PREPRINT UCRL- 81588 REV. I

Lawrence Livermore Laboratory

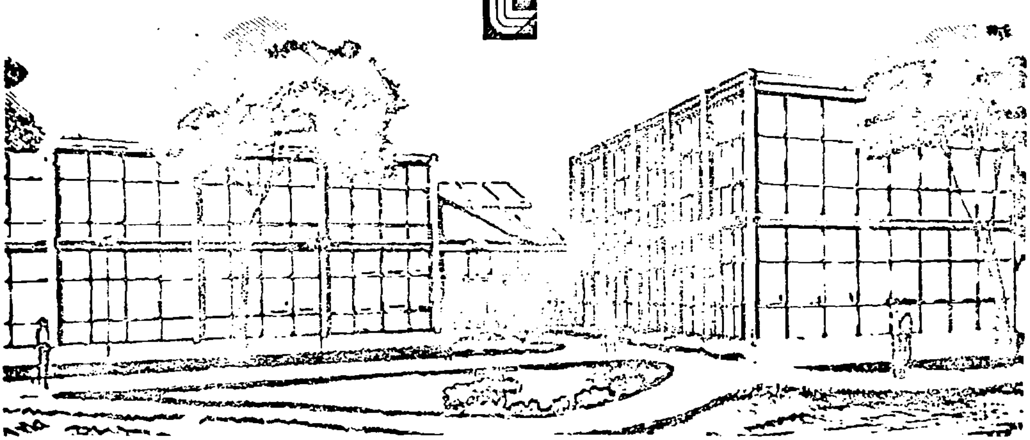
FUEL PELLETS AND OPTICAL SYSTEMS FOR INERTIALLY CONFINED FUSION

Charles D. Hendricks

May 8, 1979

MAS

This paper was prepared for submission to the
First Topical Meeting on Fusion Reactor Materials
Miami Beach, Florida, January 29-31, 1979



FUEL PELLETS AND OPTICAL SYSTEMS FOR INERTIALLY CONFINED FUSION*

C. D. HENDRICKS

Lawrence Livermore Laboratory, Livermore, California 94550, USA,

Current laser-driven ICF targets are complex sets of concentric spherical shells made from a variety of materials including the fuel (e.g., deuterium-tritium), glass, beryllium, gold, polymeric materials, organo-metallics, and several additional organic and inorganic materials depending on the particular experiments to be done. While we don't yet know what the reactor targets will be exactly, there is little reason to believe they will be just simple, low quality glass shells containing DT gas or simple spheres of deuterated polyethylene or other fuel. Consequently, we consider many of the current targets, materials, and fabrication techniques to be applicable to the long range problems of ICF reactor target fabrication. Many of our current material problems and fabrication techniques are discussed and various quality factors are presented in an attempt to bring an awareness of the possible fusion reactor target materials problems to the scientific and technical community.

We should note that "target" and "pellet" are used interchangeably and do not have any hidden variational significances.

1. INTRODUCTION

Inertial Confinement Fusion (ICF) is a process by which a fuel such as a deuterium-tritium is compressed to high densities and heated to high temperatures to cause nuclear fusion. The inertia of the compressed material confines it for the short time during which the thermonuclear burn process occurs.

In a magnetic confinement system the fuel is to be contained in a magnetic field configuration for a sufficiently long time that the high temperature fuel nuclei are able to fuse. The fuel densities are in the 10^{14}-10^{15}/cc range and confinement times are of the order of 0.1 to 1.0 second. The number densities of fuel atoms in ICF implosions are about 10^{26}/cc and confinement times are about 10^{-12} second. Of course, in both magnetic confinement and inertial confinement fusion systems, the fuel temperatures must reach 10^8 K or greater for the nuclei to overcome repulsive forces and interact. Deuterium and tritium fuse to form a helium nucleus, a neutron, and 17.5 MeV of energy.

Since the inertial confinement fusion program is still in a "research" phase, detailed specifications for reactor chamber shapes, first wall protection methods, final beam turning materials and configurations, and indeed. driver specifications or types themselves are not known. In addition, perhaps even more seriously, the reactor target configuration is not even known in any detail. This is serious because the target design is a driving or determining factor in driver power and energy, pulse length, reactor chamber design, first wall protection. energy extraction, radioactive debris disposal and many other aspects of ICF power production (not the least of which is power cost to the user). An area in which the lack of a well defined target design is almost catastrophic is that of target fabrication research, development, and production studies. While it *may be literally years before a reactor target* design is made firm, we cannot afford to wait until then to develop fabrication techniques and materials processes for building the targets. We must take what we know of present target designs and extrapolate as best we can to the future reactor target possibilities and begin our development work. In addition, it is necessary to produce targets for research experiments now. We feel that the materials research and development being done on today's and near-term future's targets is relevant to the long term future targets.

That the targets will not be simple spheres of glass filled with gaseous DT or simple spheres of solid CD_2 or CDT is almost certain. Target designs are tailored to obtain the best yield, gain, and utilization of driver capability. These considerations are not usually met by a simple, single layer sphere but require multiple shells of various materials: High-Z for reduction of fuel pre-heat, high density for pusher or tamper use, low-Z for ablators, etc. Shell thicknesses, spacings, and material properties must be determined to achieve shock timing, compression velocities, and appropriate hydrodynamic behavior of the targets resulting in desirable burn characteristics in the compressed fuel.

Target design specifications include high surface quality and material density uniformity. This is to avoid the disasterous growth of Rayleigh-Taylor instabilities. Small surface and volume irregularities can grow sufficiently during the implosion to produce non-spherical compression of the target which can result in very unsatisfactory burning of the fuel at best or total failure at worst.

*Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under Contract Number W-745-5-ENG-48.

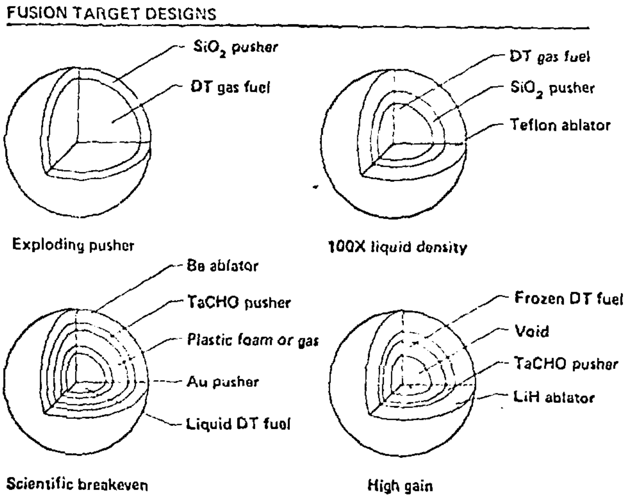


Fig. 1. Typical designs for laser fusion research pellets.

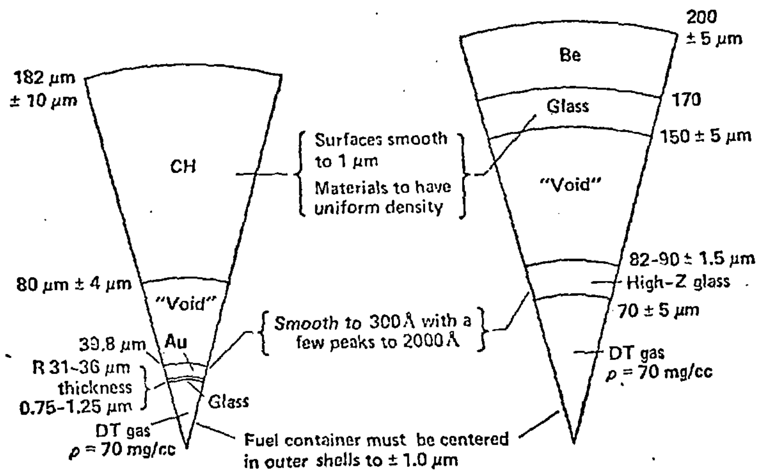


Fig. 2. Detailed pellet design specifications. The "pie"-shaped sections are taken from typical pellet designs and include dimensions and tolerances.

It is not anticipated that the quality specifications will be drastically changed for reactor target designs as compared with those for our current research target designs.

As a result of required production rates, material and geometrical surface considerations, reactor-class laser fusion targets will present some unique fabrication challenges. Targets will be required at rates of 1 to 10 per second and must be delivered accurately to the laser focal point in the target chamber and be there coincidentally with the laser pulse.

Free surfaces will be required to be extremely smooth; 1000 Å to 2000 Å peak-to-valley defects whose lateral dimensions are approximately equal to the shell thickness may be the largest permitted. Interfacial surfaces must have the same smoothness characteristics as free surfaces. Material nonuniformities or voids can be tolerated only to the same extent as the equivalent surface defects. One or more of the layers in the target may be cryogenic substances such as deuterium, deuterium-tritium, or normal hydrogen. If two separate frozen layers are to be placed, frozen and maintained in the target, surrounded by layers of polymerized fluorocarbon, beryllium, gold, glass, or other materials, there are difficulties in depositing the cryogenic material in uniform layers and keeping them frozen during the deposition or assembly of the other layers.

Since the target configurations include opaque materials, characterization by optical interferometry will be almost impossible. Microradiography can provide data on layer thickness and defects in opaque spheres. The production of targets at high rates will utilize computerized sensing, characterization, and control of the processes. Effective use of modern techniques of material deposition will permit the rapid and accurate formation of uniform high-quality layers.

Driver power and energy levels which are as yet not achieved will be necessary for economical, power-producing laser fusion systems. New gas laser development is proceeding and may provide the "Brand X" system necessary for a working power reactor driver.

The ICF reactor of the future will be a well-integrated combination of a high power, high energy driver and a low cost, high quality target. In both driver and target development, the most difficult problems to be solved are in materials technology.

2. PELLET SPECIFICATIONS

There are several critical elements in the ICF process--the fuel, the fuel container, the driver, and the peripheral apparatus such as diagnostics and energy removal systems. We are concerned here primarily with the fuel and its surroundings. The fuel may be gaseous, liquid or solid depending on the particular experiments and the type of fuel being used. A number of fuel possibilities are:

Deuterium-tritium
Deuterium-deuterium
Boron 11-hydrogen
Lithium-deuterium-tritium

The first two, of course, are gases at normal temperatures, and liquids or solids at appropriate cryogenic temperatures. Boron-hydrogen compounds range from gases to liquids to solids at normal temperatures and range in composition from diborane (B_2H_6) which boils at -92.5 °C to decaborane ($B_{10}H_{14}$) which boils at +65°C. Lithium hydride is a solid which melts at 680°C.

The structure holding the fuel is more than just a container. All of the materials and their physical characteristics, masses, atomic numbers, and radii are important aspects of the target design. A few designs for experimental targets are shown in Fig. 1. Figure 2 is a more detailed set of specifications for a typical laser fusion pellet.

3. FABRICATION PROBLEMS

Some of the problems facing us in the production of ICF targets may be seen in the specifications in Fig. 2 and Table I. The fuel completely fills the inner shell at a density of 0.070 g/cc. Other target designs may require the DT fuel to be at liquid density (0.21 g/cc at 24 K). Both the filling of pellets with liquid fuel and containing the fuel will require the solution of difficult cryogenic problems. The pellets will have to be maintained at temperatures below 24 K to avoid disruptively high internal pressures.

TABLE I. Design specifications, tolerances, and other criteria to be met by glass spheres produced for use in laser fusion pellets.

1) Diameter range 50 μm-2000 μm
2) Thickness range 1 μm-20 μm
3) Sphericity better than 5%
4) Concentricity and wall uniformity better than 5%
5) Surface finish better than 2000 Å
6) Strength--able to contain up to 100 atmospheres of DT
7) Composition--variable

Commercial glass microspheres are unable to meet these criteria.

4. GLASS SHELL PRODUCTION

For some time in ICF research it has been the practice to produce glass shells of high quality then permeate DT gas through the shells at moderately high (350°-450°C) temperatures. These DT filled shells are used as the central spheres of the multilayer targets. The fuel density obtainable by this technique is limited by the temperature at which permeation occurs, fill system pressures and the temperature at which the target is used. Surface quality of the glass is required to be very high. Early laser fusion targets were made from commercial glass shells produced for other purposes. Because surface and wall uniformity were not important, no effort had been made to produce high surface quality. Table I shows the specifications which must be met by glass spheres utilized in current experiments. A comparison of previously used commercial spheres with those currently being used is shown in Fig. 3.

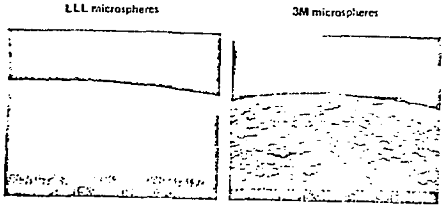


Fig. 3. Scanning electron microscope pictures of the surfaces of glass spheres taken from commercial sources (3M) and from batches of spheres produced at Lawrence Livermore Laboratory. The "tie" marks along the lower edges of the pictures are 1.0 μm apart and each mark is 0.1 μm wide.

Several techniques are being used for the production of high quality glass shells with a wide range of diameters and wall thicknesses. Our most successful method utilizes the generation of uniform droplets of an aqueous solution of glass-forming chemicals and subsequent drying and fusing into hollow glass shells. Droplets are formed by the break-up of liquid jets as first described by Rayleigh and developed more recently for special purposes including high-speed printing. The droplets of solution are initially passed through a moderate temperature (300°-500°C) region of a vertical column to remove the solvent. After passing through the drying region, the dry particles pass into a high temperature region where the material fuses to glass and the final shell is formed. Temperature and temperature gradients are important parameters as are droplet size, solute concentration, and chemical composition. A sample of hollow spheres directly from the furnaces is shown in Fig. 4. The process is sufficiently well controlled that in the batch shown almost 80% of the spheres meet the criteria indicated in Fig. 3 with regard to wall uniformity, sphericity, concentricity, size, and surface finish.

Many of our experiments require gases in the targets other than isotopes of hydrogen. For example, for diagnostic and other purposes we often need argon, krypton, neon or xenon in the shell together with the DT fuel gas. Other than neon, these gases do not diffuse readily through glass and hence are very difficult to put in after the spheres are formed. However, if the furnace column is filled with the required gas while the spheres are being formed, the spheres will contain the gas when they are completed. For example, we regularly fill our glass spheres with 0.1 to 0.3 atmospheres of argon during the production process. As needed, we also fill with krypton and xenon to about the same pressures. Fills of these and other gases are easily done at both lower and higher pressures.

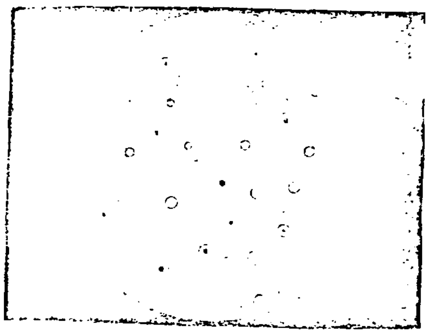


Fig. 4. An interference microscope photograph of a batch of spheres taken directly from the furnaces. About 80% of these spheres meet the diameter, wall thickness, sphericity, concentricity, and surface finish requirements for laser fusion pellet designs.

5. COATINGS AND LAYER DEPOSITION

The second layer of the multiple layered target may be a layer of polymeric material similar to Teflon. To deposit this material we chose to utilize a technique suggested by some work done by Jay Fries' group at Los Alamos. They had done some excellent work utilizing a parallel plate plasma discharge to activate a monomer which then polymerized on the surfaces of glass or metal shells. The LASL group had very good success with this method. However we were not able to get the process to produce layers to our satisfaction, so we decided to proceed along other lines. We examined both microwave and longer wavelength RF plasma discharge systems for activation of the monomers.

In our plasma polymerization process a mixture of a monomer gas (for example, perfluoro-2-butene, PFB) and argon is introduced into a helical, resonant transmission line tuned to about 13 megahertz. At appropriate power levels, a gas discharge is produced and maintained in the transmission line. The flow of the gases moves the activated material out the end of the transmission line into the region above a small dish in which the spheres to be coated are located. The dish is mounted on a piezoelectric element which is driven by an AC signal so the spheres roll and slide transversely over the surface of the dish, thus presenting their entire surfaces to the coating action of the activated material. The PFB polymerizes on the sphere surfaces in uniform layers. As with any coating technique there have been problems obtaining smooth surfaces and well-controlled layer thicknesses. The most severe problem was the surface quality which had to meet the 200-300 Å smoothness specifications as on the glass shells. By periodically bombarding the surfaces of the spheres with ions during the coating process, the growth of irregularities in the layers was avoided and the surfaces were made to the required smoothness.

The microwave-generated plasma deposition techniques also appear to have promise in a slightly different direction. While we have been able to put coatings of polymerizied materials on surfaces by this technique, we have also produced low density "foam" compositions which also have utility in target fabrication.

Looking back at Fig. 1, we see several other materials of interest to target designers. Beryllium is of particular interest because of its low atomic number (Z = 4), and its density (ρ = 1.85 gm/cc) and because it is a stable solid at room temperatures. However, beryllium poses some problems for us from a fabrication standpoint. It is considered to be very toxic; and it is, consequently, difficult to use. We have, however, deposited coatings of beryllium on glass hemishells. These hemishells can be assembled to form complete spherical shells which surround our DT filled glass spheres.

As has been the case with almost every material being deposited and every deposition process, initial efforts in the sputter deposition of beryllium yielded surfaces which were rough (on a few hundred Angstrom scale length). By controlling temperatures, sputtering power levels and gas pressure, we are able to produce "amorphous" beryllium layers with smooth surfaces. "Amorphous" in this sense means crystal structure is either absent or the crystals are small enough that no lines are found in x-ray diffraction studies of the material.

We see in Figs. 1 and 2 that many other materials may be needed for the fabrication of ICF pellets. High atomic number materials with high density are necessary in some designs. For example, a gold pusher may be needed as shown in Fig. 2. The second shell is a layer of gold about 35 μm in radius and about 7 μm thick. The inner and outer surfaces of the gold must have no peaks higher than 1000-2000 Å and must have a general ambient surface smoothness of 200-300 Å. That is, there should be at most only a few (five or six) of the 2000 Å peaks and the remaining surface features should have a peak-to-valley amplitude less than 300 Å. The severe surface quality criteria result from an attempt to avoid Rayleigh-Taylor instabilities during the implosion of the target. The fabrication problem arises from two aspects. The gold shell must be produced with the smooth surfaces required and the fuel must be put into the shell which is impermeable to the fuel. If holes or filling tubes are used, they must not introduce irregularities in the shell which exceed the smoothness specifications. Many variations are possible and indeed are often "negotiated" between pellet designers and the fabrication scientists. Such parameters as layer thickness, total mass in the layer, average atomic number, atomic number of each element present in the layer, and density are all important and must be considered together, not independently.

A material often shown in pellet designs is generically designated TACHO or CHOW. These are names given to hydrocarbons--often polyethylene--in which high-Z (such as tantalum or tunsten) materials have been dispersed. The average density may be still about that of the polyethylene while the average atomic number may be relatively high. Several techniques are used to produce the materials. The critical parameters for our purposes are the density, the average atomic number, the material uniformity on a scale of 10-100 Å, and its production and mechanical properties. One of the easiest (and probably least satisfactory) methods to produce TACHO is to disperse tantalum or tungsten particles in polyethylene. Dispersion of metal particles uniformly in the polyethylene is difficult. Local clumping of the metal is almost unavoidable even if particles of a few to tens of Angstroms could be obtained. The variations throughout the material would be unacceptable.

A more satisfactory means of obtaining appropriate materials of the TACHO type is by polymerization of organo-metallic compounds or by a combination of polymerization and co-deposition of a high-Z element such as tantalum, gold, tungsten, or uranium. It must be noted once again that surface and interface smoothness and layer uniformity are critical parameters.

As a substitute for the gold layer around the DT in the center of the pellet, we have prepared lead glass shells. These have been produced in a wide range of sizes and thicknesses with excellent concentricity, wall thickness uniformity, and surface finish. Our compositions include up to 75% by weight of lead oxide. A composition containing 50% by weight of PbO has a hydrogen permeability sufficiently high to permit filling by permeating DT through the glass wall. Thus, to fill the shells with DT at densities of a few tens of milligrams per cubic centimeter is almost as easy, in principle, as for the borosilicate, soda-lime, or other commonly-used glass shells.

6. PELLET FACTORY CONSIDERATIONS

Many of the problems and solutions discussed here have been related specifically to types of pellets to be used for research experiments leading to breakeven. However, many aspects of these same pellets are applicable to reactor pellets. It is highly likely that multilayer pellets will be required. It is clear that whatever the pellet configuration is, they must be produced at high rates (up to ten per second) at costs of 10 to 50 cents each. We have suggested techniques by which layered targets may be made cryogenically. Figure 5 shows how multilayer pellets can be made in large quantities.

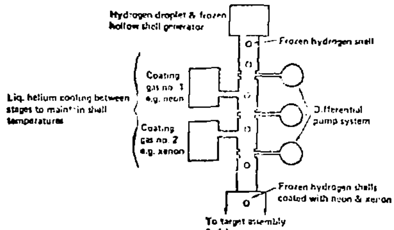


Fig. 5. A method of producing multilayer ICF pellets at high rates. The schematic diagram illustrates cryogenic pellets but the method is also applicable to other materials that need not be handled at low temperatures.

Many techniques which are currently "state-of-the-art" in other fields or in our present pellet fabrication work can be brought together to produce ICF pellets on a production basis. Pellets of glass, plastic, metal, and other material layers may be made by the techniques used for current production but in automated modes. To illustrate the successive steps in automated pellet production, let us first go back to the glass spheres. Our current production by the drop generator method produces batches in which 80% of the spheres coming from the furnace are the correct diameter, wall thickness, uniformity, and surface finish to meet the very stringent requirements set by our designers.

It is within the state-of-the-art to perform high-speed pulsed laser holography on the glass shells as they come from the furnaces. By computer processing it should be possible to check which two out of ten spheres would be rejected. (However, we think sphere production can be significantly improved to give yields even higher than we have presently.) It is possible to fill the pellets with fuel during the formation process by the method which has been described for the filling of glass shells with argon and other gases. It may be more desirable to collect the spheres from the furnaces in batches and fill the spheres in large numbers.

After the spheres are filled with fuel, they must be coated with layers of appropriate materials. The coatings can be applied in fluidized beds or on a single sphere basis. Fluidized bed techniques need more development to achieve the surface quality required. Molecular, atomic, or ion beam methods are suitable for single sphere coating. We have a ring quadrupole suspension system which works well with beam deposition techniques. Figure 6 is a schematic of a system adapted from one of our present operating support and transport structures which has a 30 cm diameter, and in which we are able to suspend pellets in stationary positions or move them from one location to another. The suspended pellets can be coated at each of the locations by different ion or molecular beams and thus multilayer pellets can be produced at high rates. Multiple suspension-coating stations can be used if necessary to provide higher rates of production.

7. CHARACTERIZATION

The development of techniques by which pellets can be produced to designer specifications must be coupled to the development of means to characterize the pellets. The most used characterization methods are: optical microscopy, interference microscopy

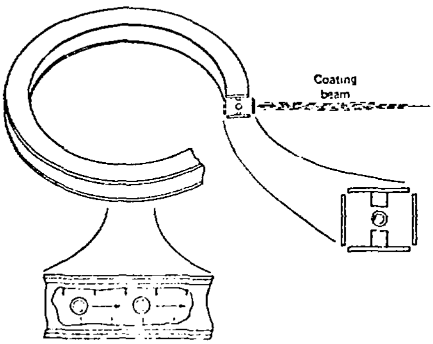


Fig. 6. A ba' n processor for ICF pellet production. C' ırged pellets are suspended in a dynamic el cric quadrupole field in a circular co' ıguration and are moved around the circle y pulsed voltages applied to electrode inserted between sections of the rail sys em. Coating beams of various materials are applied to the pellets through the spaces between the rails.

(including both transmission and reflection types), microradiography, and scanning electron microscop,. To provide pellets with the highest possible quality for implosion experiments, we must perform a complete quantitative study of the surfaces of every layer of every pellet and of the uniformity of every layer. An interference microscope study of a glass shell manipulated by hand can be very time consuming. This is particularly true at high magnification. As a result of the need for a more rapid handling system, we have developed a sphere manipulator which is controlled by a microprocessor which rotates the spheres through a prescribed pattern of rotation in the field of view of an interference microscope. The manipulator reduces the time for a full characterization of a sphere from several hours to a few minutes.

Since we also utilize the scanning electron microscope extensively, we needed a similar manipulator for the SEM. To produce a set of photographs of the entire surface a 150 μm-diameter sphere at a magnification such that one picture covers a 3 μm x 3 μm area on the sphere is a very time-consuming process. Each photograph take few minutes including time to manually manipulate the sphere to permit photography of a new area. We have automated the operation of the sphere manipulation and the SEM recording process so that a video tape of the entire surface of a sphere can be made by the SEM with no operator present. The recording of the complete surface of the sphere takes five to ten hours and provides resolution sufficient to detect 100 Å irregularities on the sphere surface.

In the process of producing a pellet for a laser fusion shot, every sphere and its coating layers are "4π" characterized to determine the presence of any defects. If there are many defects larger than about 300 Å, the sphere may be rejected. If only a few defects are present, say four or five defects which are 1000-2000 Å, the details of the irregularities are examined with the designers and a decision is made to accept or reject the pellet. The characterization is time consuming and expensive and more automated methods are coming on line very soon to cut the time for each operation and to provide computer printed contour maps of every sphere.

8. LASERS AND OTHER ICF PELLET IRRADIATORS

There are currently several operating systems for irradiating ICF pellets. Most of these are high power, high energy lasers. Some high voltage e-beam drivers are also operating as pellet drivers.

The major large laser systems are in the United States--the glass lasers at Livermore and Rochester and the gas lasers at Los Alamos. There are also several high power laser fusion research systems operating or under construction in other countries including the Soviet Union, France, Great Britain, Japan, and China.

There are high voltage e-beam driver systems being used primarily for fusion studies at Sandia and in the Soviet Union.

The development of drivers for irradiation of pellets to study inertial confinement fusion is an on-going process as is the development of pellet fabrication. Laser development is being done at several laboratories in the U.S. and in several other countries. High repetition rate glass lasers, CO_2, HF, KrF, and other gas systems are being studied toward the goal of producing the "Brand-X" laser suitable for reactor applications. High voltage e-beam machines, light ions, and heavy ions are also being studied as drivers for power system ICF pellets. The development of suitable drivers for achievement of breakeven, for research into the physics of implosion processes, and for future reactor systems will require material and scientific p rsonnel resources and much ingenuity an creativity. In the glass laser field, there have been significant advances in materials development. Phosphate and fluoroberyllate glasses have been improved and in many cases have beneficially replaced silicate glasses in laser systems. Energy and power delivered to the pellet to achieve a breakeven implosion are in the 100-400 KJ, 100-400 TW range. There are currently no driver systems operating which are capable of delivering these high powers and energies to a pellet. To avoid lens, mirror, and amplifier damage in laser systems as energy and power are increased, beam aperatures must be increased. The production of optical elements at large aperature is very expensive and technically very difficult. To use many parallel beams to achieve high energy and high power leads to problems of alignment, aiming, focusing, and reliability which are all present in single beam chains and obviously increase as more beams are added.

It should not be assumed that a change of driver type will eliminate all the problems. Ion beam systems and e-beam systems have their own peculiar problems which must be solved before they can be considered as the reactor drivers of the future.

9. CONCLUSION

The conclusion to which we come as we study the problems of pellets and drivers--optical or otherwise--is that the tasks are formidable! Pellet fabrication is not a trivial task as implied by some reports. It is also not the impossible task implied by others. We see no fundamental barriers to successful pellet fabrication. However, to produce some of the more complex designs will be technologically very difficult and will require some very good physics, chemistry, engineering, and material science developments.

Driver technology must improve to reach the energy, power levels, and repetition rates necessary to achieve breakeven and subsequently reactor class driver output. With sufficient committment of resources and dedication by creative scientists, the goals of inertial confinement fusion can be reached.