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Abstract

Thin films of silicon have been formed using a patented electrostatic deposition method which utilizes charged particle motion in an electric field. After deposition, the films are heat treated for varying times and temperatures in a programmable furnace maintained under a purified argon atmosphere. X-ray diffraction (XRD) confirmed that these films were polycrystalline in nature. These films were found to have grain sizes of about 50 microns. Solar cells were fabricated using these large-grained polycrystalline silicon films by sputtering pure gold as both front and back contacts. The cells have shown efficiencies of 1.8%. This paper reports on the growth of these large-grained polycrystalline thin silicon films and on the laser recrystallization setup to be used to increase the grain size up to 100 microns. Films grown via this electrostatic deposition method and subsequent laser recrystallization have a great potential for use in the solar cell industry.

Introduction

Single crystal silicon solar cells are the best known photovoltaic energy converter. Solar cell arrays have been used extensively to supply electric power in spacecraft for many years. The steps involved in the fabrication of these cells are very energy, labor, and material intensive (Chu et al., 1975). Also, the present cost of single crystal silicon solar cells makes their use prohibitively expensive except for applications in space vehicles and as power sources in certain remote areas. (Fan and Zeiger, 1975).

To reduce the material and processing costs, one approach involves the deposition of a thin layer of polycrystalline silicon containing a p-n junction on a suitable substrate (Chu and Singh, 1976). Polycrystalline silicon has great potential for large scale terrestrial photovoltaic device applications. Techniques for preparing polycrystalline silicon are, in general, less critical than those required to produce single-crystal silicon. Also, polycrystalline silicon shows great promise for reducing costs by circumventing many of the complex and energy-intensive steps associated with the growth of single crystals.

An electrostatic deposition process which utilizes charged particle motion in an electric field has been successfully developed as a method for the deposition of silicon films. (Gadepally et al., 1990). In their research, silicon films (coatings) were deposited on conducting, insulating, and semiconducting surfaces after making suitable changes in deposition parameters. These coatings subsequently were heat treated to establish an alternative method to generate polycrystalline electronic-grade silicon with grain sizes from four to ten microns.

This electrostatic thin film deposition technique can offer significant cost reduction in the growth of polycrystalline silicon thin films relative to conventional deposition methods such as sputtering, chemical vapor deposition (CVD), and molecular beam epitaxy.

Several experimental and theoretical results have indicated that there is an increase in efficiency of polycrystalline silicon solar cells with increasing grain sizes (Ghosh et al., 1979). It is generally accepted that grains of at least 100 microns are necessary to make polycrystalline silicon solar cells having efficiencies > 10% (Ghosh et al., 1979; Ouwens and Heijligers, 1975). If a high-power laser beam is focused on the surface of a silicon film that absorbs at the laser frequency, the film can be locally heated to temperatures high enough to cause crystallization (Fan and Zeiger, 1975). This paper reports on the growth of large grained (> 50 microns < 100 microns) polycrystalline silicon films using the electrostatic deposition method suitable for solar cell applications and on the laser recrystallization setup to be used to increase the grain size up to 100 microns.

Materials and Methods

Thin films of silicon were obtained by depositing 100 mesh electronic-grade silicon powder (purity = 98%) supplied by Cerac Inc. on single crystal silicon substrates supplied by MEMC Electronic Materials, Inc. P and n doped powders doped with boron and phosphorous (dopant level = 1×10^{15} atoms / cm^3 - 1×10^{17} atoms / cm^3) were used. Immediately prior to the deposition, the substrates were given a standard wash in a formic acid - hydrogen peroxide mixture (7:3), rinsed in deionized water, and transferred to 48% hydrofluoric acid for one minute to remove all traces of oxide from the surface. They then were given a prolonged wash in deionized water and finally blown dry using pure nitrogen (Lillington and Townsend, 1976). The powders were deposited using the electrostatic spray gun at Ameron Powder Coatings in Little Rock, Arkansas. As illustrated in Fig. 1, the silicon powder "S" in particulate form is stored in a hopper that is secured to the top of the gun. The gun has an electrode located at the front end, which is connected to a high voltage generator via an electrical line passing through the gun body. Pressurized dry air is driven through the gun which facilitates the transportation and charging of the silicon particles. The powder "S" is delivered to the gun nozzle by both gravity and a Venturi effect as seen in the detail. When the trigger of the gun is actuated, the silicon powder that is stored in the hopper is drawn into the hopper-Venturi feeder. When the exterior opening of the gun is directed toward a grounded substrate, electric field lines form between the tip of the electrode, extending from the corona region, and the grounded substrate. The charged silicon particles follow the electric field lines, resulting in the deposition of the particles onto the grounded substrate.

The powders were deposited at gun voltages ranging from 60 kV to 80 kV. The input pressure of the gun was varied from 6 bar to 12 bar. The deposition time varied between 2 and 5 seconds. The powders were deposited on both p-doped (resistivity: 1-3 ohm-cm) and n-doped (resistivity: 1-4 ohm-cm) single crystal silicon substrates. The configurations obtained were p/n, p/p, n/p, and n/n.

Heat treatment of these deposited films was carried out in a programmable Lindberg Model 51333 furnace (maximum temperature of 1500°C) for varying times and at different temperatures. The heat treatment times varied from 1 hour to 24 hours and the temperature varied from 600° C to 1370° C. In order to minimize any oxide formation, a filtered purified argon atmosphere was maintained within the furnace, which was modified for the heat treatment as shown in Fig. 2. The opening to the furnace was plugged with an ash and sali insulating material obtained from Zircar Products, Inc.. A hole was drilled through the center of the insulating material to accommodate an alumina process tube which held the samples to be heated. A removable flange having two ports was attached to the process tube. One port was connected to a gas cylinder containing argon; whereas the other port was connected to a vacuum pump. A

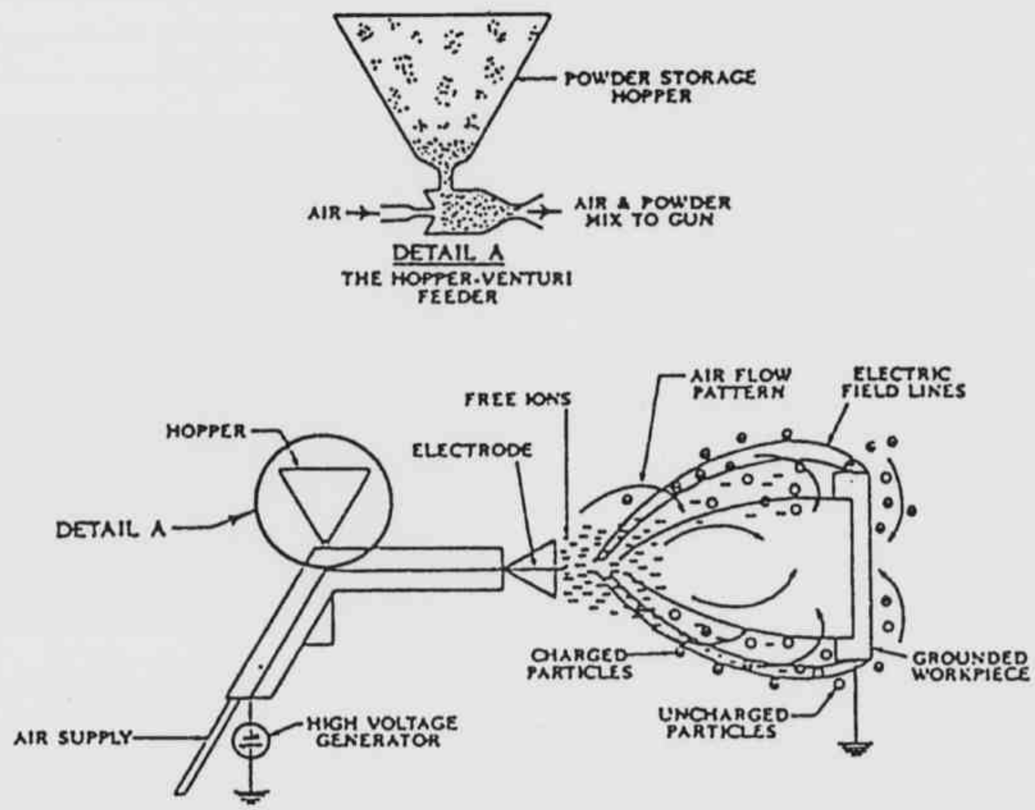

Fig. 1. Schematic of electrostatic spray gun.

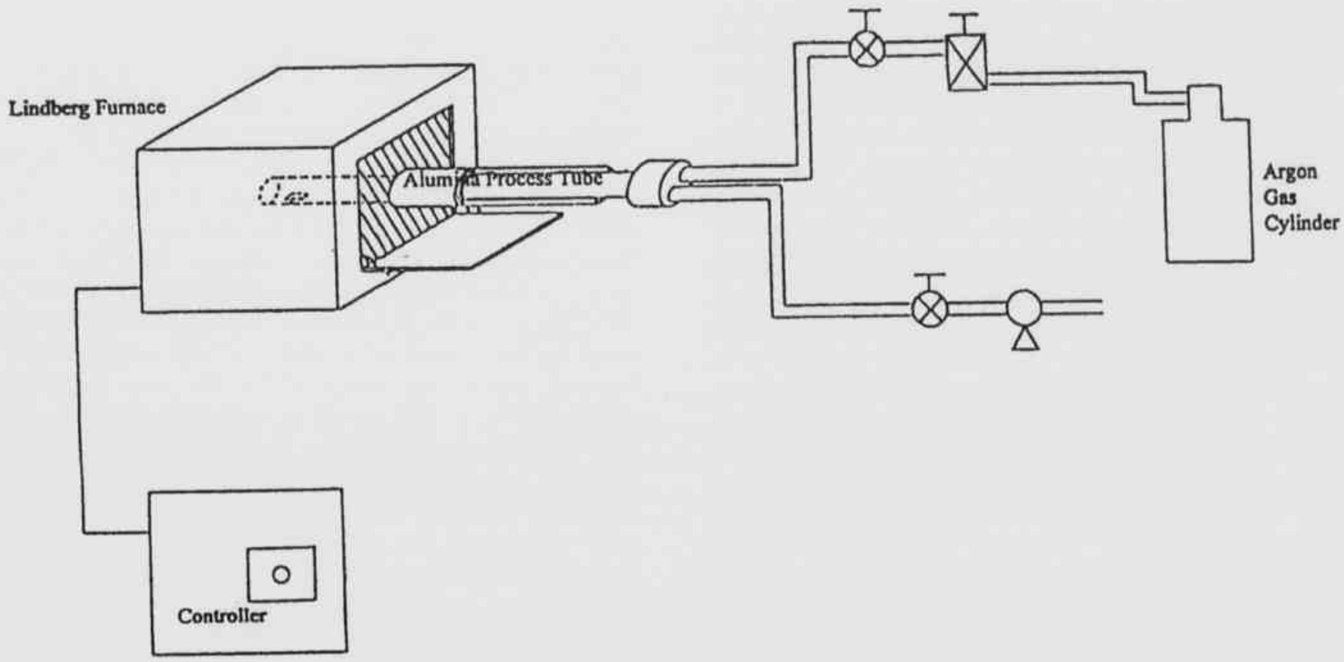

Fig. 2. Schematic of heat treatment apparatus.

Lindberg Model 58125 (818P) controller for regulating the furnace was connected to the furnace via cable wires. Prior to heat treatment, the vacuum pump was turned on to remove all traces of air from the process tube.

The primary recrystallization temperature, which is defined as the temperature at which 98% of the material is recrystallized within 0.5 hours (Ouwens and Heijligers, 1975) was found to be about 1200° C. It must be noted that no further change was observed at that temperature even if the samples were heat treated for more than 24 hours; these films were assessed using a Cambridge Stereoscan 600 Scanning Electron Microscope (SEM). Average grain diameters were found to be about 20 microns. Secondary recrystallization was observed from 1350°C upwards. As usual with secondary recrystallization, when samples are heated for a relatively short time, big grains amidst smaller grains are observed. The big grains are about 60 microns, and the smaller ones about 20 microns (Figs. 3a and 3b). The optimum time and temperature were found to be 10 hours and 1370°C, respectively. The film thickness was found to increase with the increase in the input pressure of the gun. Films deposited at an input pressure of 6 bar had a thickness of around 30 microns, whereas films deposited at an input pressure of 12 bar had a thickness of about 60 microns for a deposition time of 2 seconds. Structure and phase determination was done via X-Ray Diffraction (XRD) using a Philips Model PW 3710 X-Ray diffractometer. A Cu Kα radiation operating at 45 kV and 40 mA was used. The

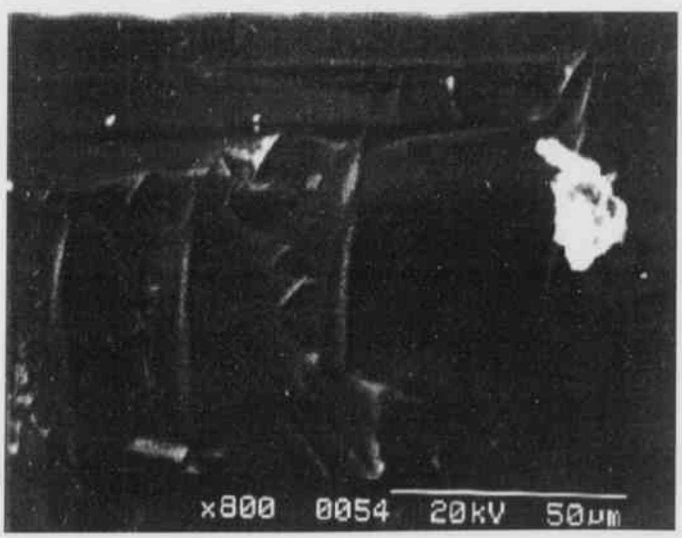

Fig. 3a. SEM photograph of a n/p sample showing average grain size. T = 1370°C, t = 10 hours.

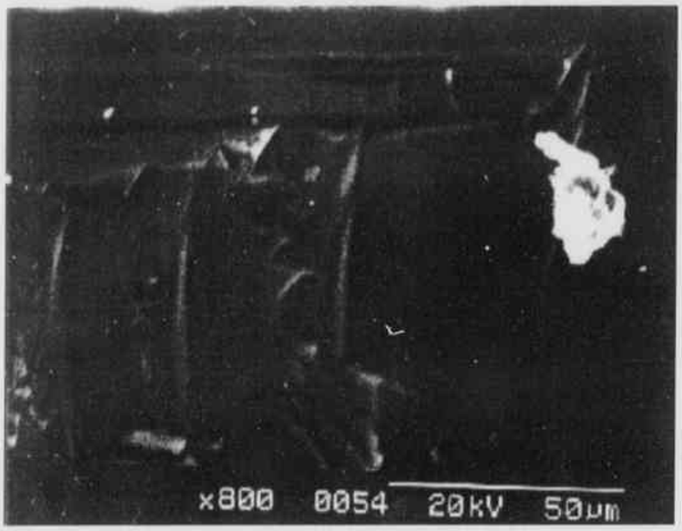

Fig. 3b. SEM photograph of a p/n sample showing average grain size. T = 1370°C, t = 10 hours.

wavelength used was 1.5406 Angstroms. The Bragg angle (θ) was fixed at 12° and the detector (2θ) was scanned from 25° to 90°. The XRD spectrums (Figs. 4 and 5) confirmed the film which was formed to be polycrystalline in nature with sharp <111>, <220>, and <311> peaks. The <111> phase was the most dominant phase.

Solar cells were fabricated by sputtering pure gold as both front and back contacts on the cells having the configurations p/n and n/p. The dark and illuminated I-V characteristics (Fig. 6) of these cells were analyzed using a Tektronix Model 576 curve tracer. The open circuit voltage was found to be about 320 mV, the short-circuit current was found to be about 9 mA. The conversion efficiency was calculated using the standard equation given by

$$\eta = \frac{V_{oc}.I_{sc}.FF}{P_{in}} \times 100,$$

where,

η = Efficiency,
V_{oc} = Open-Circuit Voltage,
I_{sc} = Short-Circuit Current,
FF = Fill Factor, and
P_{in} = Input Power.

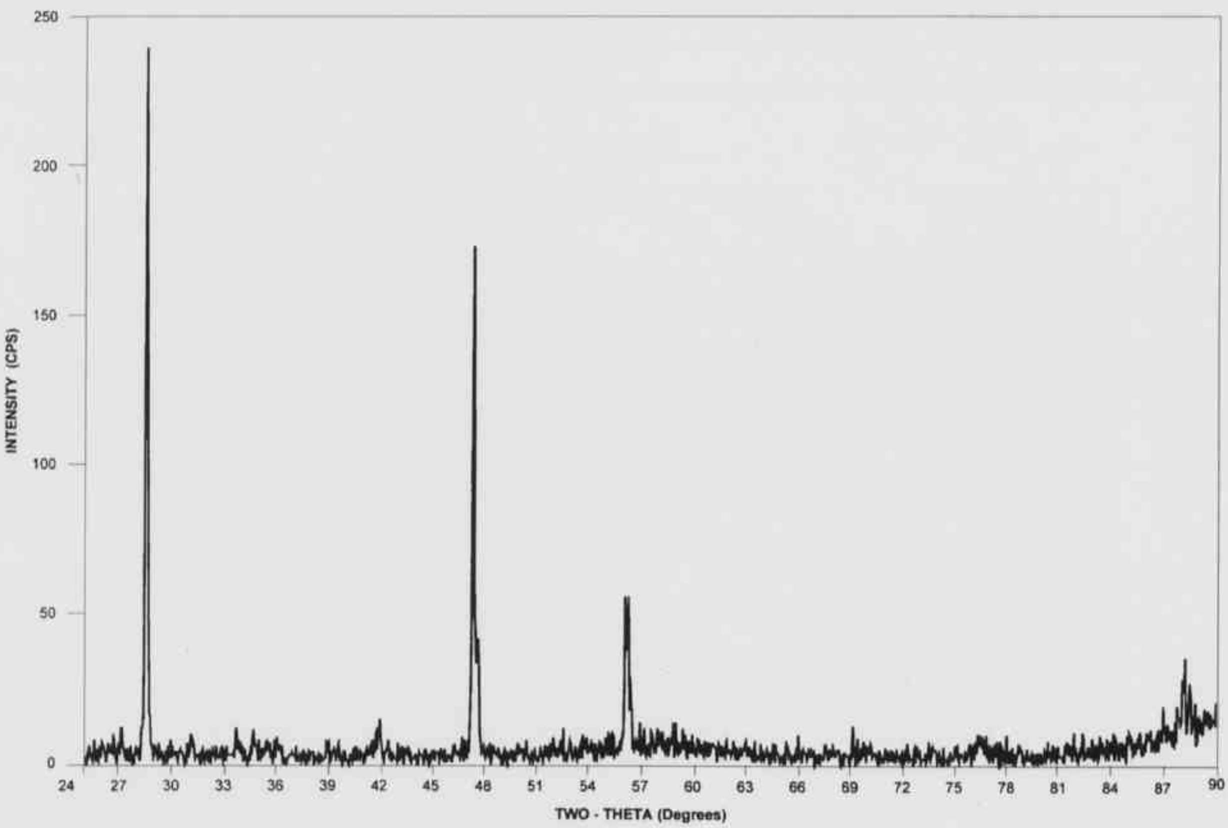

Fig. 4. XRD spectrum of n/p sample. T = 1370°C, t = 10 hours.

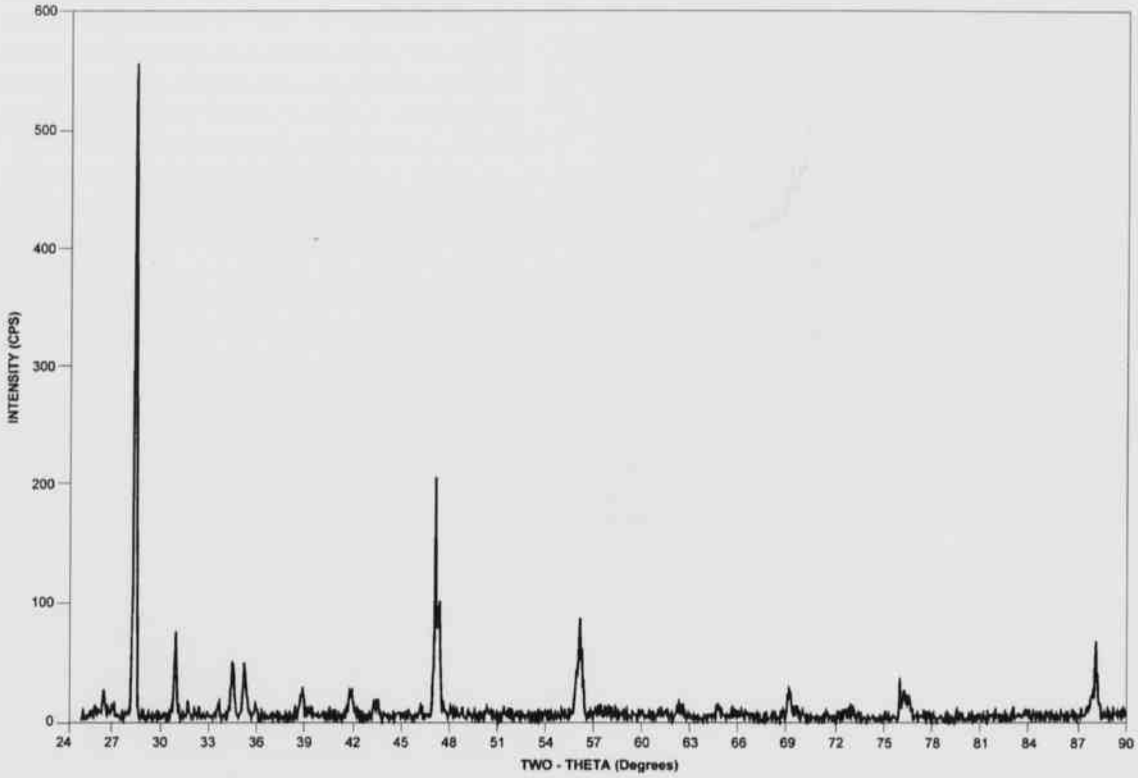

Fig. 5. XRD spectrum of p/n sample. T = 1370°C, t = 10 hours.

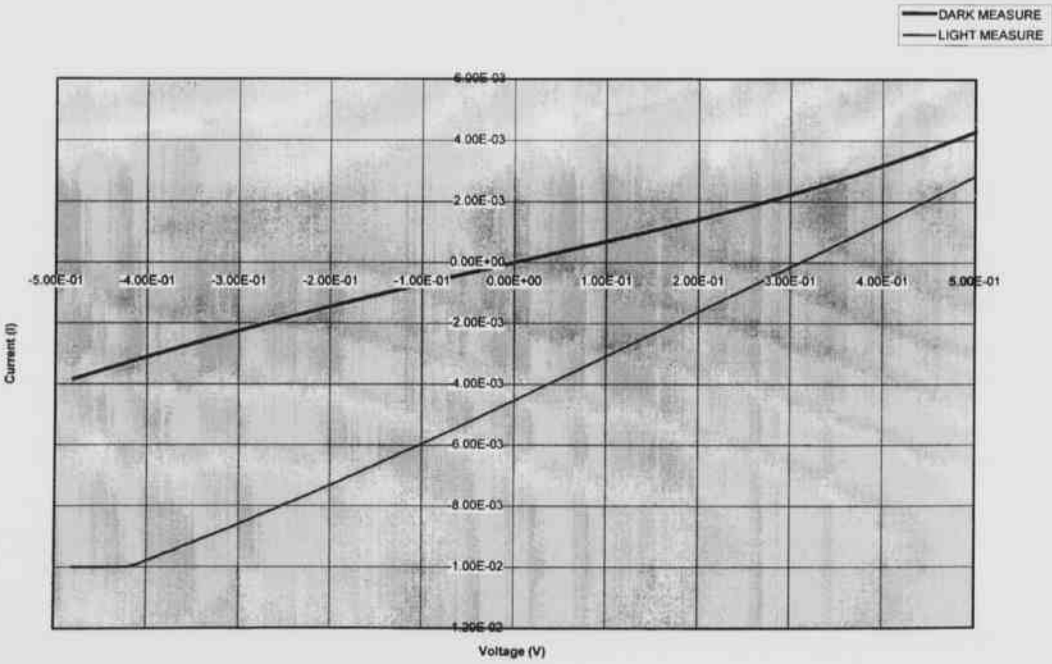

Fig. 6. Dark and illuminated I-V characteristics of fabricated solar cell. 5 mA per vertical division; 100 mV per horizontal division.

The efficiency was found to be about 1.8%. This efficiency can be improved by increasing the grain size to about 100 μm (Ghosh et al., 1979; Ouwens and Heijligers, 1975).

Laser recrystallization produces crystallites whose structure and electrical characteristics vary according to the starting material and the laser scan parameters. Generally, the crystallites nucleate in the surface region and are randomly oriented. A translatable stepper-motor-controlled vacuum stage was designed and constructed for use during laser recrystallization. A CW argon ion laser (Optilase Contact Laser System:Model 900) having total output power of 12 watts and operating at a wavelength of 488 nm is being used.

The sample during the laser recrystallization process will be mounted on a substrate holder made up of tantalum (Fig. 7). The tantalum foil is mounted on a 6 mm thick Macor ceramic block. The substrate holder is enclosed in an aluminum housing. In order to minimize any oxidation, an argon atmosphere will be maintained within the housing.

The substrate will be held at a temperature of about 450° C by heating the tantalum foil using brass strips connected to the foil.

The aluminum housing with the substrate holder will be mounted on the stepper motor controlled x-y stage. Stepper motors operating at 12 V DC, .16 A, and capable of producing 1.8° / step are being used. A complete block diagram of the laser recrystallization setup is shown in Figure 8. Since the optimal conditions for the recrystallization process were not known at the outset, a versatile computer program was written to drive the motors. The x-scan rate using these motors can be varied from 0.1 second to 2 seconds. The sample is scanned linearly which produces overlap regions where non-uniform growth of crystallites may occur.

The next phase of the research will be concentrated on finding the optimum conditions for the laser recrystallization including the laser power, scan rate, and substrate temperature.

Conclusions

Polycrystalline silicon films having grain size > 50 microns have been successfully formed using the electrostatic deposition method. These films were all observed to form at and above a temperature of 1350°C with a constant heat treatment time of 10 hours. Solar cells having efficiencies of 1.8% have been fabricated using these films. Laser recrystallization of these films will be used to increase the grain size up to 100 microns. This fabrication procedure holds great potential in the solar cell industry. This method has the potential to produce large area terrestrial devices at greatly reduced costs.

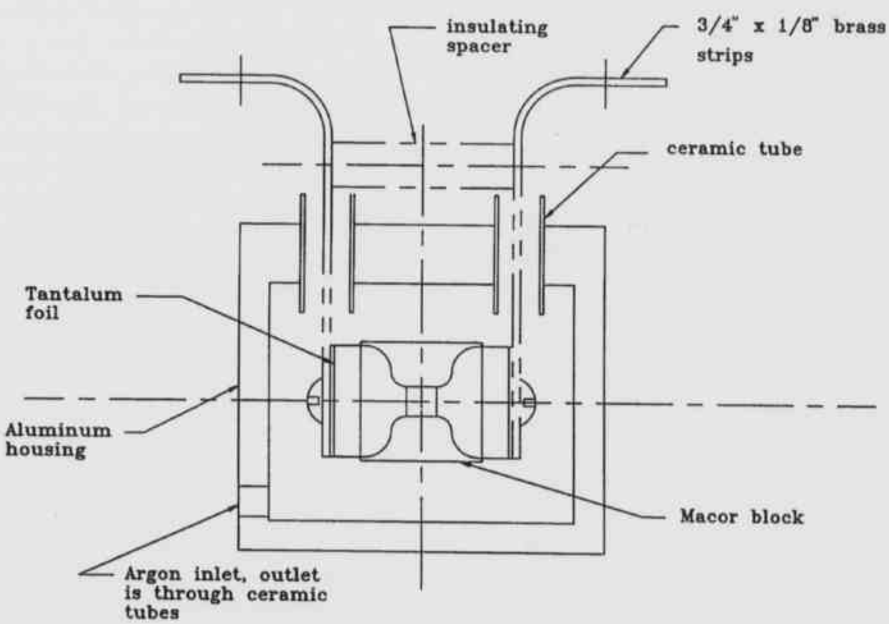

Fig. 7. Diagram of substrate holder.

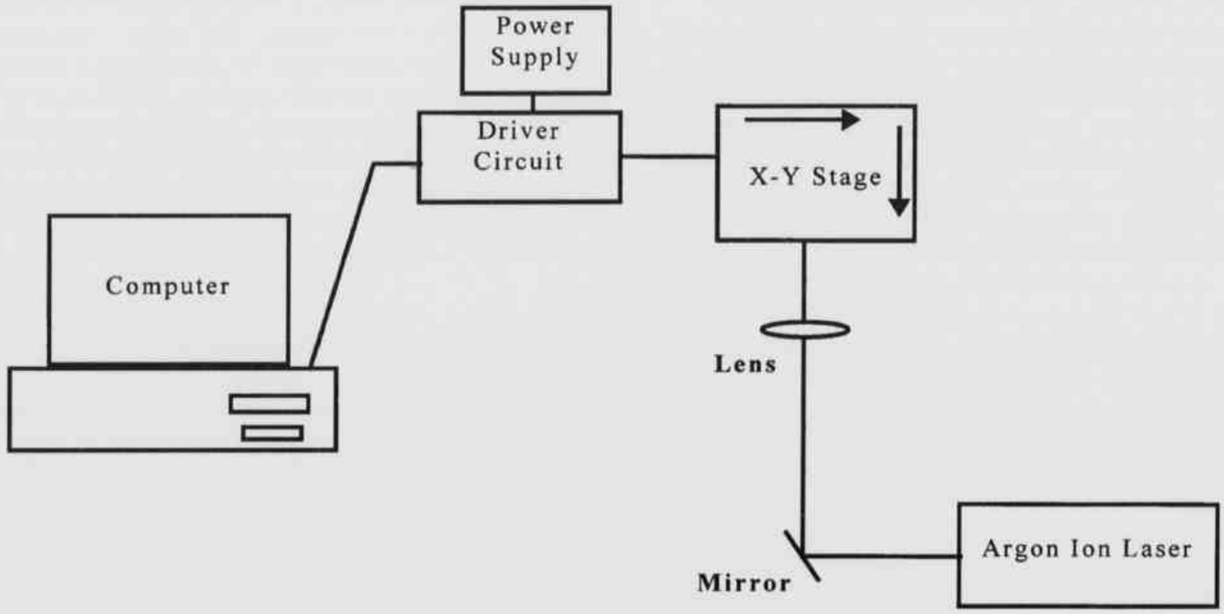

Fig. 8. Laser recrystallization setup.

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