Production of Fullerenes with Concentrated Solar Flux

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NOTICE

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PRODUCTION OF FULLERENES WITH CONCENTRATED SOLAR FLUX

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ABSTRACT

Research at the National Renewable Energy Laboratory (NREL) has demonstrated that fullerenes can be produced using highly concentrated sunlight from a solar furnace. Since they were first synthesized in 1989, fullerenes have been the subject of intense research. They show considerable commercial potential in advanced materials and have potential applications that include semiconductors, superconductors, high-performance metals, and medical technologies. The most common fullerene is C_{60}, which is a molecule with a geometry resembling a soccer ball. Graphite vaporization methods such as pulsed-laser vaporization, resistive heating, and carbon arc have been used to produce fullerenes. None of these, however, seems capable of producing fullerenes economically on a large scale. The use of concentrated sunlight may help avoid the scale-up limitations inherent in more established production processes. Recently, researchers at NREL made fullerenes in NREL's 10 kW High Flux Solar Furnace (HFSF) with a vacuum reaction chamber designed to deliver a solar flux of 1200 W/cm^2 to a graphite pellet. Analysis of the resulting carbon soot by mass spectrometry and high-pressure liquid chromatography confirmed the existence of fullerenes. These results are very encouraging and we are optimistic that concentrated solar flux can provide a means for large-scale, economical production of fullerenes. This paper presents our method, experimental apparatus, and results of fullerene production research performed with the HFSF.

FIGURE 1. A CLOSED POLYHEDRA C_{60} MOLECULE DIAGRAM

INTRODUCTION

Just a few years ago carbon was considered to exist only in a limited number of forms, such as diamond, graphite, glassy carbon, amorphous carbon, and a number of high-temperature species that existed in the vapor phase above 2000 °C. In 1984, however, mass spectrometry revealed that carbon could exist in a number of other forms ranging from C_{30} to C_{190} (Rohling, 1984).

A year later the unique stability of molecular allotropic forms such as C_{60} and C_{70} was demonstrated (Kroto, 1985). These events led to the discovery of a whole new set of carbon-based substances known as fullerenes. Fullerenes are comprised of closed polyhedra or tubes produced by carbon atoms linking together to form hexagons and pentagons, as shown in Figure 1.

The arrangement of carbon atoms in fullerenes raises the possibility of properties that have intrigued chemists, physicists, materials scientists, and medical researchers. A few examples of fullerene properties and/or applications serves to illustrate this. Fullerenes have been shown to polymerize in several ways. Metal atoms can be placed inside the fullerene cage to form encapsulated systems (UC_{28}, LaC_{82}, etc.)(Bandow, 1993), or outside the cage to form catalysts (Nagashima, 1992). The fullerene cage can be reacted with other substances to form new molecules. One example of particular interest is a possible HIV drug (Sijbsma, 1993). The tubules are of interest as fibers, nanowires, and encapsulants (Ebbesen, 1992). Fullerenes may be doped to form electronic materials or reacted to form superconductors (Holezer, 1991). Other examples of fullerene properties and applications may be found in reviews in the literature (Kroto, 1992, Taylor, 1993, Buckminsterfullerenes).

All of these applications were identified following isolation of the first macroscopic amounts of the most common fullerene, C_{60}, in 1990 (Kratshmer, 1990). Much of the work on fullerenes has been performed using smaller amounts of the material because synthetic approaches to these forms of carbon had yielded limited quantities. Indeed, the major roadblock to commercialization of some fullerene applications lies in the lack of a large-scale method of producing and isolating material.

BACKGROUND

All fullerene production methods require a source of small, gas-phase carbon clusters (one to possibly ten atoms). Two sources of these clusters exist. One is the combustion of hydrocarbons such as benzene in an oxygen-deficient flame to produce a soot (along with other combustion products, such as H_2O, CO, and CO_2) (McKinnon, 1992). Although the percentage of fullerenes in the soot may be relatively high, the yield of fullerenes compared to the mass of hydrocarbon consumed is rather low. The second source of gas-phase carbon clusters uses an energy source to vaporize elemental carbon at temperatures above 3000°C. The vaporized carbon then condenses into carbon soot. A number of energy sources have been used to vaporize the carbon, including lasers, plasmas, induction heating (Peters, 1992), and arcs struck between graphite rods. The arc method has proven to be the most useful. However, even the best methods yield only tens of grams of fullerenes per hour. Despite considerable effort, none of these methods has been brought to large-scale production. This led us to investigate the production of fullerenes with a solar furnace.

Highly concentrated solar energy offers the potential for efficient fullerene production (Chibante, 1993). The carbon source, graphite, efficiently absorbs solar light, and the solar beam can be directed and shaped to accommodate a variety of reactor designs (Fields, 1993). This light energy can be deposited directly on the graphite source. Cold-wall reactor systems can be used, thus mitigating materials problems at the required 3000°C-or-greater temperature range. Reaction chambers that are free of contaminants like oxygen and water can be easily designed. The process can be easily scaled to production size. In addition to these advantages there are other significant benefits. The condensation process where carbon vapors form fullerenes can be controlled by appropriate designs of optics and reactor geometries. Greater yields than in any other process may be attainable due to the relatively low intensity of ultraviolet light (Chibante, 1993). These features can also help minimize cost and energy requirements.

The solar furnace at NREL has the design features necessary to support experiments in fullerene production. A diagram of NREL's HFSF is shown in Figure 2. The heliostat directs solar radiation onto a stationary, primary concentrator. The primary concentrator has 25 individual facets that focus the concentrated solar beam off-axis through an attenuator and a shutter and onto a target (Bingham, 1991). This furnace can deliver a maximum power of 10 kW. Using only the furnace's primary concentrator, the concentrated solar beam has a 12-cm diameter and a peak flux of 250 W/cm^2. When additional concentration is required, as in our fullerene experiments, a reflective secondary concentrator (designed and built by the University of Chicago) (Lewandowski, 1991) can be placed at the furnace's focal point. The secondary concentrator has demonstrated concentrations of over 20,000 suns (2000 W/cm^2 under clear-sky conditions).

EXPERIMENTAL APPARATUS AND METHOD
Equipment

The reaction chamber we designed for our experiments is shown in Figure 3. The vertical apparatus is a vacuum sealed system with gas flow ports that allow the experiments to be conducted in an oxygen-free environment. It is sealed on top with a quartz window on top of a Viton O-ring. The gas inlet port and secondary concentrator are beneath the quartz window. A needle valve outside the chamber regulates gas flow into the chamber. The carbon sample sits at the exit aperture of the secondary concentrator, which is the region of highest solar flux. The secondary concentrator and the horizontal surface just beneath it have internal channels connected to ethylene-glycol cooling lines. The bottom section of the reaction chamber is connected to a pumping system fitted with a mechanical pump, a turbomolecular pump, a butterfly valve, and a residual gas analyzer (RGA).

During an experiment, concentrated sunlight passes through the reaction chamber's quartz window and into the secondary concentrator. The concentrated solar flux (approximately 1300 W/cm^2) impinges on the top surface of the carbon sample, which vaporizes and condenses as soot on the chamber's walls. Throughout the experiment, an inert gas (we used argon) flows from the inlet port, through the secondary concentrator, over the carbon sample, and out of the vessel through the vacuum pumping port. In addition to providing an oxygen-free environment, the flowing gas also helps cool the system and keeps the secondary concentrator clean by sweeping the carbon vapor away from the reflective surface.

We used graphite as the carbon source for all our fullerene experiments. Samples were cut from a 0.75-cm-thick slab of 99.999% pure graphite from the Kurt J. Lasker Co. We cut several 1-cm-diameter cylinders from this slab. For our preliminary experiments we used unmodified graphite cylinders. For subsequent experiments we modified the cylinders either by
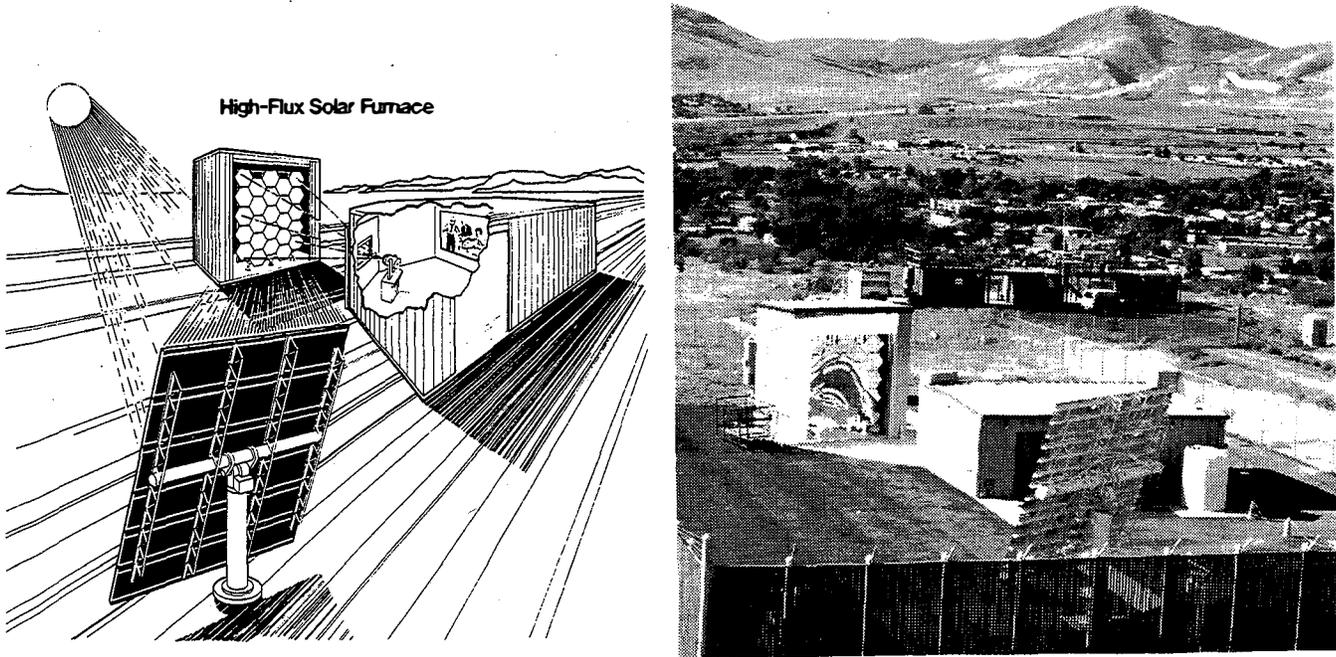
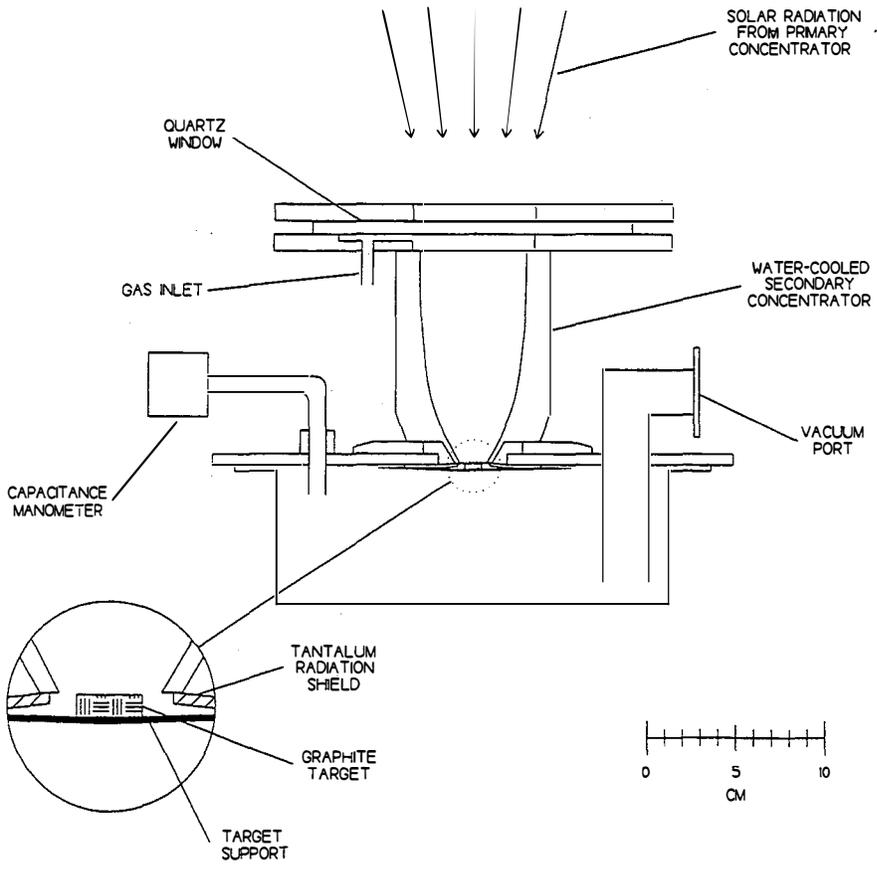FIGURE 2. DIAGRAM AND PHOTOGRAPH OF NREL'S HIGH FLUX SOLAR FURNACE

FIGURE 3. REACTION CHAMBER DESIGNED FOR FULLERENE EXPERIMENTS

3

bevelling the top edge to form a conical shape or by cutting a small, hemispherical cavity into the top surface.

We used a circular-foil calorimeter at the exit of the secondary concentrator to measure the flux. The calorimeter is water cooled to tolerate the high flux at the exit of the secondary concentrator. A normal incidence pyrheliometer (NIP) was used to measure direct-normal irradiance.

Procedure

Prior to beginning the experiments, we tested the experimental apparatus, calibrated the gas flow equipment, and measured the solar flux available at the carbon sample top surface. We used helium to perform preliminary leak-detection tests on the reaction chamber. No leaks were found. Using the turbomolecular pump, we were able to pump the vessel down to 5×10^{-5} torr.

We used the circular-foil calorimeter to measure the flux at the secondary-concentrator exit with the attenuator in several different positions. The attenuator was used to control the level of power reaching the reactor system. We also measured the flux at varying distances from the concentrator exit with the attenuator fully open.

For each experiment, we placed a graphite sample at the exit of the secondary concentrator, assembled the reaction chamber, and positioned the apparatus so the secondary concentrator was at the HFSF's focal point. We connected the reaction chamber to the vacuum system and cooling lines and then subjected it to a pumping and purging process to clear it of air and water vapor. We pumped the chamber down to 50 torr, then backfilled the vessel with argon to 400 torr. This pumping and purging was repeated four times. We then closed the argon inlet valve and pumped the vessel down to 5×10^{-5} torr to ensure that it was not leaking.

We performed several experiments with different experimental conditions. We adjusted the argon flow to the proper level and adjusted the butterfly-valve setting to bring the chamber to the desired experimental pressure (either 50 torr or 100 torr, depending on the experiment). Once the chamber pressure was steady, the chamber was illuminated with the concentrated solar beam for approximately three minutes to "bake out" any impurities in or on the graphite sample. The sample was then removed from the chamber, weighed, and replaced. The above procedures were repeated, beginning with the pumping and purging and finishing with the chamber pressure adjusting, after which we exposed the graphite sample to the concentrated beam for anywhere from 30 seconds to two minutes. Following the exposure we opened the reaction chamber and removed the soot from the condensing surfaces with a small brush, bottled it, and reweighed the graphite sample. The percent yields for our soot samples were determined by taking toluene extractions and matching their high-pressure liquid chromatography (HPLC) retention times with extracts from commercially available soot samples (Bucky USA). The yield analysis was performed on an Isoo Model 2360 HPLC instrument using a reverse phase column (Waters µ Bondpack C18 30 cm) with toluene/methanol (45:55 at 2 mL/min) as the eluant.

EXPERIMENTAL DESIGN
Incident Flux Measurements

We performed flux measurements to ensure that the graphite sample temperature was going to be in the range for vaporization. The results of the flux measurements are shown in Figures 4 and 5. All these data were normalized to a direct-normal irradiance of 950 W/m^2, which was representative for the days the tests were conducted. In Figure 4, the measured peak flux is plotted as a function of the attenuator opening. The plot shows that, with the attenuator fully open, a flux of over 1300 W/cm^2 (corresponding to a concentration just under 15,000) was available at the exit of the secondary concentrator. The data in Figure 4 was measured with all 25 of the primary concentrator facets uncovered. Similarly, all the facets were uncovered for the fullerene experiments.
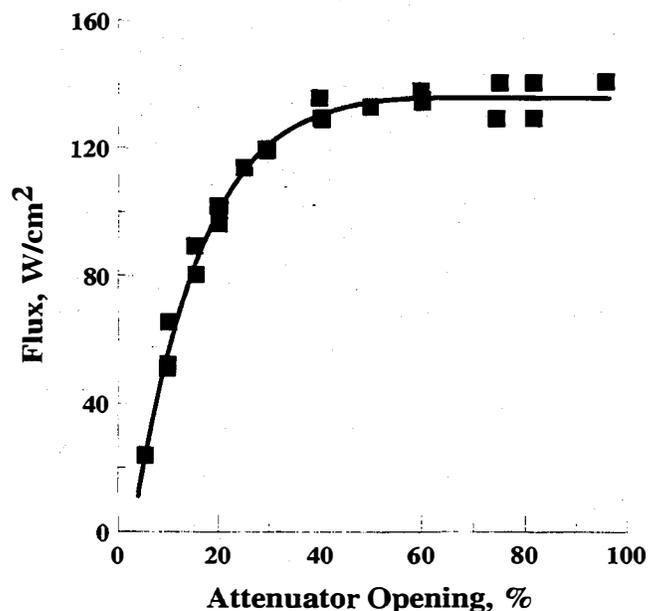
FIGURE 4. SOLAR FLUX AT CPC EXIT AS A FUNCTION OF ATTENUATOR OPENING

In Figure 5, the flux is plotted as a function of various distances from the secondary-concentrator exit. We compared these measurements to the flux predicted by a Monte Carlo computer code (SOLFUR) (Jorgensen, 1991) written to explicitly model various optical components of solar furnaces. Using an additional code specific to reflective secondary concentrators, the set of rays from SOLFUR was traced from the entrance plane of the secondary concentrator to a plane placed at various distances below the exit. The measured data in Figure 5 was gathered with 19 of the primary concentrator facets uncovered.

4

1150 W/cm² (which corresponds to a direct-normal irradiance of 950 W/m² and a concentration of 12,000 suns), the sample temperature reaches the 2500°C range. This is lower than the 3000°C-temperature range achieved in the other fullerene production methods. This may be a result of the simplified nature of the heat transfer analysis performed here, or it may be that there is a phenomenon associated with solar energy that allows the vaporization of carbon to occur at lower temperatures.
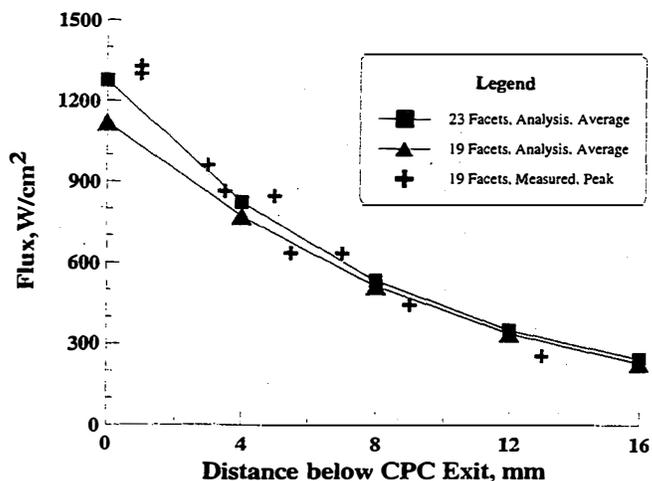
Examination of the different heat-transfer components in Equation 1 reveals that at a flux of 1150 W/cm², if heat loss due to thermionic emission were neglected the sample temperature increase by almost 50°C. Similarly, if convection were neglected, the temperature would increase by about 150°C.

FIGURE 5. SOLAR FLUX AS A FUNCTION OF DISTANCE FROM CPC EXIT

Heat Transfer Analysis

We performed a simplified energy balance analysis on the experimental sample to get an estimate of its temperature once it reached steady state conditions. We considered radiative emission, thermionic emission, and convection as the three dominant modes of heat transfer. We did not consider the effects of conduction because the configuration of the sample holder was not conducive to conduction from the sample to the sample holder, which made the temperature of the bottom surface of the carbon sample difficult to estimate. The energy balance equation we used is shown below in Equation 1.

$$\alpha I A_{top\ surface} = \varepsilon \sigma A_{sample}(T^4_{sample} - T^4_{chamber})$$
$$+ 4\pi m \sigma^2 h_p^{-3} e^{-\phi/\sigma T} A_{sample} T^3_{sample} \quad (1)$$
$$+ h A_{top\ surface}(T_{sample} - T_{argon})$$

where,
α = absorptivity of graphite, 0.925 (Touloukian, 1972),
I = solar flux incident on sample (W/m²),
ε = emissivity of graphite, 0.74 (Touloukian, 1972),
σ = Stefan-Boltzmann constant,
$A_{top\ surface}$ = area of top surface of graphite sample,
m = mass of electron,
h_p = Planck's constant,
ϕ = work function, 4eV (Kittel, 1968),
h = convective heat transfer coefficient, 850 (W/m²K) (Gardon, 1962 and Chan, 1993).

In Figure 6 the carbon sample temperature, calculated from Equation 1, is plotted as a function of solar flux incident on the sample. The plot shows that at a flux of approximately

FIGURE 6. CARBON SAMPLE TEMPERATURE AS A FUNCTION OF INCIDENT SOLAR FLUX

Experimental Optimization

Several variables in the experimental conditions had the potential of affecting fullerene yields in our experiments. We wanted to know the effect of these variables to determine how to adjust them for optimal fullerene yield. To determine the influence of the different experimental variables, we used the Plackett-Burman design of balanced incomplete blocks to design a set of experiments. The Plackett-Burman design is a two-level method ("plus" denoting high level and "minus" low level) of experimental design where as few as N experiments can be used to study the effect of as many as N-1 variables (Stowe, 1966). The variables are usually associated with an experimental parameter (for example, gas pressure), but they can also be dummy variables. Dummy variables are not associated with any experimental parameters. They add more experiments to the

design and thereby refine the statistical calculations of variable effect and standard error.

According to the Plackett-Burman design, the effect of any one variable on the experimental result is determined by the difference between the average value of the results for the positive runs less the average value of the results for the negative runs (see Equation 2).

$$E_{variable} = \frac{\sum R_{(+)}}{No.of(+)runs} - \frac{\sum R_{(-)}}{No.of(-)runs} \qquad (2)$$

where,

$E_{variable}$ = effect of variable on results (in our case, % fullerene yield),
$R_{(+)}$ = results from experiments with variable at positive value,
$R_{(-)}$ = results from experiments with variable at negative value.

The standard error in the effect is calculated with Equation 3.

$$SE_{effect} = \sqrt{\frac{\sum (E_{dummy})^2}{n}} \qquad (3)$$

where,

$S E_{effect}$ = standard error of any effect calculated with Equation 3,
E_{dummy} = effect of dummy variable(s) on result,
n = number of dummy variables.

In our experiments we considered the effects of seven different variables. According to the Plackett-Burman method, this required a set of eight experiments. The experimental variables and their high and low values are given in Table 1. Table 2 gives the distribution of the high and low parameters throughout the set of eight experiments.

TABLE 1. VARIABLES AND PARAMETERS USED FOR THE PLACKETT-BURMAN EXPERIMENT DESIGN.

Variable	High Parameter (+)	Low Parameter (-)
A = Solar flux	1300 W/cm^2	1150 W/cm^2
B = Position of sample	at secondary concentrator exit	2 mm below concentrator exit
C = Sample shape	cavity	cone
D = Length of exposure	2 min	1 min
E = Gas flow rate	100 mil	50 mil
F = Type of gas	He	Ar
G = Gas pressure	100 torr	50 torr

TABLE 2. PLACKETT-BURMAN MATRIX FOR EXPERIMENTAL PARAMETERS.

Run	A	B	C	D	E	F	G
1	+	+	+	-	+	-	-
2	+	+	-	+	-	-	+
3	+	-	+	-	-	+	+
4	-	+	-	-	+	+	+
5	+	-	-	+	+	+	-
6	-	-	+	+	+	-	+
7	-	+	+	+	-	+	-
8	-	-	-	-	-	-	-

TABLE 3. MASSES OF SOOT PRODUCED DURING TWO LEVEL EXPERIMENT AND CORRESPONDING FULLERENE YIELD.

Run	Soot Mass (mg)	% Yield of Fullerenes
1	7.0	3.2
2	7.6	12.8
3	0	NA
4	4.7	7.3
5	0	NA
6	0	NA
7	12.6	13.9
8	0	NA

TABLE 4. RELATIVE EFFECTS OF EXPERIMENTAL VARIABLES AND THE STANDARD ERROR ON PERCENT FULLERENCE YIELDS

Variable	Effect
Position of sample (B)	9.3
Length of exposure (D)	4.05
Gas flow rate (E)	4.05
Solar flux (A)	1.3
Dummy (F)	1.3
Sample shape (C)	0.75
Chamber pressure (G)	0.75
Standard Error	1.3

We calculated the effects of the different experimental variables and the associated standard error using Equation 2 and Equation 3. These statistical results are given below in Table 4.

The relatively large standard error in the effect calculations eliminates the significance of the effects calculated for three of the variables: the solar flux (A), the shape of the sample (C), and the chamber pressure (G). The calculated effects of the remaining three variables, however, are outside the standard error range. The position of the sample relative to the secondary concentrator exit had the most significant effect.

The effects of the length of exposure and the gas flow rate calculated to be the same. The fact that no soot was ever produced when the carbon sample was in its low position increased the likelihood for two effects to calculate to the same value. The fact that the four experiments with low sample position resulted in zero soot production obviously lessened the impacts of these runs. It is likely that the effects of the length of exposure and the gas flow rate were not equally significant to the fullerene yield. It should be noted that the effect of the rate of gas flow was actually a negative value; in other words, a low gas flow rate was more conducive to fullerence production. The high gas flow probably cooled the graphite sample and inhibited vaporization.
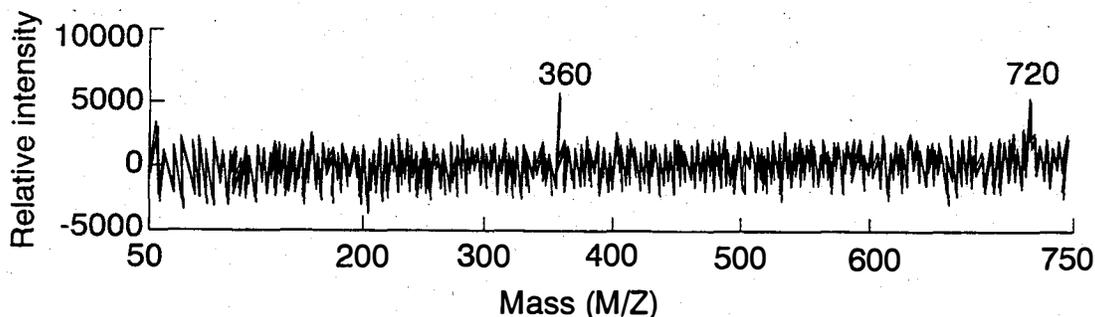
FIGURE 7. MASS SPECTRUM OF C_{60} IN SOOT PRODUCED AT THE SOLAR FURNACE

Preliminary Experiments

We performed a series of preliminary experiments to determine the high-low limits given in Table 1. For example, in one set of experiments, we observed that very little soot was produced for exposure times less than one minute. But as we lengthened the exposure time, the temperature of the experimental apparatus became dangerously high after approximately two minutes. During these preliminary experiments, we also discovered that no fullerenes were forming when we used helium. We therefore kept the type of gas constant throughout our set of eight experiments, and the variable "F" became a dummy variable. (Originally, we did not include a dummy variable in our set of experiments.) We speculate that the superior heat transfer characteristics of helium, as compared to argon, prevented the carbon sample from reaching a high enough temperature for vaporization.

During the preliminary experiments we used mass spectrometry to confirm the existence of C_{60} in our collected soot. Figure 7 shows the mass spectrum of a 1 mg soot sample we produced during our preliminary experiments. The mass spectrum shows significant peaks at 360 amu and 720 amu for the doubly and singly charged C_{60} molecules.

EXPERIMENTAL RESULTS

We performed our set of eight experiments under the conditions prescribed in Table 2. Table 3 gives the mass of soot produced during each of the experiments and the percentage of C_{60} in each soot sample.

Table 3 shows that only half of the experiments resulted in appreciable soot production. Examination of the data reveals that all of the experiments with the sample at the concentrator exit resulted in a measurable amount of soot, but no soot was produced in the experiments where the sample was 2 mm below the concentrator exit. This indicates that our negative value for the sample positioning was too extreme.

Of the soot-producing experiments, run 7 showed the most promising results. This experiment resulted in the largest amount of soot and the highest yield. Although the difference in the percent yield between runs 7 and 2 is probably within the margin

of experimental error (13.9% vs. 12.8%), the difference in the amount of fullerenes produced was significant. Run 7 produced approximately 1.8 mg of fullerenes compared to just under 1 mg in run 2. Runs 2 and 7 were both performed with a high exposure length and low gas flow rate, but run 7 was performed with a low flux parameter, a cavity-shaped sample, and low chamber pressure. Our experience during these experiments was that the cavity-shaped samples often showed much greater signs of vaporization activity than did the conical samples. Sample shape is likely the parameter responsible for the large soot (and hence fullerene) production in run 7. Figure 8 shows the retention-time plot from the HPLC analysis of the soot run 7.
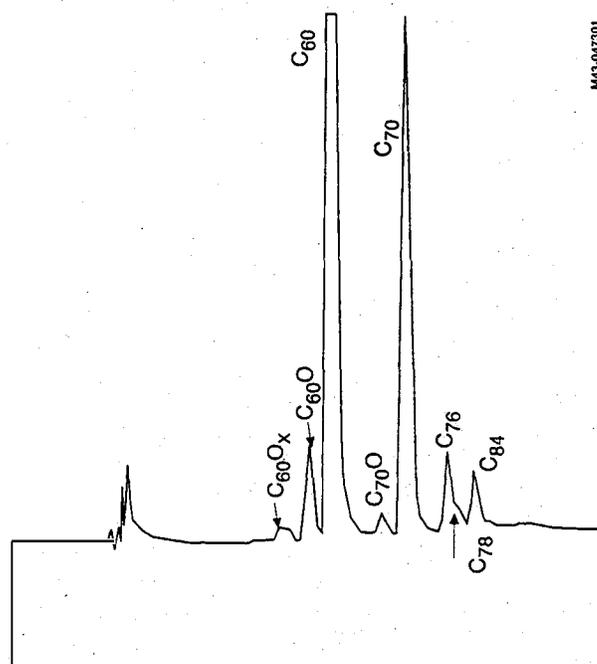

FIGURE 8. HPLC OF TOLUENE EXTRACT FROM SOOT PRODUCED AT THE SOLAR FURNACE

CONCLUSIONS AND RECOMMENDATIONS

Although the low sample position was too extreme to realize the full benefit of the Prackett-Burman design, the effect analysis wasstill beneficial. The calculated effects revealed that the carbon sample should be positioned at the concentrator exit and the lower gas flow rate and the longer exposure times were both conducive to high yields. Another, more refined, set of two-level experiments would help us more completely understand the effects of different experimental variables.

The 14% fullerene yield from our soot sample was very encouraging. It warrants further exploration into solar fullerene production. Areas to be considered for yield and amount improvements include optimizing the reactor geometry, further investigation of experimental variables, and scale-up to larger system sizes.

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