N71-23775

NASA TECHNICAL MEMORANDUM

NASA TM X-64543

PROCESS AND APPARATUS FOR MAKING DIAMONDS

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September 4, 1970

NASA

George C. Marshall Space Flight Center
Marshall Space Flight Center, Alabama

MSFC - Form 3190 (September 1968)

TECHNICAL REPORT STANDARD TITLE PAGE

1. REPORT NO.	2. GOVERNMENT ACCESSION NO.	3. RECIPIENT'S CATALOG NO.
TM X-64543		
4. TITLE AND SUBTITLE Process and Apparatus for Making Diamonds		5. REPORT DATE September 4, 1970 6. PERFORMING ORGANIZATION CODE
7. AUTHOR (S) J. R. Rasquin, Marvin Estes, and David Webb		8. PERFORMING ORGANIZATION REPORT #
9. PERFORMING ORGANIZATION NAME AND ADDRESS George C. Marshall Space Flight Center Marshall Space Flight Center		10. WORK UNIT NO. 11. CONTRACT OR GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS		13. TYPE OF REPORT & PERIOD COVERED Technical Memorandum 14. SPONSORING AGENCY CODE

15. SUPPLEMENTARY NOTES

Prepared by Product Engineering and Process Technology Laboratory, Science and Engineering

16. ABSTRACT

This paper describes a new and unique method of synthesizing diamond grit. The grit is polycrystalline in nature and of sizes typically used for grinding and lapping processes.

EDITOR'S NOTE

17. KEY WORDS	18. DISTRIBUTION STATEMENT
	Unclassified – Unlimited John R Rasquin

19. SECURITY CLASSIF. (of this report)	20. SECURITY CLASSIF. (of this page)	21. NO. OF PAGES	22. PRICE
Unclassified	Unclassified	24	$3.00

MSFC - Form 3292 (May 1969)

TABLE OF CONTENTS

	Page
SUMMARY	1
INTRODUCTION	1
The Production of Shock by Magnetic Induction	2
Experimental Data	6
CONCLUSIONS	14
REFERENCES	16

LIST OF ILLUSTRATIONS

Figure	Title	Page
1.	Diamond making apparatus	3
2.	Current waveform from the magnetic hammer – 3 kV on capacitor bank, 91A/cm, 100 ns/cm sweep time per cm	5
3.	Diamond crystal – largest dimension approximately 0.1 mm	7
4.	Diamond crystal – largest dimension approximately 0.75 mm	8
5.	Diamond crystal – largest dimension approximately 0.38 mm	9
6.	Diamond crystal – largest dimension approximately 0.14 mm	9
7.	Diamond lines for film no. 1	13
8.	Diamond lines for film no. 2	13

LIST OF TABLES

Table	Title	Page
1.	d Values of X-Ray Diffraction Test, Film No. 1	11
2.	d Values of X-Ray Diffraction Test, Film No. 2	12

TECHNICAL MEMORANDUM X-64543

PROCESS AND APPARATUS FOR MAKING DIAMONDS

SUMMARY

This paper describes a new and unique method of synthesizing diamond grit. The grit is polycrystalline in nature and of sizes typically used for grinding and lapping processes.

INTRODUCTION

This experiment is a spinoff from the investigation of intense magnetic fields as used in the manufacture of space vehicles.

The interest in the production of industrial diamond abrasives in the United States and, indeed, the whole world has greatly increased as the level of technology has advanced. The new harder and tougher structural materials being developed are increasingly difficult to machine with conventional abrasives.

Diamond is unique as an abrasive. Its hardness and cutting ability are unrivaled by any other substance. The abrasive comes in many forms: loose; mixed with a carrier, such as oil; or embedded in a carrier, such as bronze. It is used in grinding and polishing wheels or loose as in a buffing or lapping compound.

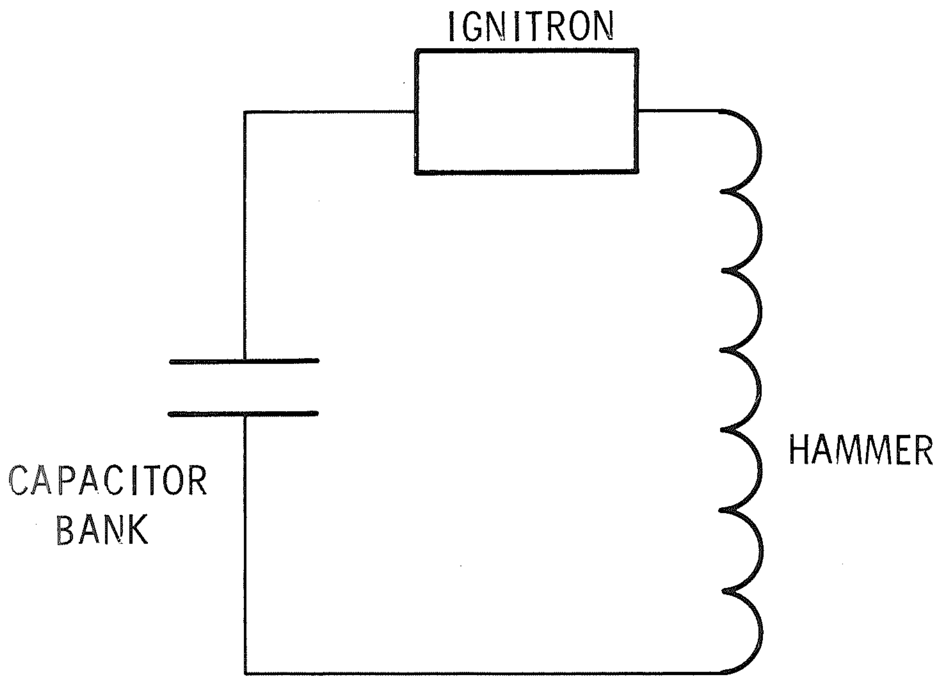
The abrasive has one bad feature; it is expensive. Many ways of producing cheap diamond have been sought down through the years. It was not until this century however that a relatively cheap industrial diamond has been synthesized. The General Electric Company makes industrial diamond by a process [1,2] of extremely high temperatures and pressures and recently has even been able to grow gem quality stones by another method. Recently, also P. S. De Carli and J. C. Jamieson have shown that diamond can be produced by shock induced in graphite by high explosives [3,4]. Allied Chemical Corp., owner of the De Carli patent [5], is presently offering diamond synthesized by this method. The De Carli method specifically uses a shaped charge of explosives to create the shock wave.

This paper describes a novel and inexpensive method of producing shock in graphite to synthesize diamond without using high explosives.

The Production of Shock by Magnetic Induction

The apparatus shown in Figure 1 is made with an exponential horn of solid hardened steel that tapers from a large end to a small end. The small end is 0.6 cm in diameter and the large end approximately 14 cm in diameter. The horn is approximately 30 cm long. A magnetic coil is positioned adjacent to the large end of the horn with a 1.2-cm thick copper disk positioned between the horn and the magnetic coil. A 0.01-cm thick piece of Mylar is used as an electrical insulator between the copper disk and the magnet coil. An anvil block with a small shallow pocket 0.02 to 0.03 cm larger than the small end of the horn is positioned below the small end of the horn so that the small end fits into the pocket. The pocket is filled with loose graphite, and the whole assembly is bolted together with 5-cm thick steel end plates and 1.6-cm diameter steel bolts.

The apparatus shown in Figure 1 was used to produce the shock waves in graphite. This experiment was designed around the magnetic hammer which is shown schematically as follows:



The magnetic hammer is described in a patent [6] awarded to R. J. Schwinghamer and L. E. Foster. The magnetic hammer is powered by a capacitor bank of 360 μF and is capable of being charged to 10 000 V.

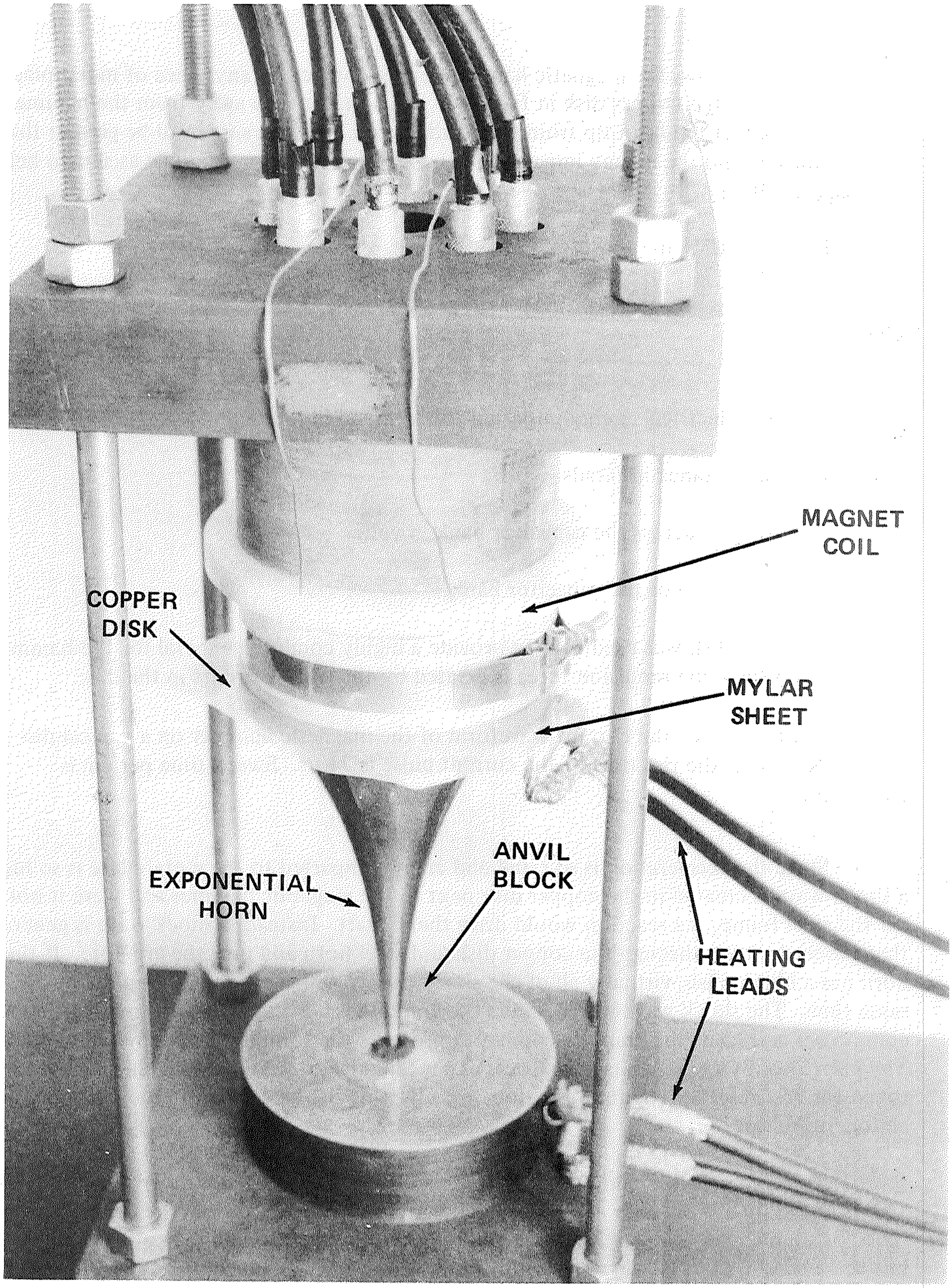


Figure 1. Diamond making apparatus.

By Lenz's Law, the magnetic hammer applies a force on any piece of metal placed near the coil; e.g., the copper disk in Figure 1. Thus the force is away from the hammer. The acceleration of the horn tip from the hammer coil has been shown to be greater than 75 000 cm/sec^2 and the energy imparted to the metal plate is considerable as shown below. (See page 16, item 4.)

$$E = \frac{1}{2} CV^2 n$$

where

E = the electrical energy imparted to the metal in joules

C = capacitance in farads

V = voltage across the capacitor bank in volts

n = efficiency of the capacitor bank.

A copper disk was used here to provide a highly conductive metal for the hammer coil to act on since the repulsion force is created by the eddy currents in the disk.

Figure 2 shows the current waveform of the magnetic hammer on a typical discharge. Note that the rise time of the current pulse is 71 μs. Sweep time per cm is 100 μs/cm.

Since the acceleration is so rapid and energy imparted to the metal plate is so high, a shock wave is created in the copper disk next to the magnetic hammer and were it not for the steel frame, the reaction would drive them apart. Instead, a shock wave is generated that proceeds down through the copper disk and steel horn and into the graphite. If the horn has a high polish, virtually all of the shock energy will arrive at the graphite at the same time. The shock wave which enters the graphite is a high-speed pressure front which compresses and heats the graphite enough to promote the formation of diamond crystals. The horn also, by virtue of its shape, acts as a mechanical amplifier and as such it has to be tuned for best results. The tuning is done by adjusting the length by grinding for maximum action at the tip.

An acoustic wave, which travels at the speed of sound, travels down the horn and arrives at the tip a short time after the shock wave, since the shock wave is faster than the speed of sound. The acoustic wave does not benefit the process and may even be detrimental, but there is no way to eliminate it at this time.

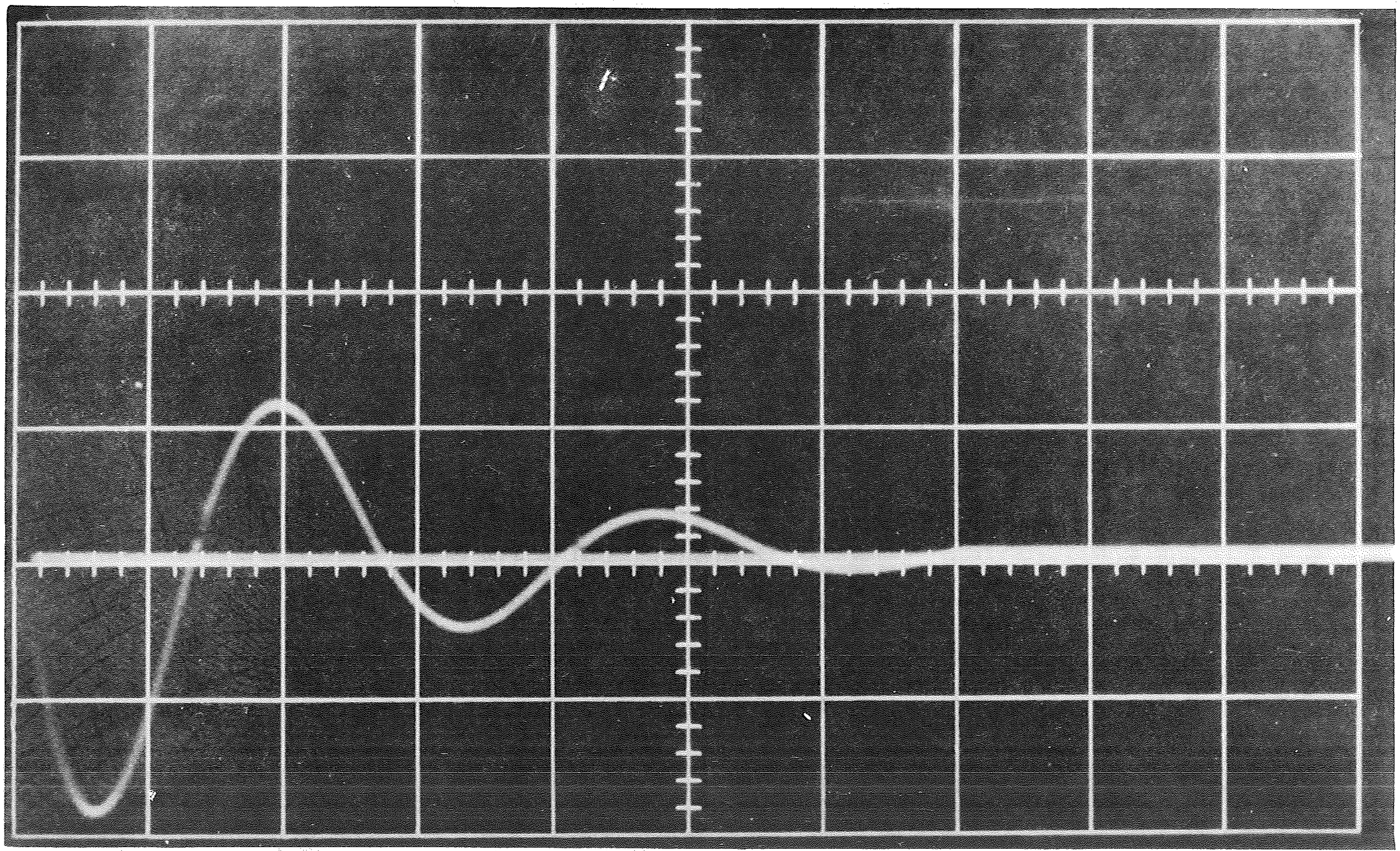


Figure 2. Current waveform from the magnetic hammer – 3 kV on capacitor bank, 91A/cm, 100 ns/cm sweep time per cm.

This laboratory has no instrumentation to measure such high pressures, but as is shown by De Carli, the pressure has to be at least 1×10^{10} N/m^2 for the creation of diamonds. See References 3 and 4 for further information.

The exponential horn has a curvature described by the equation $y = ce^{ax}$ where x and y are points on a plane defined by the two coordinate axes, a and c are constants, and e is the transcendental number (2.7183) used as the base of natural logarithms. This curve is then made into a figure of rotation about the x axis.

Experimental Data

Experimental shots were made at capacitor bank energy levels of 180, 720, 1620, 2880 and 4500 joules. Laboratory experiments indicate that transfer efficiency from this capacitor bank to a work piece is approximately 2 percent. Using this figure as the value for n in the equation on page 4, the energy available for shock creation becomes 3.6, 14.4, 32.4, 57.6 and 90 joules.

The diamond material formed is so small that it is difficult to identify with a microscope; however, the crystals shown in Figures 3 through 6 are virtually certain to be the diamonds. The reason for this is that only a tiny amount of the graphite is changed into diamond on each shot and the only method this laboratory had to separate the diamonds out of the graphite was to reflux the contents of the pocket in a mixture of concentrated sulfuric and nitric acid which oxidizes most of the graphite. The solution was then diluted with distilled water and filtered through high grade, ashless, Whatman filter paper. Millipore manufactures a filter paper more suited to this process but none of this material was on hand at this time. The Whatman filter paper was dried and then slowly decomposed. The remaining residue was mixed with neutral, filtered, Canadian balsam. X-ray patterns were made, each yielding the following information:

Parameters for Pattern No. 1 (May 22, 1970)

General Electric XRO-6 X-ray machine

X-ray diffractor powder camera with copper anode

25 kVp, 20 mA

Wavelength: 1.5405 Å

Time of file exposure: 4 hr

Type of film: Kodak F0034DA, medical, no screen

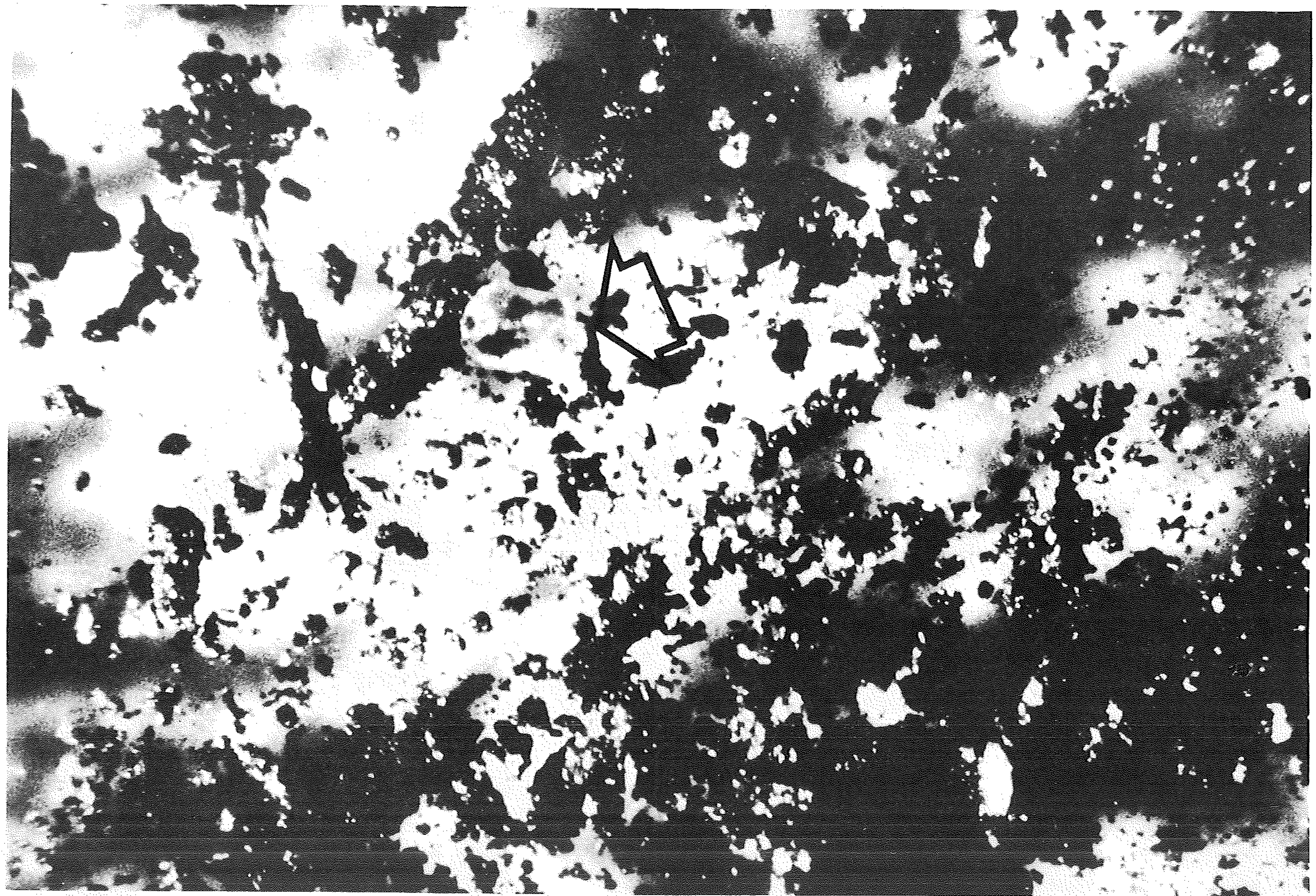


Figure 3. Diamond crystal – largest dimension approximately 0.1 mm.

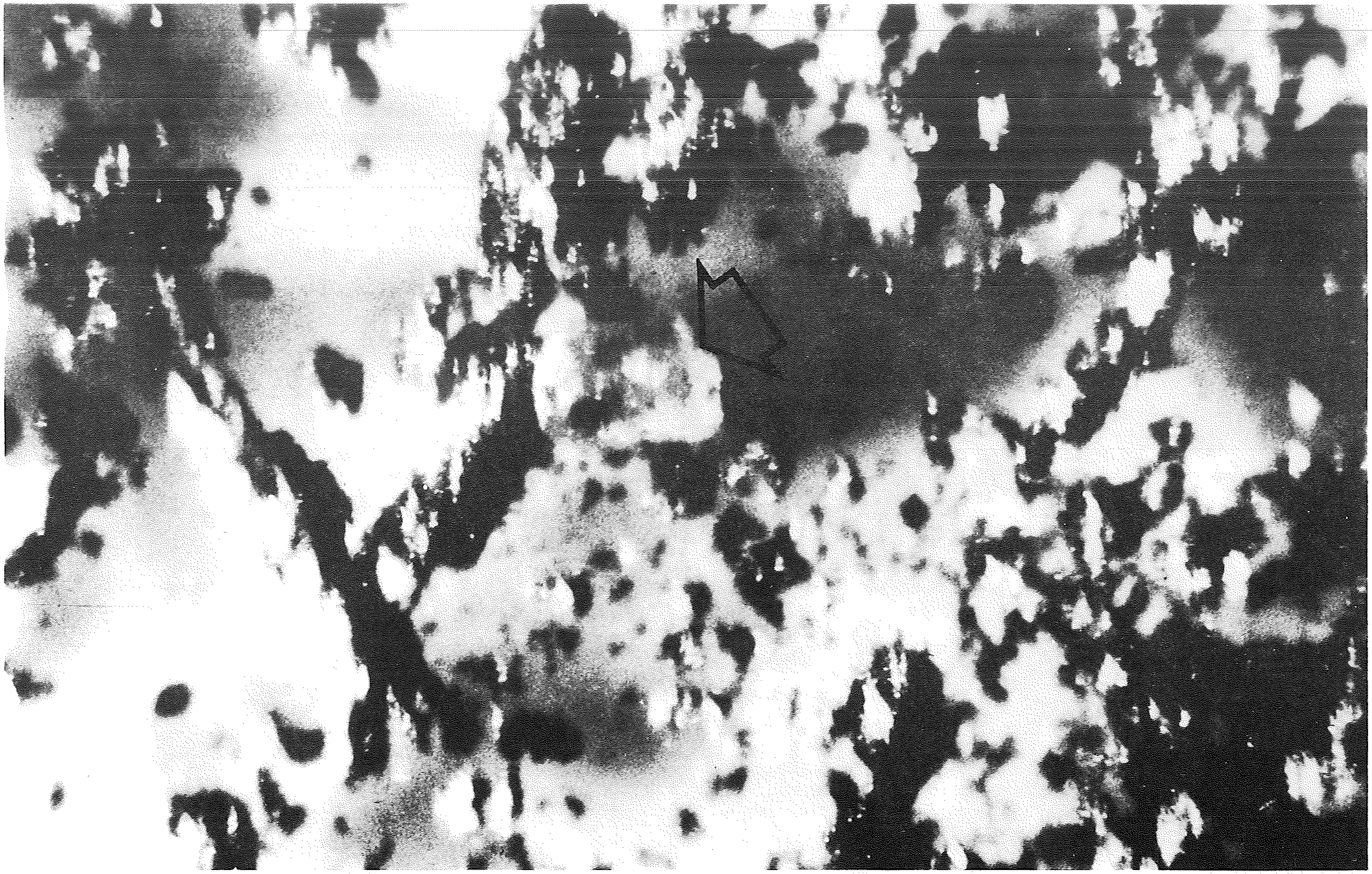


Figure 4. Diamond crystal – largest dimension approximately 0.75 mm.

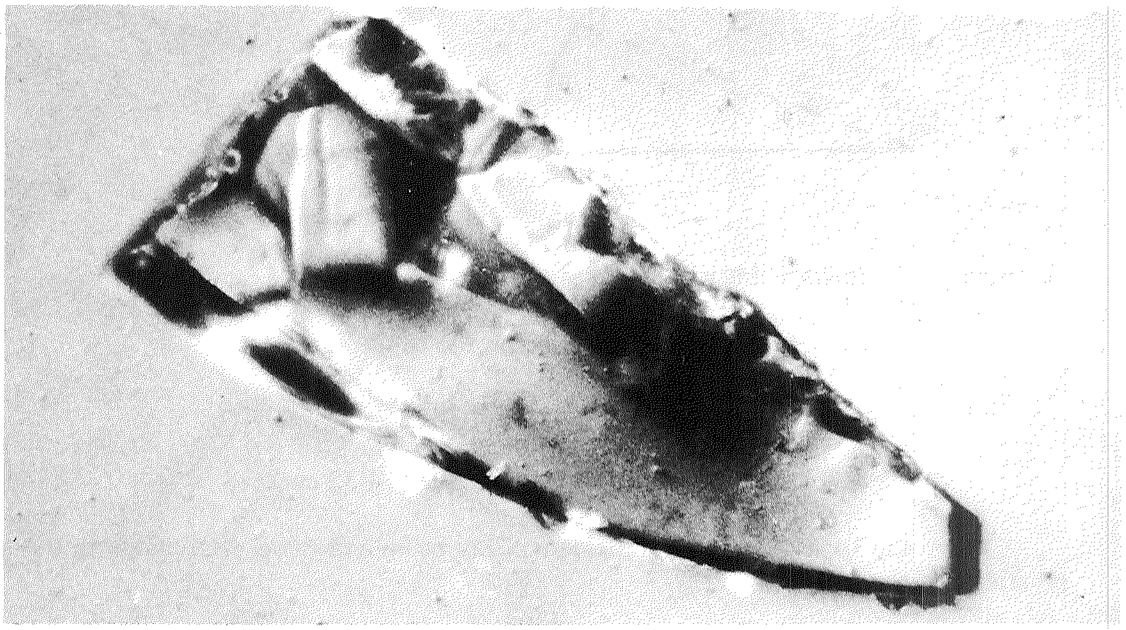


Figure 5. Diamond crystal – largest dimension approximately 0.38 mm.

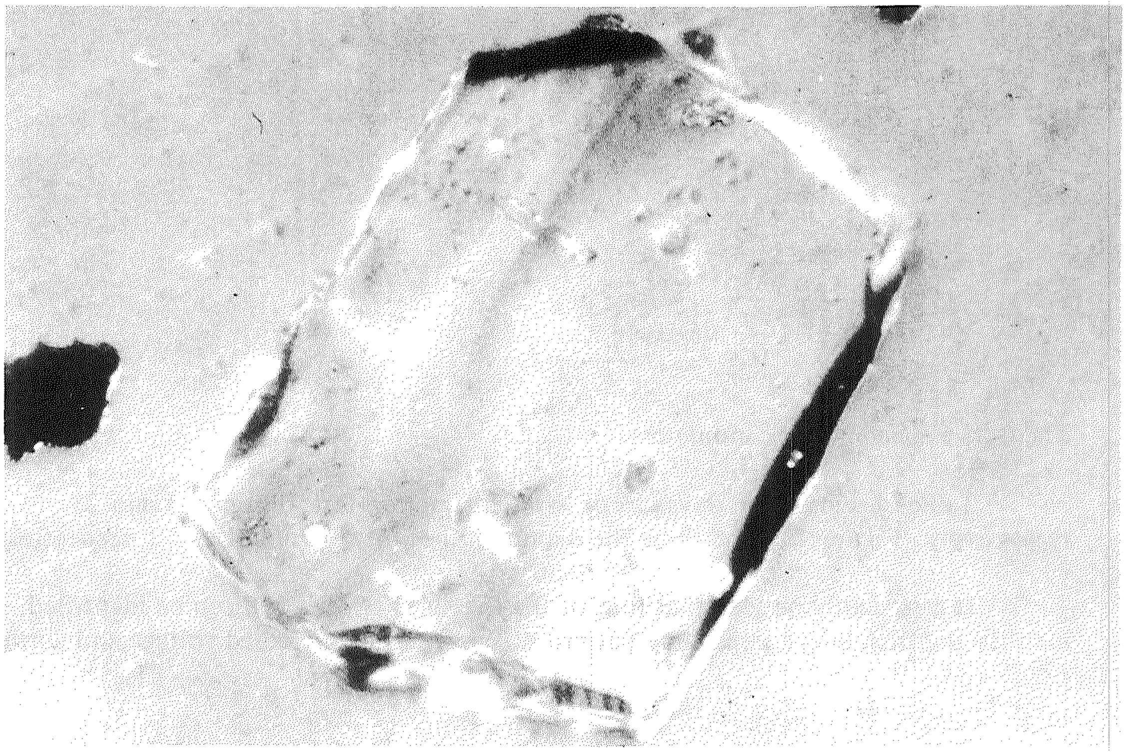
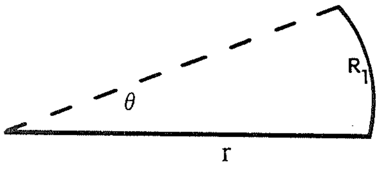


Figure 6. Diamond crystal – largest dimension approximately 0.14 mm.

The equation below was used to derive the d values.



$a = S_1 - R_1$

where

a = the line spacing on the film (cm).

S_1 = the diameter of the first circle on the film.

R_1 = the radius of the first circle on the film.

r = the distance of the film from the sample (14.32 cm).

θ = the angle between the incident X ray beam and the crystal planes in the crystolite of the specimen.

d = the lattice spacing (nm).

λ = the wavelength of the X rays (1.5404 Å).

Then

$$d = \frac{\lambda}{2 \sin \theta} \quad \text{(Bragg's Law).}$$

$$\theta = \frac{a}{2\pi r} (360^\circ).$$

$$d = \frac{\lambda}{2 \sin \left(\frac{180\,a}{r} \text{ degrees} \right)} .$$

The lattice spacing is in nanometers.

Tables 1 and 2 give d values of X-ray diffraction test, film nos. 1 and 2, respectively. Figures 7 and 8 show the diamond lines for film nos. 1 and 2, respectively.

It can readily be seen that four of the five lines of diamond can be identified. To identify the fifth line, a new X-ray pattern was made using increased voltage and current.

TABLE 1. d VALUES OF X-RAY DIFFRACTION TEST, FILM NO. 1 (FIG. 7)

Line	Uncorrected a	d	Book d Values (diamond)
1	3.13	3.31	
2	4.92	2.28	2.06
3	5.43	2.08	
4	6.68	1.71	

Parameters for Pattern No. 2 (May 22, 1970)

40 kVp

30 mA

Time of film exposure: 4 hr

Wavelength: 1.5405 Å

Type of film: Kodak F0034DA

Film no. 1 identifies the fifth line which does not appear on film no. 2. The d values for the films were computed by a desk model calculator. The uncorrected $S_1 - \theta_1$ is included for possible future reference. The lines not attributed to diamond can be attributed to residual graphite, Canadian balsam cement, and the glass sample holder. Both films positively identified diamond.

Another experiment was tried in which an additional capacitor bank was connected between the horn and the anvil as can be seen in Figure 1. Since the graphite has a very much greater resistance than the horn or anvil, when this capacitor bank was discharged, its energy appeared in the graphite in the form of heat. Isolation is maintained between the two capacitor banks by the sheet of Mylar between the coil and the copper disk. The shock wave was applied 100 μs after the heat was applied. No difference in the diamond yield was observed with graphite. However, when charcoal carbon was tried, a minor explosion occurred with a spit of flame and smoke almost as though a small charge of black powder had been set off. No diamond material was observed in the debris.

TABLE 2. d VALUES OF X-RAY DIFFRACTION TEST, FILM NO. 2 (FIG. 8)

Line	Uncorrected a	d	Book d Values (diamond)
1	4.96	2.27	
2	5.74	1.97	
3	6.31	1.81	
4	6.91	1.66	
5	7.24	1.59	
6	7.54	1.53	
7	8.07	1.44	
8	8.56	1.37	
9	9.24	1.28	
10	9.51	1.25	1.26
11	9.75	1.22	
12	10.04	1.19	
13	10.23	1.18	
14	10.53	1.15	
15	11.44	1.08	1.08
16	11.94	1.04	
17	12.42	1.01	
18	14.38	0.91	0.89
19	17.97	0.81	0.82

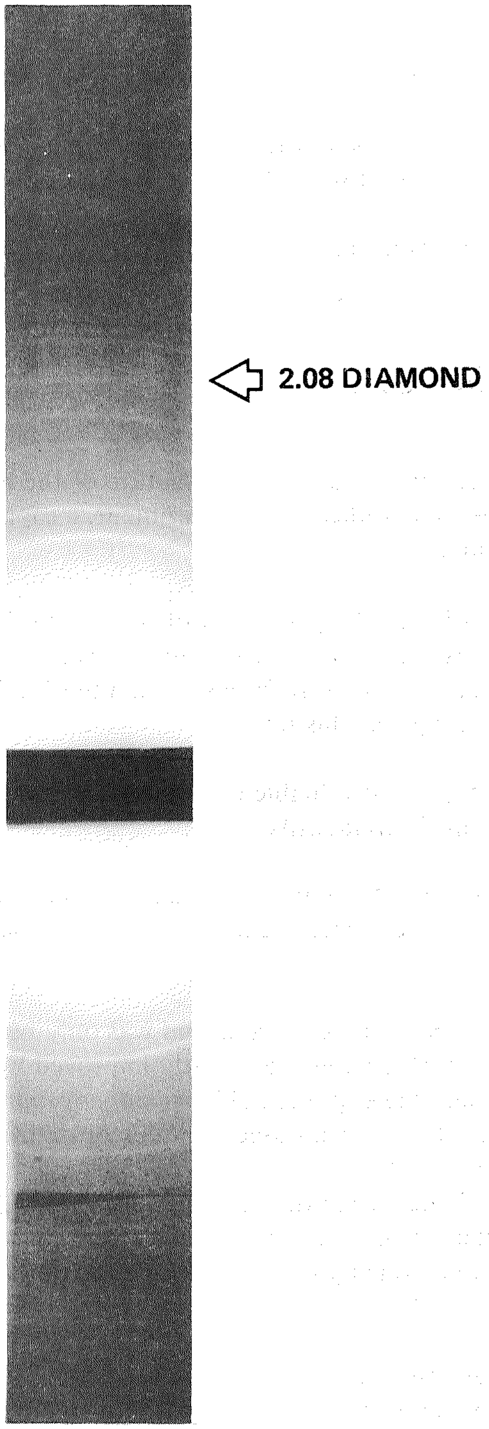


Figure 7. Diamond lines for film no. 1.

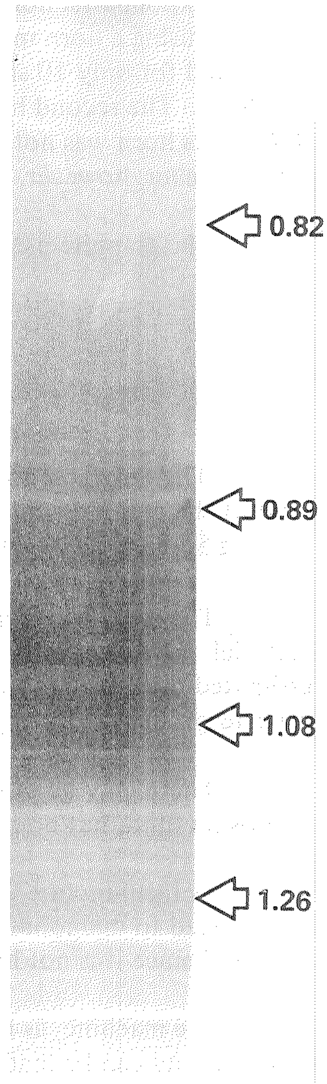


Figure 8. Diamond lines for film no. 2.

Exponential horns of two different materials were used. The first attempts were made by using a horn made of 4130 steel. This horn worked very well, but it was too soft and lasted for only 10 shots. The crystals shown in Figures 3 through 6 were made with this horn. The second horn was made of maraging steel which was hardened to Rockwell 40C. This horn was not damaged after many shots, but the diamonds produced were very much smaller; however, these were the ones used for the X-ray diffraction tests.

The graphite used was a laboratory grade graphite produced by the Fisher Scientific Co.

CONCLUSIONS

The only cost to perform the experiment was the manufacture of the horn and the cost of the graphite and X-ray film strips. To duplicate the experiment, a capacitor bank costing $5000 to $10 000 would be required.

The main advantage of the process is the likely low cost of producing the diamond material. The electricity used costs pennies, and graphite is certainly cheap enough. It is estimated that this process should produce diamond grit for less than a tenth of what it costs to produce it by any other method known at this time.

There may be other crystals of unique and valuable properties that can be produced by this method, but no attempt has been made to identify them.

At this juncture, the process is not very efficient. Less than 1 percent of the graphite is converted to diamond in one shot; however, the graphite not converted to diamond can be reused since it is not lost.

The machine, as built for the experiment, is very crude and was only built to prove that diamond can be made this way. The machine could be greatly improved by determining the exact velocity of the shock wave through the exponential horn and appropriately adjusting the length for maximum effect. The materials used could be optimized.

Better yields should be obtained by increasing the velocity and amplitude of the shock wave. This could be done by shortening the rise time of the current pulse through the hammer coil. For instance, this could be done by crowbarring the circuit at the current peak or perhaps some other means could be found.

There may be some other forms of graphite or carbon that would give a better yield. That tried by this laboratory was a questionable grade of graphite and charcoal carbon.

If one were going into the business of making diamond grit, a machine should be constructed that would be automatic in operation. The graphite would be loaded and removed automatically and the capacitor bank cycled automatically. The machine could also be made very large.

Finally, it is believed that although this is an excellent method for producing industrial grade diamond crystals, a need for more development work is indicated.

REFERENCES

1. Bundy, F. P.; et al.: Man-Made Diamonds. Nature, July 9, 1955, p. 51.

2. Bundy, F. P.: Direct Conversion of Graphite to Diamond in Static Pressure Apparatus. J. Chem. Phys., vol. 38, no. 3, Feb. 1, 1963, p. 63.

3. De Carli, P. S.; and Jamieson, J. D.: Formation of Diamond by Explosive Shock. Science, vol. 133, no. 3466, June 2, 1961.

4. De Carli, P. S.: Shock Wave Synthesis of High Pressure Phases; Comments on the Origin of Meteoritic Diamond. Science and Technology of Industrial Diamonds. Proceedings of the International Industrial Diamond Conference. Vol. I: Science, Industrial Diamond Information Bureau, Oxford, 1967.

5. U. S. Patent Number 3,238,019.

6. U. S. Patent Number 3,360,972.

APPROVAL

TM X-64543

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This document has also been reviewed and approved for technical accuracy.

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