B_4C/GRAPHITE PELLETS FOR NUCLEAR REACTOR SHUTDOWN SYSTEMS

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An economical procedure has been developed for producing large quantities of boronated graphite pellets for use in nuclear reactor emergency control systems. The pellets were formed by tumble abrading right cylindrical sections cut from extruded rod stock.

Boronated graphite pellets were selected for this application since boron produces no radioactive products nor is gamma radiation emitted under neutron bombardment. In addition the boronated graphite pellets flow by gravity and are thermally and chemically stable in the reactor environment.

The nominal 7/16" diameter pellets produced met all required physical, mechanical, and chemical specification requirements.

The current material used for emergency control of the graphite moderated Hanford N reactor is a dispersion of natural samarium oxide in 3/8" diameter aluminum oxide (Sm_2O_3/Al_2O_3) ceramic pellets. Samarium oxide was chosen because of its large neutron capture cross section and "assumed" fast burn out. These pellets have been used from start-up of the Hanford N reactor in 1964 until the summer of 1977. The pellets are stored in hoppers above the reactor core. In the event that the control rods cannot be inserted into the core to shut the reactor down, the pellets are dropped into the core to serve the same purpose. The control channels are filled with pellets during routine testing of the emergency control system and the pellets are retrieved and returned to the hoppers. Problems have arisen in the use of these pellets since some pellets have remained in the graphite moderator causing reactivity losses which are more or less permanent since samarium transmutation daughter products have neutron capture cross-sections similar to samarium. Also, samarium and other rare earth elements normally found as impurities in the samarium become extremely radioactive when irradiated causing gross radiation and contamination problems. For example, one Sm_2O_3/Al_2O_3 pellet may read as much as 500 Rem after irradiation in the N reactor.

For this reason it was decided to replace the Sm_2O_3/Al_2O_3 pellets during a convenient reactor outage. Boronated graphite pellets, as natural B in the form of B_4C in graphite, were selected since the effective boron isotope, B^{10}, undergoes the following reaction when bombarded with neutrons:

$$_0N^1 + {}_5B^{10} \longrightarrow {}_3Li^7 + {}_2He^4$$

Thus no radioactive products are formed nor is gamma radiation emitted.

The boronated graphite pellets must be thermally stable when dropped into the reactor core operating at 750°C. They must be shaped such that they will flow and must not wear nor break excessively when dropped. The pellets must, of course, shut the reactor down, be "burnable" to eliminate radioactivity loss if pellets remain in the graphite moderator, and must not become highly radioactive in the event they are inadvertently irradiated.

The boronated graphite pellets were produced from (1) minus 60 mesh nuclear grade B_4C, (2) high purity, low ash petroleum coke based graphite flour, and (3) a coal tar pitch binder.

Previous attempts to mold boron carbide loaded graphite spheres proved uneconomical because of excessive tooling costs. The extreme abrasiveness of the boron carbide in the soft graphite matrix caused rapid punch and die wear. In addition, the large number of pellets required for this application would have necessitated long processing times because of the capacity restrictions of conventional molding equipment.

Alternate methods of achieving a suitable "spherical" form were investigated. The extrusion of rods of boronated graphite had been demonstrated in the prior production of "burnable poison" rods. It was felt that such rods could be reduced to right cylindrical pellets with length equal to diameter and the pellets then either warm tumbled as "green" pellets or tumble abraded as heat treated pellets.

Experiments conducted with small lots of trial material indicated that the baked boronated graphite rod stock could be accurately reduced to the desired pellet configuration at the production rates required. Attempts to form pellets from the green, pitch bonded rod stock were generally unsuccessful because of distortion and structural cracking.

After demonstrating the feasibility of the basic processing procedures, several small test lots were produced. Pellets capable of meeting the proposed specification were obtained using the process procedures developed. The production lot of material was produced by preparing a primary blend of boron carbide and graphite. Cross blending procedures were used to ensure uniform distribution of the boron carbide. The coal tar pitch binder was added and the boronated graphite rods were formed by extrusion. The rods were thermally processed to carbonize the pitch binder and to provide the necessary thermal stability for the service environment. Following the baking operation, the boronated graphite rods were sawed to right cylindrical pellets. The pellets were then dry tumbled to abrade the sharp pellet edges and achieve the final, acceptable quasi-spherical configuration.

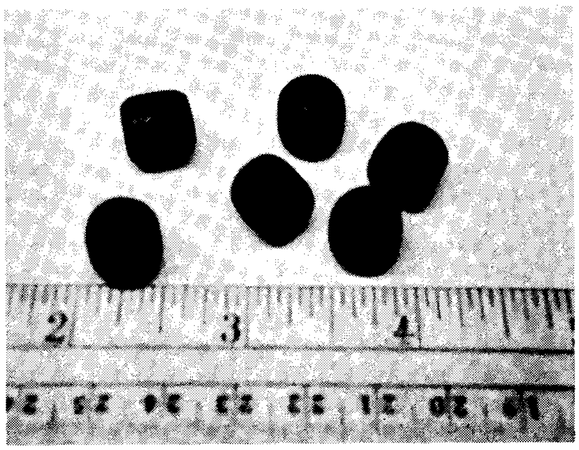
A boron content of 65 to 105 grams of natural boron per foot of 3" ID channel was specified to provide adequate poisoning of the core without prolonging "burnout". A density of not less than 1.50 g/cc was required to assure pellet flow from the hoppers and thru the core. A weight loss specification of no more than 1% when heated to 850°C was imposed to prevent property changes during repetitive testing of the emergency control system. It was required that not more than 1 pellet per thousand be broken and not more than 0.5 wg't percent of the pellets be lost after ten drops of 30 feet to assure that usage will not cause deterioration, since broken pellets tend to slow pellet flow and "dusting" of the pellets might cause contamination problems.

Finished pellets were measured for boron content using a modification of the ASTM method for ferro-borons (ASTM E-31). A boron level of 81 ± 5 grams B/foot of 3" I.D. channel was determined. Physical property tests on the finished pellets results in an apparent density of 1.50 g/cc or greater. Thermal stability was verified by measuring pellet weights before and after thermal treating to 850°C for 12 hours in an inert atmosphere. Wear and breakage were determined by dropping pellets thirty feet into a graphite crucible ten times. Pellet sizing was determined by passing the pellets thru a perforated plate onto a plate with perforations smaller than the pellet dimensions. In all cases the pellets met or exceeded material requirements when MIL-STD-105D inspection standards were applied.

In addition, irradiation and activation analysis of sample pellets by United Nuclear Industries (UNI) personnel indicated that activity and contamination levels were better by a factor of 10^3 when compared to the present Sm_2O_3/Al_2O_3 pellets. Thus activity levels will be measured in millirems rather than in Rems, as at present.

Flow tests carried out at UNI indicated that the boronated graphite pellets will flow from the storage hoppers well within the allowable time limits. However, since they are neither as spherical nor as dense as the current Sm_2O_3/Al_2O_3 pellets, they do not flow as readily in the return troughs.

A photograph of sample pellets is shown below as Figure 1.



These sample pellets were taken from the finished lot of ten tons of pellets which were produced on schedule, met or exceeded all requirements, and provide a satisfactory replacement for the previously used Sm_2O_3/Al_2O_3 material.