Personal Experiences in High Pressure*

By H. Tracy Hall, Distinguished Professor, Brigham Young University, Provo, Utah

On December 16, 1954, I discovered how to make diamonds. Others have claimed prior discovery. (The first claim to synthesis was made 142 years ago!) but there was something unique about mine. My method could be reproduced by others. Moreover, my method grew diamonds so rapidly and in such profusion that commercial production quickly followed. Now, industrial diamond grit is manufactured by every major nation, and the output can be measured in tons.

The way to discovery was not easy, and I should like to tell you a little concerning it and concerning subsequent events.

I always had wanted to work for General Electric, and I had informed my fourth grade school teacher of this desire in 1928. However, upon completion of the Ph.D. degree in chemistry twenty years later, I found this company to be disinterested in acquiring my services. I was persistent in seeking employment, however, and was hired—reluctantly—by the G.E. Research Laboratory in the fall of 1948.

Three years later I became the new addition to a small, loosely-knit group whose assigned task was to synthesize diamonds. There were two facets to this problem. Firstly, thermodynamics indicated that high pressure and high temperature would be needed. Equipment capable of generating the estimated pressure and temperature did not exist. Secondly, the chemistry of diamond formation was not known; i.e., did diamond form directly through polymorphic transformation of graphite, or were other starting materials required? Perchance carbonates, carbon monoxide or other carbon-containing compounds might react with other agents to produce diamond. Catalysts also might be needed.

The problems of the chemistry fell to my lot and for a time I eagerly pursued them. It soon became apparent, however, that the lack of suitable high pressure/temperature equipment was the more immediate barrier to a solution of the diamond-synthesis problem. High-pressure equipment had been partially planned, but was a long time away. The major portion of this equipment was an expensive, large, double-ram hydraulic press which was to provide driving and clamping forces for piston-cylinder type devices not yet designed. This hydraulic press was so expensive that the need for it had to be justified to the company president before purchase was permitted. Delivery time on this equipment was eighteen months.

I was impatient and began to ponder means of generating pressure and temperature that would not require this yet-to-be-delivered press, but could be used in an ancient hydraulic press already on hand. Although equipment design was outside my assignment, I designed, and was allowed to have constructed, a device later named the "Half-Belt." The Half-Belt gave higher steady-state pressures and temperatures than ever before had been achieved simultaneously. But because my colleagues felt negative about it when I proposed to build an improved version, the "Full-Belt" or just plain "Belt," the proposal was rejected, although the cost was less than a thousand dollars. I fretted about this for a time and then decided on a sub-rosa solution. Friends in the machine shop agreed to build the Belt, unofficially, on slack time. This took several months. Ordinarily, it would have taken only a week. The Belt, built of hardened steel, operated so successfully, in my view, that I desired to have the critical components constructed of cemented tungsten carbide. This would allow much higher pressures to be generated. Management, however, would not approve the purchase of the carbide.

There was some confusion at this time as to whom I was responsible to. My former supervisor assumed that I was on loan to Project Superpressure (the diamond project code name). I wasn't certain about this and neither was the manager of Superpressure. (Later it was decided that I belonged to Superpressure). At any rate, having been stopped by the Superpressure people, I appealed to my former supervisor and spoke at a seminar of his group concerning the Belt. He and his group were impressed and shortly thereafter permission was received to buy the carbide components.

With carbide, I soon advanced into pressure-temperature territory far beyond that known to man before. Pressures of 120,000 atmospheres or 1,800,000 pounds per square inch (on today's pressure scale*) simultaneously with temperatures of 1,800 degrees centigrade were maintained for several minutes.

These extreme conditions were thought to be more than sufficient to cause the direct transformation of graphite to diamond, but the sought-for change would not occur. I attempted many hundreds of indirect (mainly "carbon releasing") ap-

**At the time, pressures were thought to exceed 150,000 atmospheres.*

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proaches over a period of about a year but to no avail, and I was becoming discouraged. Then, one wintry morning, I broke open the sample cell after removing it from the Belt. It cleaved near a tantalum disk used to bring in current for resistance heating. My hands began to tremble; my heart beat rapidly; my knees weakened and no longer gave support. My eyes had caught the flashing light from dozens of tiny triangular faces of octahedral crystals that were stuck to the tantalum and I knew that diamonds had finally been made by man. After I had regained my composure, I examined the crystals under a microscope. The largest, about 150 microns across, contained triangular etch and growth pits such as I had observed on natural diamonds. The crystals scratched sapphire and other hard substances, burned in oxygen to give carbon dioxide, and had the density and refractive index of natural diamond. A few days later, an x-ray diffraction pattern unequivocally identified the crystals as diamond.

This first successful experiment contained troilite (FeS) and graphite at a pressure near 70,000 atmospheres and a temperature near 1,600 degrees centigrade. Troilite is a mineral associated with the microscopic diamonds found in the Canyon Diablo meteorite. It now seems certain that the meteoritic diamonds were formed by the transient pressure and temperature generated on impact of the meteorite with the earth.

I was able to repeat this experiment successfully a dozen times in the next two weeks, learning during this period that iron alone could cause graphite to transform to diamond, but that sulfur alone would not. Tantalum also showed a beneficial effect. On Dec. 31, 1954, Hugh Woodbury, a company physicist, successfully duplicated my December 16th experiment and became the first person to duplicate the diamond synthesis claim of another. There had been a long history of chicanery, tomfoolery, bad faith, and downright fraud during the period of diamond-synthesis claims. Consequently, on January 18 and January 19 of 1955, official duplication experiments were carried out under the watchful eyes of company officials. I was not allowed to be present while Hugh Woodbury and Richard Oriani (a company metallurgist), using independent sources of FeS and graphite from those that I had used, each made three runs according to my procedure. I breathed a relieved sigh when diamonds were made in all six runs.

These runs, as well as the original run, were made in my Belt apparatus, which was actuated by the ancient press previously mentioned. This press used water for hydraulic fluid and leaked so badly that rubber footwear, mop, and bucket were standard accessory equipment. In addition, the press's hydraulic lines had been wrapped with rags to reduce the overhead water spray. Historic as this old Watson Stillman Press had now become, it was relegated to obscurity, for officially it was said that the first diamonds were made in the beautiful new double-ram press that now had been delivered.

The News Spreads Fast

Management, thus convinced of the authenticity of my synthesis, held an impressive press release on February 15, 1955. Within the next two days, most U.S. newspapers carried as front page news the story that diamonds had been made at the G.E. Research Laboratory in Schenectady, New York. Company advertising executives were quick to note that the value of this newspaper coverage far exceeded the cost of the Superpressure project. The press release gave no details concerning high-pressure equipment or the method of synthesis. A number of important U.S. scientists voiced objections to company officials for this secrecy. In a move to ward off further adverse comment and establish credibility, the company engaged the services of Nobel Laureate P. W. Bridgman, who wrote an article for *Scientific American* certifying that diamonds indeed had been made at G.E. But the secrecy continued for five more years.

In April of 1955 I decided to leave the company that I had aspired to work for in my youth. There were several reasons for doing this, most compelling among them the continued lack of financial support for the things I wished to do. I had come to "understand" the politics of the expensive double-ram press. After committing itself to this approach, management could not face a cheap alternative. But after synthesizing diamond I expected that money to be no object and could understand no longer. Incidentally, concerning the double-ram press, the two rams were tied together and a Belt was built for use therein.

On September 1, 1955, I assumed new responsibilities as director of research and professor of chemistry at Brigham Young University. Meantime, company officials had informed me that I could not use my Belt apparatus for high pressure/temperature research at the University, and this complication in my plans was reinforced by a secrecy order from the U.S. Department of Commerce.

I made several trips to Washington to confer with Commerce Department officials and some important scientists to determine how I might be able to continue research at high pressure. The "solution" to my problem dawned one day when a man from the Commerce Department said, "Hall, why don't you invent another apparatus?" I didn't appreciate the idea particularly, realizing that this might delay my work for several years. I asked the Commerce man if they would not also place a secrecy order on any apparatus I might invent that had the same pressure/temperature capabilities as the Belt. He said that they would not. I asked for a letter to that effect whereupon he assured me that I did not need a letter.

It would take money to experiment with new apparatus ideas. I approached the National Science Foundation (NSF). They were eager to help, but were cautious in view of the possible proprietary, national, scientific, and personal-interest conflicts that they sensed might exist. My first funds came from the Carnegie Foundation through the assistance of Philip

Abelson, who was very sympathetic to my dilemma. This broke the ice and funds soon also came from NSF. Then I was approached by the Army, Navy, Air Force, Atomic Energy Commission and other organizations proffering more money than I could accept in view of my administrative and teaching duties, which left only minimal time for research. The people who approached me from the government agencies were unanimously against the secrecy order and could not understand how such a thing had come to pass during peacetime. Furthermore, the defense agencies and the AEC had attempted to penetrate the secrecy. They were able to penetrate the Government secrecy, but always were stopped by the proprietary secrecy. Consequently, they heartily encouraged me to invent a new apparatus that would be free and open for all to use. To me, a frustrated inventor in my attempts to get at the heart of the matter, it appeared that the proprietary secrecy hid behind the Government secrecy and vice versa. It required four years of effort by various interests to have the Government secrecy removed, and several months following this for the proprietary secrecy to end. For all these years the secrecy plagued and hindered my efforts.

Secret and Not Secret?

I had many ideas on which to work, but constantly wondered where the dividing line was between what was secret and what was not. Again, I journeyed to Washington to consult with Commerce. They could not enlighten me other than to say that if a new apparatus invention did not infringe the Belt invention, I would have no problems. But they declined to make this judgment, stating that under the circumstances I was the only person who could make the determination and must personally assume the risk of violating the secrecy order (2 years in jail, $10,000 fine).

While I had funds sufficient to pay for the machining of parts, I ran the lathe myself, in secret. I also personally attended to the hardening of the alloy steels and the fabrication, assembly, and testing of the various components and devices. There was no way I could make tungsten carbide parts, and I considered having such parts made outside too risky. However, I managed to obtain 60,000 atmospheres at 2,000 degrees Centigrade in alloy steel devices without carbide. I concluded, though, that these particular devices were too much like the Belt and might violate the secrecy order. It was indeed hard to move away from the Belt idea. I worked a great deal with piston-cylinder devices, with a stepped piston, and with a device called the Black-Hawk Special. For diversion, I tried for extremely high temperatures at modest pressures to 10,000 atmospheres in a sort of "confined" exploding wire device. One day I connected the full 20,000 watts available from the Lab mains to this device and it exploded, spewing molten refractory and metal and depositing them as a ring around the inside of an 8-foot-diameter concrete pipe used for a safety enclosure. The estimated temperature was 60,000 degrees Centigrade.

A Tetrahedral Press

By summer 1957 I had the device that eventually freed me for a continuation of my high-pressure researches. It could do everything that the Belt could and more. I called it the Tetrahedral Press. But never have I had so much anxiety and so many sleepless nights. It all revolved around ascertaining that the tetrahedral press did not infringe on the Belt and thus did not violate the secrecy. I finally decided that it positively did not and submitted a paper on the new apparatus to the Review of Scientific Instruments, filed for a U.S. Patent, and prepared a talk for the spring 1958 meeting of the American Chemical Society in San Francisco. The ACS talk drew a very large crowd and was the starting point for a new type of experience for me. Soon, hundreds of scientists from all over the world were to come to Provo to learn of the high-pressure methods. Over fifty commercial, governmental, and educational organizations within the next few years were to engage me as a consultant. I was constantly called upon to lecture, and the press, radio and T.V. insistently sought me out.

In the midst of this came another traumatic period when a letter from the U.S. Department of Commerce containing a secrecy order on the Tetrahedral Press arrived. The man from Commerce had not honored his word. Included in the secrecy order was a directive that I should inform everyone that knew of the Press that it was now a secret and conveyance of this secret to another was an act subject to the 2-year prison sentence and $10,000 fine. What a problem! How could I possibly inform the thousands of worldwide readers of the *Review of Scientific Instruments* of this? And what of those at the ACS meeting? Again a trip to Washington became necessary. Commerce finally decided that I would only be required to inform those who had personally seen the Tetrahedral Press plus those who had sent written requests for reprints describing the device. At this time, this amounted to over one hundred persons, some of them foreigners. I followed the directive, but confess to having felt rather awkward about it all, particularly in writing to the foreign scientists. Some American scientists, upon receipt of my letter, thought the whole affair horrendous and so informed the Commerce Department. In exasperation, I considered giving up the field of high pressure.

Secrecy Is Lifted

Fortunately this secrecy order lasted for only a few months. The secrecy was lifted in the following manner. A hearing was being held in Washington concerning the secrecy order. Commerce was adamant on maintaining the secrecy; those in opposition had just lost another round in the struggle; and the meeting was being closed when a messenger arrived. At first he was refused admission, but on presenting credentials, he could not be excluded. He was from the Pentagon and had a message from the top. The written message, simple and direct, amounted to the Pentagon's "pulling rank" on Commerce. It was a directive that the secrecy order be lifted immediately on the Tetrahedral Press, the Belt, and the method for synthesizing diamonds. A plea by G.E. for delay to get its patents in order was granted, but secrecy on the Tetrahedral Press was terminated immediately. The secrecy order, in effect, gave several additional years of patent protection (and without disclosure) to the Belt and diamond patents. The patent on the Tetrahedral Press was granted (including all claims as initially filed) on the first office action five months before the patent on the Belt was issued, giving resounding support to my conviction that the Tetrahedral Press did not infringe on the Belt.

The lifting of the secrecy order ushered in a period of tranquility that I had not enjoyed for six years. A great many things of interest have happened in the decade since secrecy ended that I do not have time to tell. Suffice it to say that my activities in high pressure have expanded and continued, and that I have been involved in several interesting discoveries.

Fifteen years ago very little research was being done in high pressure. Today, there are 600 high-pressure laboratories and 1,000 high-pressure researchers, and 1,000 high-pressure papers are published each year. It seems possible that much of the impetus for this expansion stems from that December day of 1954 when I discovered how to make diamonds.