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Laser Induced Synthesis of Silicon Carbide

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Abstract

Silicon carbide powder was prepared from ethylene and silicon using the 10.6 μm radiation from a TEA CO_2 laser .Different pulse energies of laser and ethylene flow rates were examined .The products were identified and tested via X-ray , FTIR and particle size techniques.

التحضير المحتث بالليزر لكاربيد السليكون

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الملخص

تم تحضير كاربيد السيليكون من ألأثيلين والسيليكون باستعمال ليزر ثاني اوكسيد الكاربون ذو الطول الموجي 10.6 نانومتر بأستعمال نبضات طاقية وسرعة جريان مختلفة للأثيلين وشخصت النواتج بواسطة طيف حيود ألأشعة السينية وألأشعة تحت الحمراء وتم قياس حجمها الحبيبي.

Introduction

Laser induced chemistry includes various types of reaction. Although some reactions are stimulated by UV/visible radiation, there are many other reactions that involve infra-red multiphoton excitation and novel laser-induced chemistry [1]. This is the case for the preparation of compounds of new characteristics especially in the ceramics field. One of the important areas in which laser methods have proven potential is the development of new methods for the chemical synthesis of powders. Ceramic inorganic powders with a high degree of thermal stability show much promise for application in mechanical, electronic and chemical engineering and have been prepared by such new laser methods [1].

There are several methods that are used to produce a ceramic powder including plasma, laser and oven. As far as the laser methods are concerned the wavelength of the laser is a characteristic parameter. The CO_2 laser can be considered as the most suitable and available laser for preparation of powders because its wavelength is compatible with an infra-red absorption band for many molecules such as, SiH_4 ,C_2H_4, NH_3 …….etc: in addition, it is commercially available with high efficiency and a suitable output power.

One of the most important ceramic powders is silicon carbide SiC which has important uses in grinding and cutting because of its abrasive power, hardness and stability. Silicon carbide exists as α, SiC of which 18 hexagonal types and 23 rhombohedra types are known and β, SiC, which is cubic with a pseudo – diamond structure. Most of the commercial silicon carbide is hexagonal. i.e. α-SiC and is black with a greenish. The β, SiC is produced at very high temperature in vacuum as transparent light yellow crystals. Both the α and β-modifications are colorless if they are absolutely pure [2 ,3].

Cannon et al discuss a novel process [4] in which (SiC) powder was produced by a CO_2 laser. L.sun .et al-[5] studied the deposition of SiC from tetramethyl silane using a CO_2 laser whereas the effect of deposition parameter were investigated by Crocker and Shaw [6]

The aim f the present investigation is to prepare and characterize the SiC powder using the TEA CO2 laser and study the effect of preparation parameters.

Experimental Details

The light source

A commercial TEA CO_2 laser system was used in this investigation. . The maximum pulse energy was 1 J and the pulse duration 100 ns .A facility for changing the gas pressure ratio provided a control on the output energy and the pulse shape .A diffraction grating blazed for the 10.6 μm radiation allowing for line emission selection was used. To observe the output laser lines a CO_2 spectrum analyzer (model 16 A Optical Engineering) was used.

Reaction chamber

The chamber which was used in this work was 400 mm in diameter and 450 mm in height. It was made from brass and had two windows orthogonal to the nozzle axes and with the germanium windows of 25 mm diameter. The laser beam was focused by a molybdenum concave mirror of 500 mm focal length. The focused beam was alligned to the reaction zone directly under the nozzle mouth .The opposite side of the nozzle was joined to the filter ,which was made of stainless steel .While the opposite side of the window group consists of a laser beam reflector about 1m from the chamber side .The experimental setup can be shown in figure (1). The vacuum system consisted of a rotary pump type (Alcaleel) joined to the other side of the filter to evacuate the system in order to obtain the required pressure. The pressure inside the chamber was measured by a Pirani gauge type 1001 and a pressure sensor type PRM 10 was used, both manufactured by the Edwards company.

The materials

Ethylene gas: ethylene gas, which was used in the preparation of SiC, was provided by the General Company for Petrochemical Industries, Basrah with a purity of (99.9%).

Argon gas: A high purity argon gas (99.999%) was used as an inert gas because of its being transparent to the 10.6 μm wavelength.

Silicon powder: Silicon powder with a mesh of 23 produced by the Prolabo Company was used as silicon source for SiC synthesis. Its purity was 99.9%.

Synthesis of SiC

The chamber was evacuated first to 10^{-3} mbar and flushed with Ar gas and then the mixture of ethylene and silicon powder was pumped through the nozzle with different flow rates .The pressure inside the chamber was maintained at 200 mbar. The reactants entering the vacuum chamber were exposed to 10.6 μm laser radiation of different pulse energies for 60 seconds. The product was collected on special filter paper

type (A/E).Then it was heated at 800 **°C** for 1hr.The unreacted silicon was removed by treating the products with a mixture of 3.5:5 of HF solution and concentrated HNO_3 [7].The insoluble SiC was then filtered off and then rinsed and washed several times. Finally the isolated powder was dried at 200 **°C** [8].

Analysis and characterization

X-ray diffraction

The x-ray diffraction technique was used for the identification of the crystal structure and the modification of SiC samples .A Philips diffractometer type PW-1840 with Cu-K α-radiation and Ni filter was used. The speed of the goniometer and chart was 1degree/min.

Particle size distribution and surface area measurements.

The particle size distribution was obtained using Fritsch particle seizeanalysette 22.It is capable of recording and calculating the particle size of the sample as a histogram and then calculating the surface area of the powder.

The FTIR technique

Shimadzu FTIR spectrometer model 8300 was used to identify the nature of the chemical bond and hence the type of the compound .It has a resolution of 4 cm^{-1}.

Results and Discussion

In order to study the effect of the ethylene/silicon ratio and pulse energy on the yield obtained and chemical structure and shape of the SiC, a set of experiments were done with different flow rates of ethylene and pulse energies: the results are shown in table (1).

Table(1) .Yield of SiC obtained at different flow rate of C_2H_4 and different pulse energies

Flow rate of C_2H_4 liter / min	Yield of SiC at 1 J (gm)	Yield of SiC at 0.850 J (gm)	Yield of SiC at 0.700 J (gm)
2.25	0.050	0.030	0.010
3.34	0.070	0.038	0.017
4.42	0.138	0.075	0.050
5.60	0.110	0.079	0.066

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It is clear that the yield of SiC increases with increasing pulse energy for all the flow rates studied, also the yield increases with increased flow rate of ethylene in the three pulse energies tested except the 1 J and 5.6 liter/min flow rate where the yield declined: this is due to the saturation in absorption [9-10].

The FTIR spectra of three samples prepared at three different energies are shown in figures (2, 3, and 4). Two samples show the presence of oxygen as identified by the peaks at 1089 and 1166.9 cm^{-1} representing the Si-O stretching vibrations in the Si-O-Si bonds [11] while the third sample does not show these peaks. It was observed also that only one sample contains hydrogen in addition to oxygen identified by a peak at 2287.4 cm^{-1}, for $H\text{-}SiO_xC_x$.

The identification of the Si-C bond in SiC appears in the three samples at 833.2 cm^{-1} 837 cm^{-1} respectively [12].

Among the other important vibration frequencies are those representing the first harmonic of the Si-C vibration at (1652.9 cm,$^{-1}$1622 cm^{-1} ,1521.7 cm^{-1} ,1649 cm^{-1} , and 1396 cm^{-1} , and broad bands at 1512.1cm^{-1} and 1651 cm^{-1}.

The x-ray diffraction analysis [13] of three chosen sample prepared at different energies demonstrates the presence of β- SiC as in the major peaks in figures (5,6,7) θ = 35.6, 35.5 and 35.8 respectively and two small peaks appear at (θ = 4.9 ,38.2) (θ = 41.3 , 59.9) and (θ = 41.4 , 59.9) respectively. The α. SiC is characterized by the presence of two peaks at (θ = 34.9, 38.2), (33.5, 37.7) and (34.0, 38.2) for the three samples. Finally the medium peaks at (θ= 47.3, 28.4) and (47.3, 28.6) reflects the presence of minor amount of silicon in samples two and three.

The examination of the particle size distribution indicates that the sample prepared at 700 mJ contains a smaller particle size than that prepared at 850 mJ as in the figures (8) where 40 % of the produced powder has a particle size of less than 2 μm and 10 μm respectively. There are also larger particle sizes in the 850 mJ sample .This phenomenon of nucleation was expected because the dissociated product includes solid materials which acts as mother nuclei for growth to larger particles.

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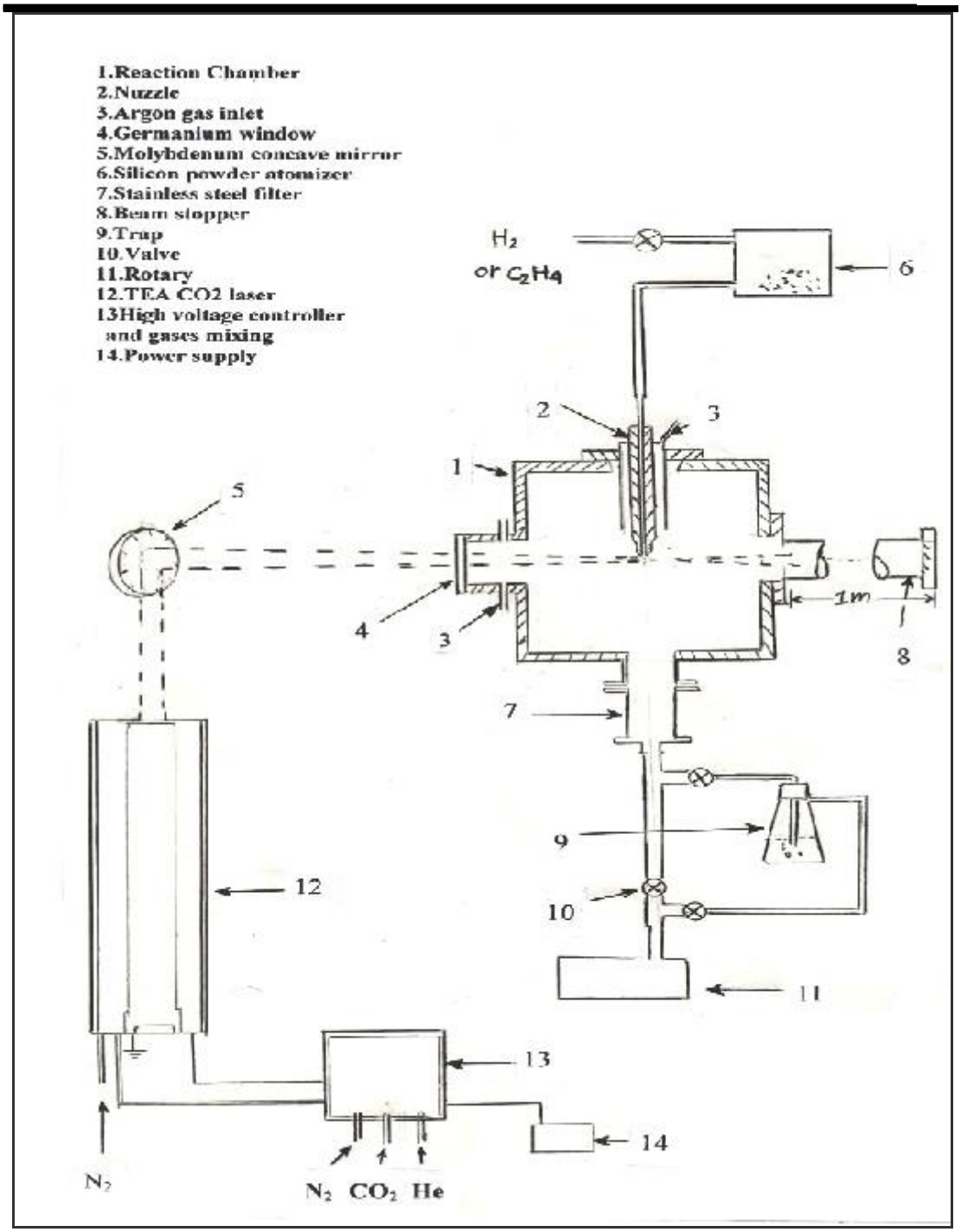

Figure (1) The experimental set-up

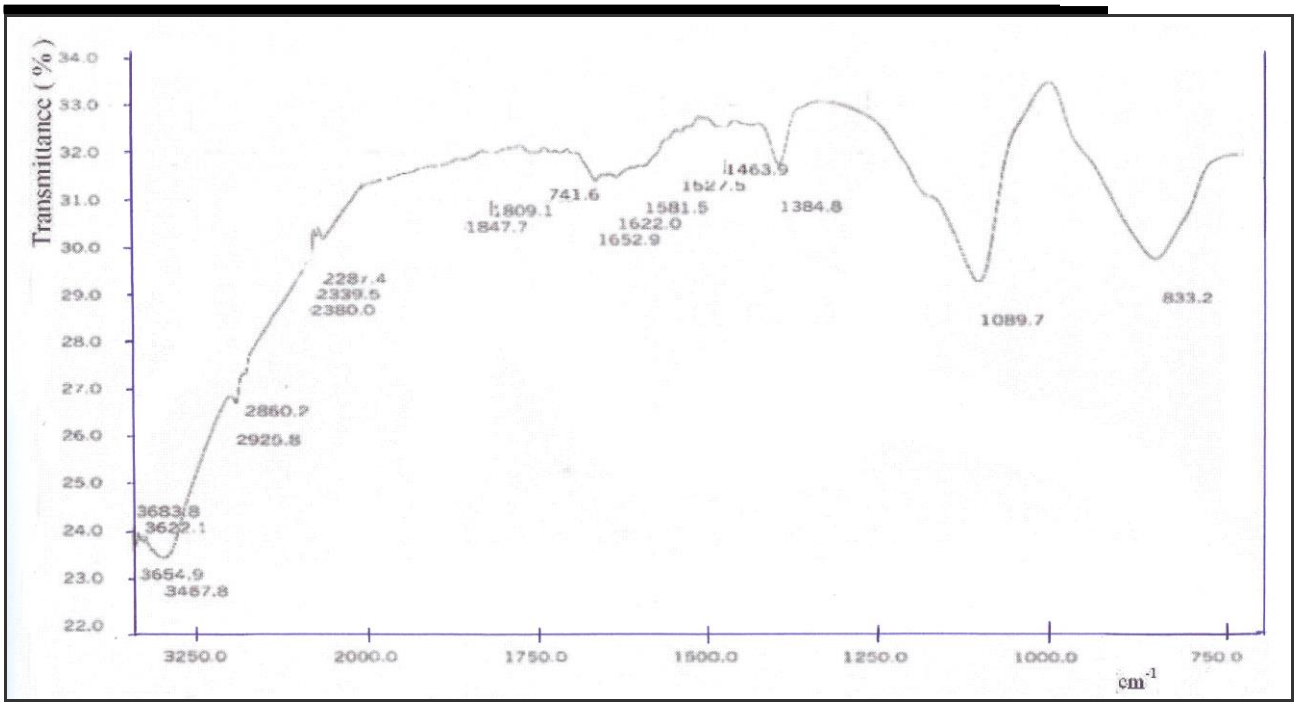

Figure (2) the FT-IR Spectrum of SiC prepared at 1 J/pulse

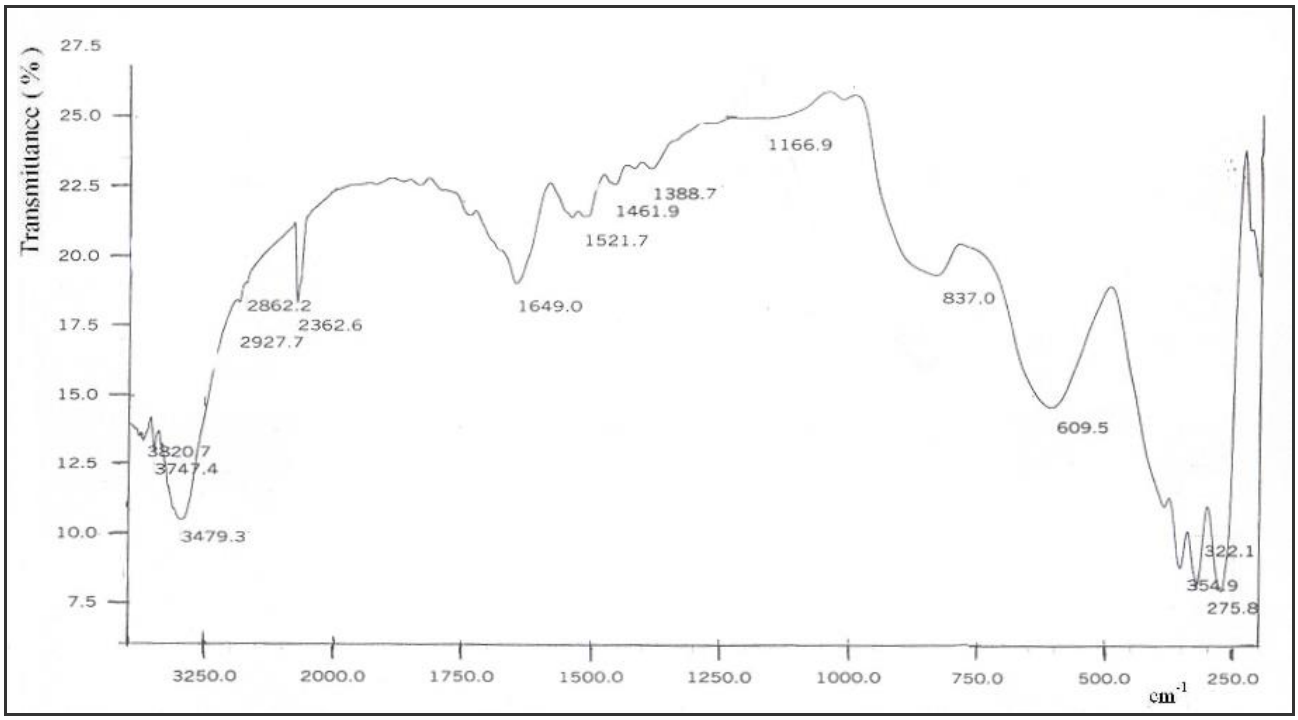

Figure (3) the FT-IR Spectrum of SiC prepared at 850 mJ/pulse

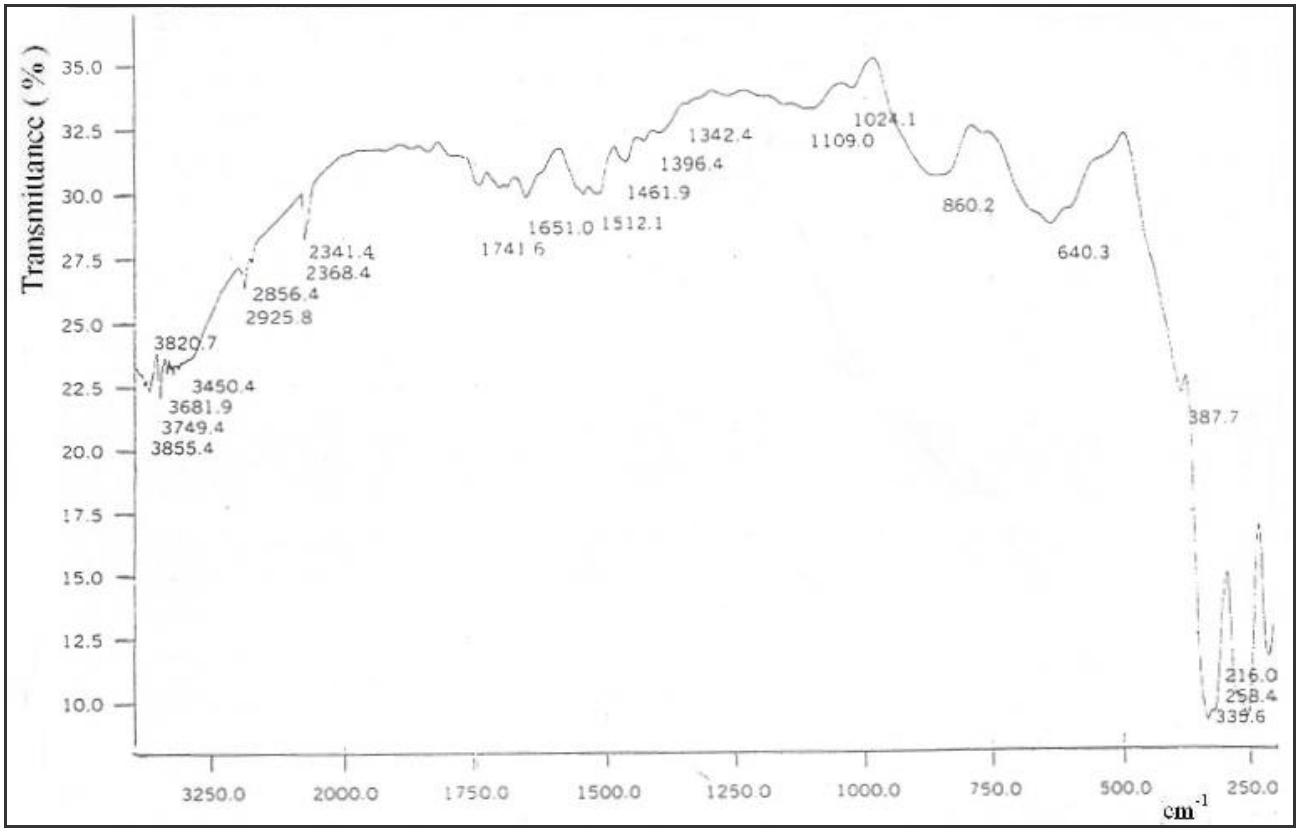

Figure (4) the FT-IR Spectrum of SiC prepared at 700 mJ/pulse

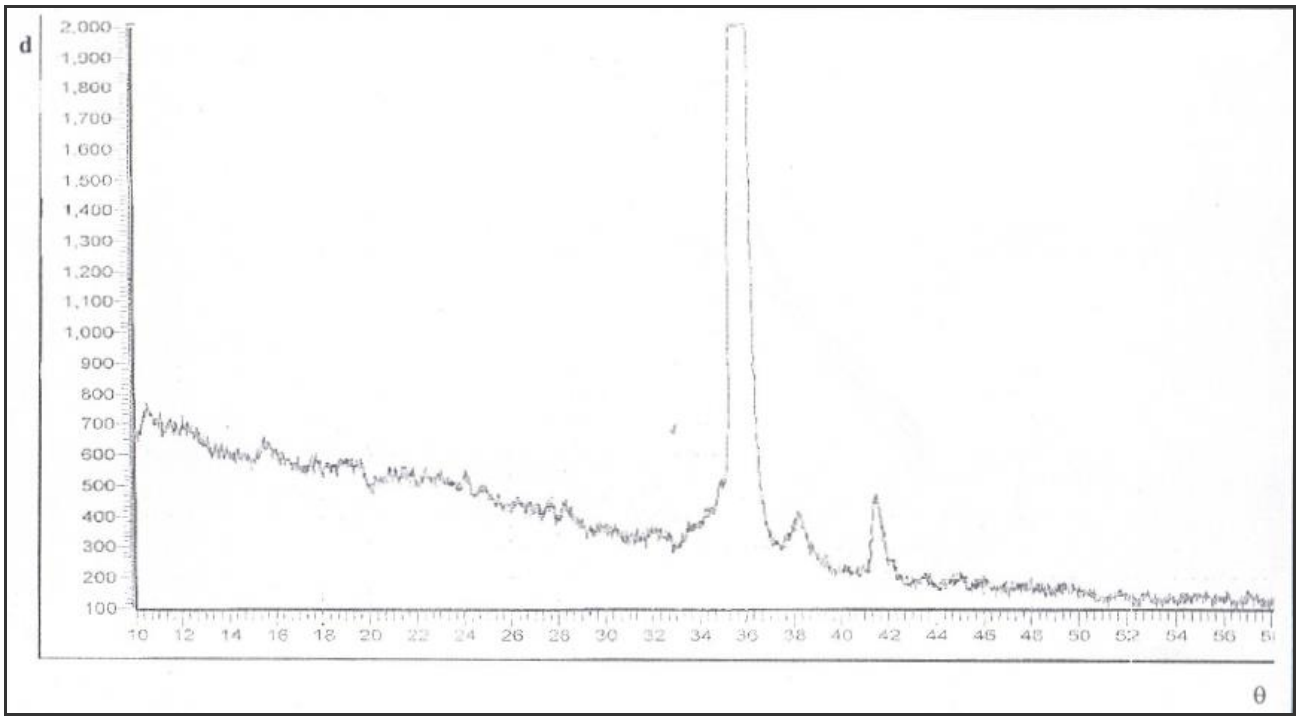

Figure (5) the X-ray spectrum of SiC prepare at 1J/pulse

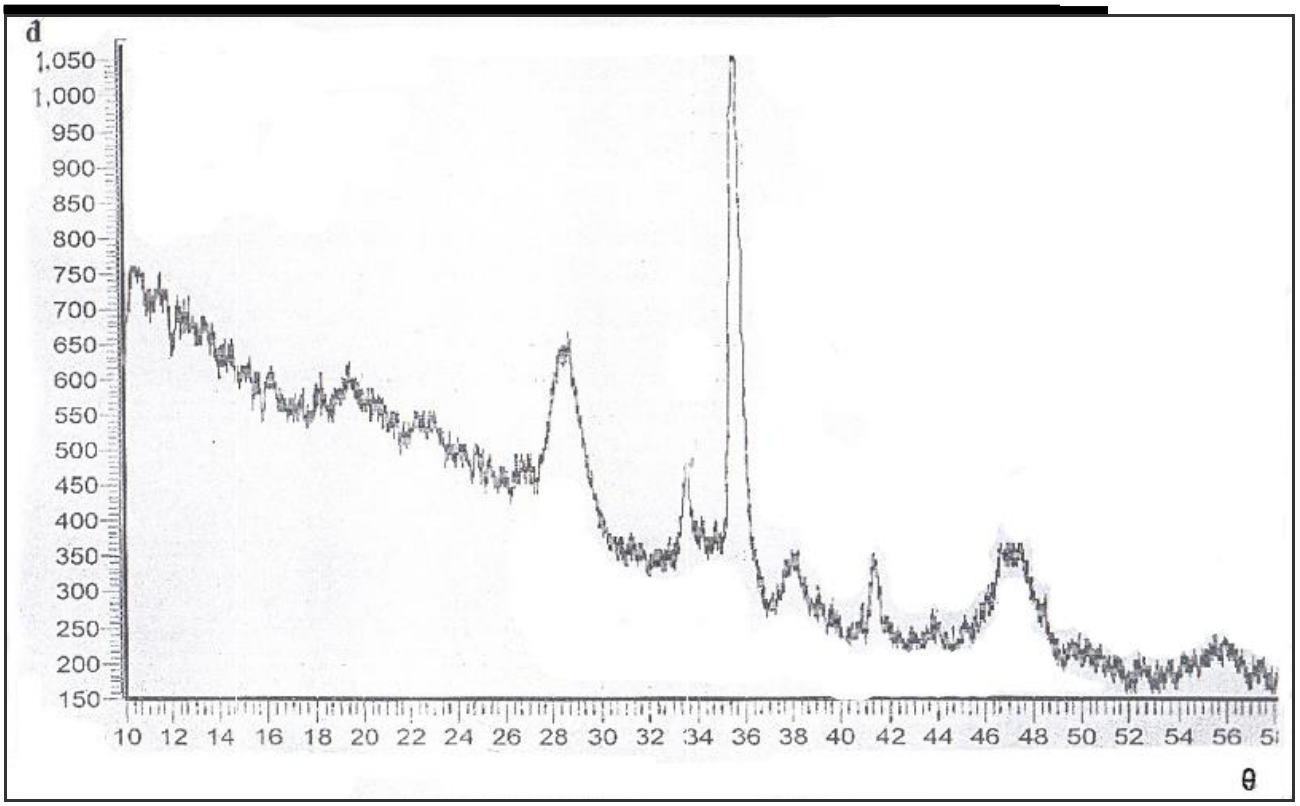

Figure (6) the X-ray spectrum of SiC prepare at 850 mJ/pulse

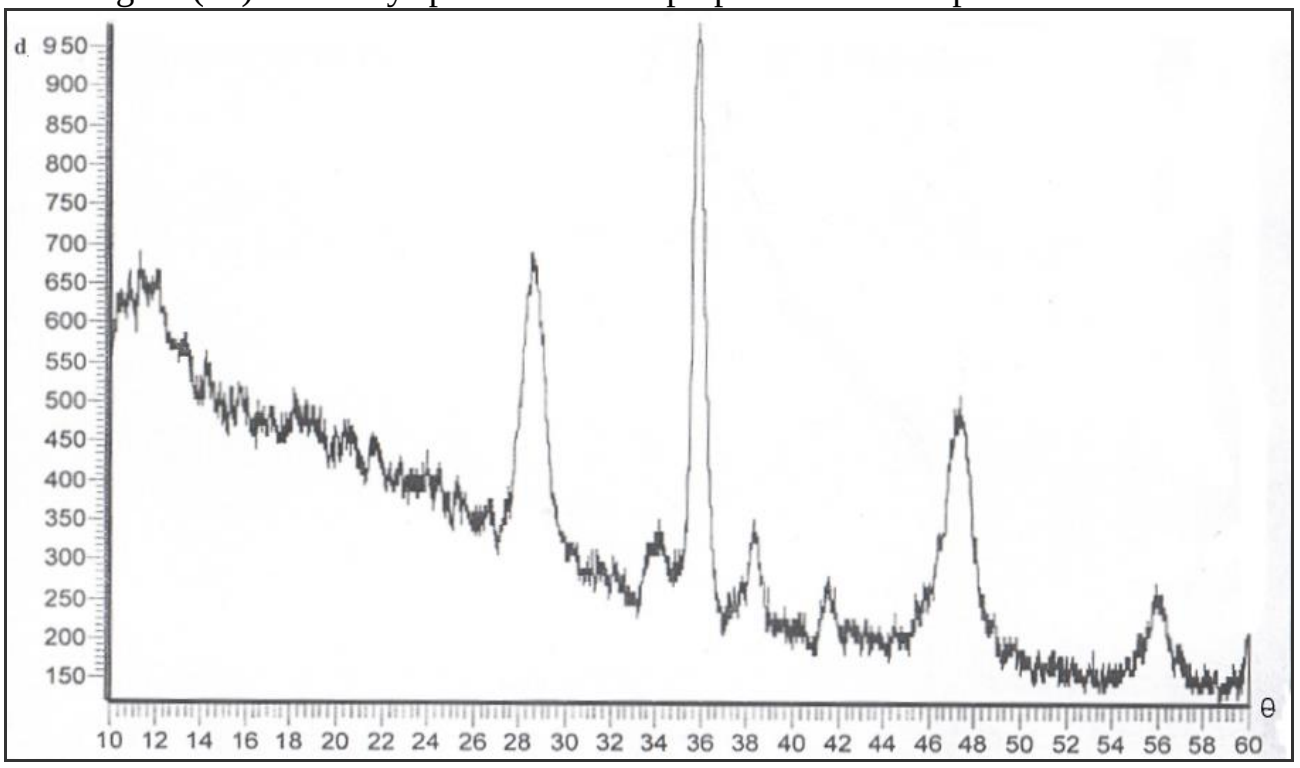

Figure (7) the X-ray spectrum of SiC prepare at 700 mJ/pulse

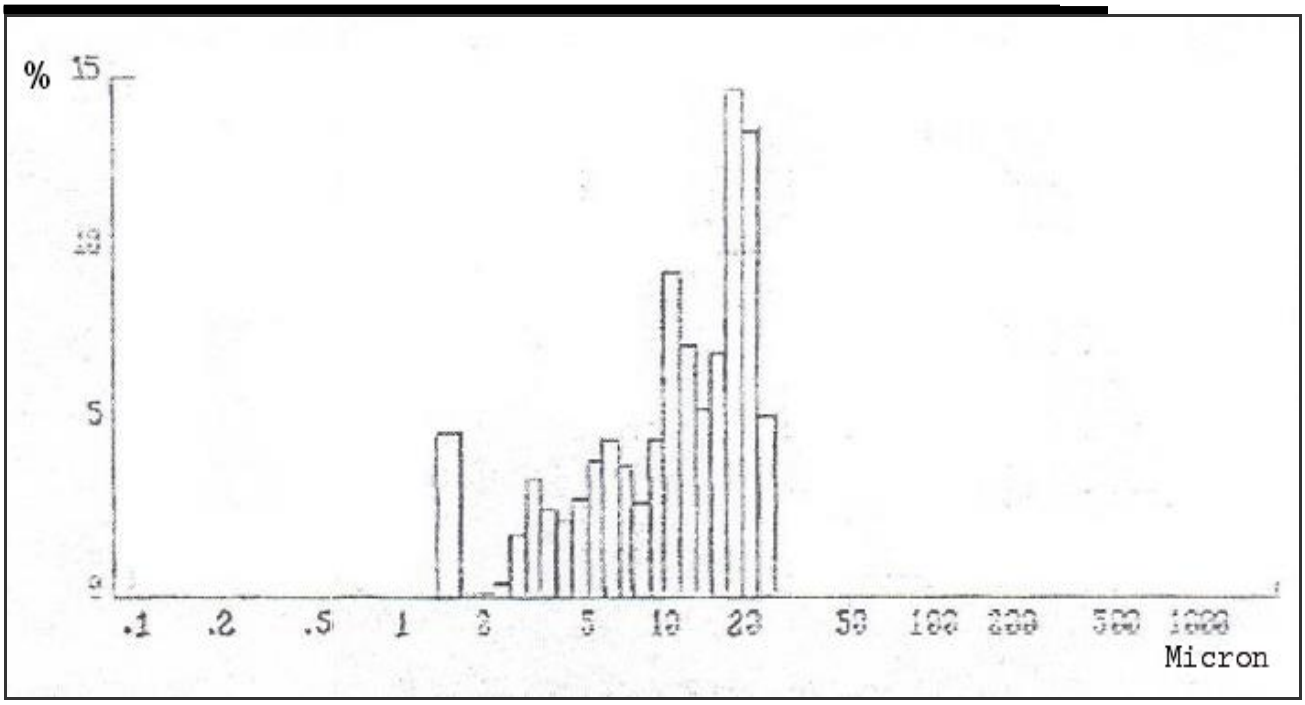

Figure (8) the histogram and frequeny disibution of SiC particles prepared at 850 mJoule/pulse of laser radiation