and preclude its burning.

Analysis of a LOCA in an Argonaut reactor predicts peak fuel temperatures of about 120°C [Chen, 1981]. This, coupled with the insignificant stored energy of the graphite suggests no change in the conclusions already reached during the evaluation related to license renewal. The likelihood of graphite fires was reviewed in NUREG/CR-2079 [Hawley, 1981].

Reactors with MTR fuel and the PULSTAR reactor have their fuel located in a water pool. In accidents in which the water level in the pool remains above the core top the graphite could not burn. During a LOCA the maximum fuel plate surface temperature for any of these reactors is 500°C and for many it is much lower except for two cases where it has been calculated to reach 510°C and 582°C. In these two cases, however, emergency core cooling spray systems are activated during a LOCA and the actual fuel temperature would be much lower than the calculated fuel temperatures [NUREG 0928, Section 14.1.3, p. 14-3; NUREG 1059, Section 14.1, p. 14-2]. The stored energy is unlikely to raise the temperature to 650°C under non-adiabatic conditions that exist. Also, the graphite will not burn if the conditions to sustain burning are not present. If the fuel plate surface temperature is always less than 500°C, the heat losses from the graphite by radiation to the cooler structures of the pool coupled with convective cooling by the cooler air in contact with the graphite should preclude conditions necessary for graphite burning.

The Safety Analysis Report [GE, 1981] for the General Electric Nuclear Test Reactor was reviewed for potential impacts of graphite stored energy on the safety analysis of the reactor. The loss-of-coolant accident analysis in the report predicts maximum fuel temperatures of 300-320°C depending on assumptions about peaking factors. Such temperatures pose no danger to the aluminum clad fuel. However, there is no indication that the loss in thermal conductivity of irradiated graphite, or the releasable stored energy in the irradiated graphite, have been included in the thermal analysis. The reduced thermal conductivity could in principle lead to higher local graphite temperatures which in turn could result in some stored energy release. Since in this postulated accident the graphite acts as an effective heat sink, the potentially higher graphite temperatures could have an impact on maximum fuel temperatures. Without a numerical analysis accounting for the space dependence of the thermal conductivity, for the time dependence of the rate of energy release, and for the concomitant changes in thermal conductivity of the graphite, it is not possible to estimate the impact of the irradiated graphite on the course of this postulated accident. However, in connection with Amendment No. 9 to the General Electric license, the NRC staff evaluated the consequences of a postulated maximum hypothetical accident which assumed, nonmechanistically, that all of the fuel in the core melted (NRC Safety Evaluation, Section 3.4, dated June 30, 1969). This scenario encompasses any potential impact of degraded thermal properties of irradiated graphite on the consequences of a loss-of-coolant accident. The resulting radiological doses to an individual at the site boundary under the extremely conservative assumptions of the analysis were well below the allowable 10 CFR Part 100 guidelines.

The MTR-D<sub>2</sub>O reactors have the graphite located away from the core, in a cavity with restricted air interchange. In the analysis of loss-of-coolant scenarios of the SAR for the National Bureau of Standards reactor [NRC, 1983c], NRC staff agreed that a LOCA will not result in melting of the fuel. Under such conditions it appears implausible that the graphite could be subjected to temperatures compatible with burning.

# 7. FORT ST. VRAIN - GRAPHITE STORED ENERGY

Fort St. Vrain operates at temperatures that preclude accumulation of stored energy. There are no known problems associated with stored energy in graphite for operating temperatures associated with HTGR's.

### 8. SUMMARY

### 8.1 Graphite Burning

The factors needed to determine whether or not graphite can burn in air are the graphite temperature, the air temperature, the air flow rates, and the ratio of heat lost by all possible mechanisms to the heat produced by the burning reactions [Schweitzer, 1962a-f]. In the absence of adequate air flow, graphite will not burn at any temperature. Rapid graphite oxidation in air removes oxygen and produces CO2 and CO which, along with the residual nitrogen, suffocate the reaction causing the graphite to cool through unavoidable heat loss mechanisms. Self-sustained rapid graphite oxidation cannot occur unless a geometry is maintained that allows the gaseous reaction products to be removed from the surface of the graphite and be replaced by fresh reactant. This necessary gas flow of incoming reactant and outgoing products is intrinsically associated with a heat transfer mechanism. When the incoming air is lower in temperature than the reacting graphite, the flow rate is a deciding factor in determining whether the graphite cools or continues to Experimental studies on graphite burning have shown that for all the geometries tested which involved the conditions of small radiation and conduction heat losses, it was not possible to develop self-sustained rapid oxidation for graphite temperatures below about 650°C when the air temperatures were below the graphite temperature. At both high and low flow rates, the graphite was cooled by heat losses to the gas stream even under conditions where other heat loss mechanisms such as radiation and conduction were negligible.

At temperatures above about 650°C, in realistic geometries where radiation is a major heat loss mechanism, graphite will burn only in a limited range of flow rates of air and only when the air temperatures are high. At low flow rates, inadequate ingress of air restricts burning. At high flow rates, the rate of cooling by the flowing gas can exceed the rate of heat produced by oxidation.

Studies have shown that burning will not occur when there is no mechanism to raise the graphite temperature to about 650°C [Schweitzer, 1962a-f]. If the temperature is raised above 650°C, burning will not occur unless a flow pattern is maintained that provides enough air to sustain combustion but not enough to cause cooling. Since the experiments were designed to minimize all heat losses other than those associated with the air flow, 650°C can be considered a lower bound for burning.

### 8.2 Stored Energy in Graphite

Fast neutron irradiation of graphite results in the development of stored (Wigner) energy. For a research reactor that has accumulated 30 cal/g of graphite after years of operation, this energy corresponds to about 1/250 of the energy released by combustion. Existing data show that for graphite irradiated at temperatures of 30°C or above, the stored energy that can be released at 650°C saturates at a value that is less than 1/30 of the combustion energy.

Analyses of the Windscale Accident and the Chernobyl Accident have shown that stored energy releases were not initiating events nor did they play any significant role in the evolution of the accidents. Although precise details of the buildup and release of stored energy vary with reactor geometry and factors relating to reactor operation, this review and analysis did not uncover any substantiated evidence or credible scenario in which stored energy releases were responsible for an accident leading to graphite burning [Fleck, 1958; Kouts, 1986].

In assessing the role of stored energy releases in graphite burning scenarios only the stored energy released below the burning temperature was considered pertinent. Stored energies released at or above the burning temperature are a small fraction of the energy released by the burning process.

A large volume of literature exists on the accumulation of stored energy at different irradiation temperatures and different fast neutron exposures. Total accumulation of stored energy is a complex phenomenon that depends upon many factors related to reactor geometries, fast flux distributions, graphite properties, reactor operating schedules and other conditions. At irradiation temperatures above about 150°C, the rate of accumulation of total stored energy is very low with negligible releases occurring if the graphite temperature remains below the graphite threshold burning temperature of 650°C. At about 30°C and at low total exposures, the total stored energy increases at a near linear rate of about 40 ± 10 cal/g per 100 MWd/AT [Nightingale, 1962]. As the exposure continues, the rate of accumulation of total stored energy decreases, and the stored energy that can be released below 650°C saturates and then appears to decrease [Nightingale, 1962; Neubert, 1957; Woods, 1956]. From existing data, an upper bound on the stored energy that can be released below 800°C is 280 cal/g if the graphite was irradiated at 30°C. If the graphite was irradiated at 70°C, data indicate that the maximum stored energy releasable below 700°C is about 150 cal/g. The saturation value for an irradiation temperature of 135°C is about 50 cal/g released below 700°C.

Although there appears to be significant differences in the estimates of total accumulated stored energy calculated in the past [Hawley, 1981; NRC, 1983a, 1983b], these values have little relevance to graphite burning conditions. The total stored energy is always greater than, and is not directly proportional to, the stored energy that can be released below the threshold temperature associated with graphite burning. It requires about 200 cal/g of stored energy to raise the graphite temperature from 30°C to 650°C if there are no heat losses. Similarly, it requires about 190 cal/g to raise the graphite temperature from 70°C to 650°C and 180 cal/g to raise it from 130°C to 650°C. The evidence on maximum stored energy releasable below 650°C shows that if graphite is irradiated at 70°C, or above, the maximum energy released below 650°C is not sufficient to raise the temperature to the burning temperature even under the hypothetical conditions of a spontaneous release under totally adiabatic conditions. In an assumed adiabatic LOCA scenario, the decay heat in any nuclear reactor should be the major source for raising graphite temperatures.

The analyses and conclusions on stored energy releases and graphite burning conditions described above provide a meaningful method of categorizing nuclear reactors with respect to stored energy releases below graphite burning temperatures:

- (1) Any reactor containing graphite in which the lowest irradiation temperature is 70°C or higher, can be excluded from stored energy safety concerns.
- (2) Any reactor in which the graphite is irradiated at temperatures below 70°C but has received a total fast neutron exposure that is less than 500 MWd/AT (3.5 x  $10^{19}$  nvt) can be excluded from stored energy safety concerns.
- (3) Those reactors which have graphite that has received more than about 500 MWd/AT (3.5 x 10<sup>19</sup> nvt) of fast neutron irradiation below 70°C without thermal anneals or subsequent re-irradiation at higher temperatures require detailed heat transfer analyses to determine if the graphite is capable of reaching 650°C in an accident that heated it initially to about 100°C. We emphasize again that graphite temperatures exceeding 650°C are necessary but not sufficient conditions to initiate and support burning.

In order to separate reactors into these categories, it is necessary to determine only the total fast neutron exposure reached by graphites irradiated at temperatures below  $70\,^{\circ}\text{C}$ .

One pound of graphite releasing a stored energy of 200 cal/g is equivalent to running a 100-watt light bulb for one hour. Recognizing that such releases cannot occur unless another energy source raises the graphite temperature above its operating temperature, spontaneous stored energy releases cannot be considered credible initiating events for graphite burning phenomena. Since the maximum energy that can be stored below 700°C is about 1/30 of the

combustion energy, the single release of stored energy that might occur during a graphite burning accident is an insignificant portion of the total energy released in the first few minutes of burning reactions. These conclusions are consistent with analyses of both the Windscale and Chernobyl accidents.

### 8.3 Safety Assessment

Consequences of graphite burning accidents depend upon the amount of graphite that can burn, and the inventory of radionuclides that can be released. Both the amounts of graphite and the inventories of radionuclides in the Chernobyl and Windscale reactors were many orders of magnitude greater than in NRC-licensed research reactors operating in the U.S.

Analyses of the actual reactor accidents in which graphite burning occurred and analyses of hypothetical accidents show that some mechanism must lead to either fuel or graphite heatup under conditions where air is available. The review of a number of research reactors representing the various classes or types of research reactors currently licensed to operate in the U.S. (e.g. the TRIGAS, ARGONAUTS, PULSTAR, GE-NTR, MTR-D<sub>2</sub>O, and MTRs) found that under normal operating conditions their design features and/or environments should preclude graphite being heated to a temperature at which burning could be initiated. In addition, under LOCA conditions it was judged to be plausible that the potential for cooling the graphite by passive means (e.g. radiation, conduction, natural convection) also should preclude graphite burning.

### 9. CONCLUSIONS

After review and analyses of existing information on graphite burning, stored energy accumulations and releases, and causes of the Windscale and Chernobyl accidents, we have concluded that the above phenomena are sufficiently well understood to allow the following evaluations of U.S. research reactors and Fort St. Vrain.

The conclusions of these analyses are that the potential to initiate or maintain a graphite burning incident is essentially independent of the stored energy in the graphite and depends on other factors that are unique for each research reactor and for Fort St. Vrain. However, in order to have self-sustained rapid graphite oxidation in any of these reactors certain necessary conditions of geometry, temperature, oxygen supply, reaction product removal and favorable heat balance must exist.

The reactors considered in this review have all undergone safety evaluations and have been granted operating licenses by the NRC. There is no new evidence associated with the analyses of either the Windscale Accident or the Chernobyl Accident that indicates a credible potential for a graphite burning accident in any of the reactors considered in this review. Nor is there any new evidence that suggests that detailed case-by-case safety analyses of the role of graphite in NRC licensed reactors are warranted.

# 10. GLOSSARY

BGRR Brookhaven Graphite Research Reactor

BNL Brookhaven National Laboratory

BTU/Hr British Thermal Units per Hour

cal/g Calories per gram

CBG Committee to Bridge the Gap

CO Carbon monoxide

CO<sub>2</sub> Carbon dioxide

FSAR Final Safety Analysis Report

LOCA Loss-of-coolant accident

MWd/AT Megawatt days per adjacent ton

NRC Nuclear Regulatory Commission

nvt(th) Exposure in terms of thermal neutrons per  $cm^2$ 

O<sub>2</sub> Oxygen

SAR Safety Analysis Report

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