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Effect of working pressure on the structural and morphological properties of gold nanoparticles prepared by a dc magnetron sputtering technique

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Abstract

Gold nanoparticles were prepared using the dc magnetron sputtering method at multiple argon working pressures [(0.55, 0.85, 1.1, 2.5)×10^{-1} mbar]. The effect of the working pressure on the microstructure and surface morphology of the deposited thin films were investigated in this work. The structural properties of the sputtered films were analyzed using X-ray diffractometer (XRD), atomic force microscopy (AFM), and the scanning electron microscopy (SEM). The XRD results hinted at the presence of polycrystalline films, attributed to the different pressures. The Au nanoparticles showed FCC structures, with an average particle size of (31.25 to 97.65) nm. The nanoparticles were determined to be spherical, as per the SEM. The AFM images confirmed that the thin films are made up of spherical particles that are evenly distributed (in terms of size). The average surface roughness of the films increases alongside the film's thickness within 70.35 – 146.7 nm.

Keywords: Au nanoparticles; Sputtering plasma; Structure properties; Morphology

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Introduction

Metallic nanoparticles is the subject of many studies due to their favorable electronic, mechanical, and optical properties vis-à-vis its bulk counterparts. Gold (Au) nanoparticles is fast becoming notorious, especially in the fields of biomedical, catalysis, plasmon-based sensors, and fluorescence [1]. The interesting physical, optical, and synthetic properties of Au nanoparticles are due to its high surface-to-volume ratio and the enormous amount of atoms at the nanoscale. Recently, many advancements were made in biomedical applications with better biocompatibility in disease diagnosis and therapeutics. Au ,Ag and Cu nanoparticles under 10 nm are exceedingly required for biomedical applications such as treating urinary tract infections (UTI) [2,3]. Au nanoparticles can be prepared using a multitude of methods, such as thermal decomposition of silver compounds, chemical and photo reduction of reverse micelles, reduction in solutions, and recently, via bio-or green-synthesis and radiation-assisted routes [4]. However, most of these methods reported problems pertaining to control of shape and size, and also the occurrences of agglomeration/aggregation. Magnetron sputtering plasma has been used to fabricate thin films, nanostructured coatings, and nanoparticles of metallic oxides and semiconductors films [5-7]. The effectiveness of magnetron sputtering plasma technique makes it a viable option for synthesizing metallic nanoparticles [8], due to the fact that it lack the common problems associated with the aforementioned methods [9]. This study involves the optimization of the operating sputtering pressure(s) to grow uniformly sized and distributed nanostructured Au nanoparticles.

Experimental Procedures

Au thin films were deposited onto glass substrates using a DC magnetron sputtering system. This system consists of planar electrode of an Au metal target (99.9 % pure) having a diameter of 5 cm. The diameter of the vacuum chamber was 31 cm, and it was 37 cm in height. The glass substrate and target was separated by a distance of 5 cm. The development chamber is first emptied at a pressure of 1×10^{-5} mbar using rotary and diffusion pumps. Au thin films were prepared at a 2 kV discharge voltage and 90 mins deposition time for all samples. The operating sputtering gas of pure 99 % argon is pumped into the chamber, at multiple variants (0.55, 0.85, 1.1, 2.5×10^{-1} mbar), at a current density of 10 mA/cm^2. The films and its preparation parameter is listed in Table 1 (thickness of the films are also listed).

Table (1) Working pressures and related thickness data

sample	Pressure(mbar)	Thickness(nm)
Au1	0.055	70.35
Au2	0.085	106.23
Au3	0.110	120.12
Au4	0.250	146.70

The structural properties of the Ag nanoparticles were determined using X-ray Diffraction (XRD) (Rikagu Mini-2 using CuKα1, λ = 0.15406 nm radiations), Morphological observations of films surface was analyzed by Scanning Electronic Microscope (SEM) (TA Instruments USA, DSC Q10). The films surface topography was analyzed by atomic force microscopy (AFM) (Hitachi: H-7500; Resolution: 2 Å).

Results and Discussions

Figure (1) shows the XRD patterns of the Au nanoparticles thin films measured in this work. It can be seen that the dominant peak at around (37.734°) match with (111) plane indicating a strong preferred plane along this direction. Moreover, the intensity of this plane increased with increasing of chamber pressure. In addition, for prepared films (Au3 and Au4), the peaks with low intensity observed at

(43.956), (64.196) and (77.447) are indexed as (200), (220) and (311) planes. The observed diffraction peaks of samples correspond to of the face center cubic structure (FCC) of Au, which in agreement with (JCPDS card No. 96-901-2431) [9-11].

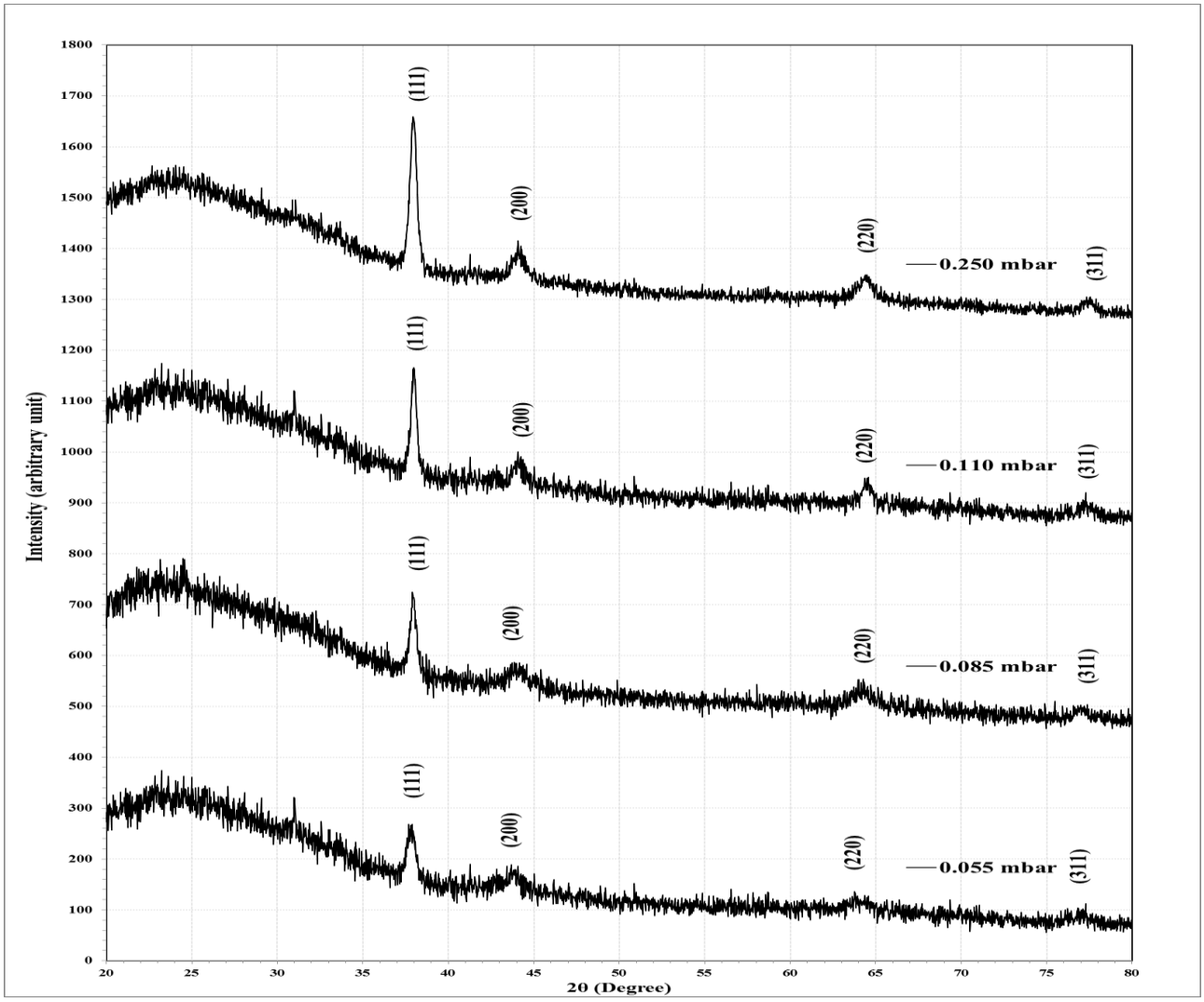

Fig: (1): X-ray diffraction of gold thin films deposited at different gas pressures

:Crystallite size was calculated using the Scherrer formula for different working gas pressures ,which is given by [10,12-14]

$$D = (0.9\,\lambda)/(\beta \cos\theta)$$

where λ is x-ray wavelength, β is width at half maximum (FWHM) of intensity, and θ is the Bragg angle. Table (2) shows the parameters of the structural properties. The XRD plots confirmed the presence of pure Au nanoparticles. For Ag films, the XRD results are showing a increase in the crystallite size were measured using Scherrer's equation as a parameter of working pressure increases. The average crystallite size of Au thin films prepared at different pressures was found to increase from 14.5 to 21.3 nm with an increase of pressure from 0.55 to 2.5×10^{-1} mbar, as shown in table 2. The sputtering yield and rate of the deposition reaction increases at higher working pressure and the crystallites grow faster resulting in a larger size.

Table(2): The XRD parameters of gold films nanoparticles.

P (mbar)	2θ (Deg.)	FWHM (Deg.)	d_{hkl} Exp.(Å)	C.S (nm)	d_{hkl} Std.(Å)	hkl
0.055	37.8350	0.5798	2.3760	14.5	2.3500	(111)
	43.7740	1.1100	2.0664	7.7	2.0352	(200)
	63.9740	1.2396	1.4541	7.6	1.4391	(220)
	76.9820	1.2096	1.2376	8.4	1.2273	(311)
0.085	37.9400	0.4318	2.3696	19.5	2.3500	(111)
	44.0570	0.9680	2.0538	8.9	2.0352	(200)
	64.2160	1.2888	1.4492	7.3	1.4391	(220)
	77.1030	0.9908	1.2360	10.3	1.2273	(311)
0.11	37.9770	0.3951	2.3674	21.3	2.3500	(111)
	44.1370	0.7528	2.0502	11.4	2.0352	(200)
	64.4580	0.7594	1.4444	12.4	1.4391	(220)
	77.3050	1.0672	1.2333	9.5	1.2273	(311)
0.25	37.9560	0.4574	2.3687	18.4	2.3500	(111)
	44.1380	0.8930	2.0502	9.6	2.0352	(200)
	64.3780	1.0022	1.4460	9.4	1.4391	(220)
	77.4270	0.9212	1.2316	11.1	1.2273	(311)

SEM images of the Au nanoparticles in the thin films on the glass substrates are shown in Figure (2). It can be seen in 0.055mbar working sputtering pressure that the surface is flat and the average size of the particles is (20-30 nm), while it can be seen in 0.085mbar, that the thin film' surface are transforming, but by bit, from a flat surface into granular surface, with particle sizes of ~ (35– 50 nm). It can be seen in 0.11mbar , that agglomeration is evident, and the particle sizes are 50 nm, which could be due to parametric error. The images of Au nanoparticles in 0.11mbar are spherical, measuring ~ (30-50 nm). The surface of the Ag nanoparticles are generally consistent and free of splits, demonstrating excellent adhesion and normality. The SEM images show the spherical characteristics of the Au nanoparticles. The sputtered particles diameter increased in tandem with working pressure, where increasing the working pressure to 0.25 resulted in its clustering on the substrate due to increasing temperature. The spherical shape of the particles were reported by Wang et al. [15,16].

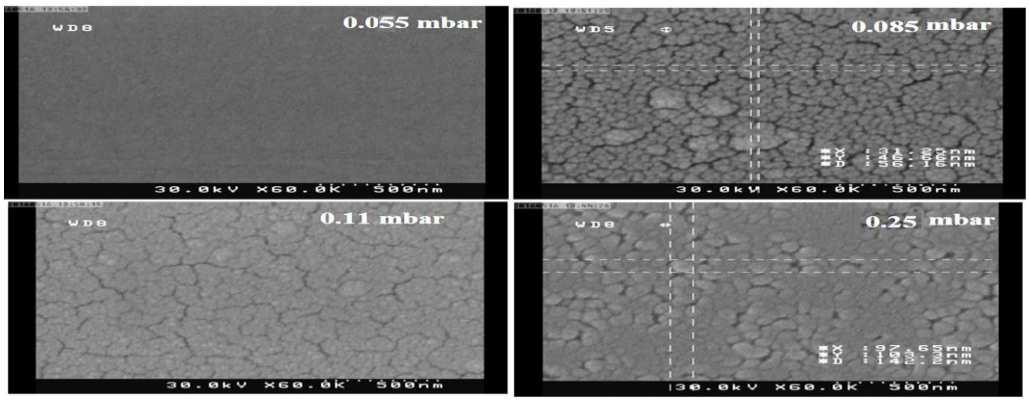

Fig (2) :The SEM images of gold thin films deposited on glass substrate at different working pressures.

Figure (3) show the AFM images of the samples. The surface morphology of the Ag nanoparticle thin films has a mass thickness, d_m, of (1.82, 7.60, 8.09, 7.37) nm for (Au1, Au2, Au3, Au4), respectively. It can be seen that variations are evident in the shapes and sizes of the metallic particles. Figure 3(a, b, c, d) show particles that are symmetrical and spherical, with triangular, hexahedral, and semi-pentagonal particles well distributed throughout the samples, *sans* aggregation. AFM data are shown in Table 3, where the grain sizes are (9.55 – 14.4 nm) and the RMS roughness are (0.468 and 2.35 nm).The deposited film show broad size/shape distributions. It is also evident that gas pressure(s) and grain size (*s*) are directly proportional, which could be due to increased sputtering rate and agglomerated small grains forming larger grains that increases the substrates' temperature.

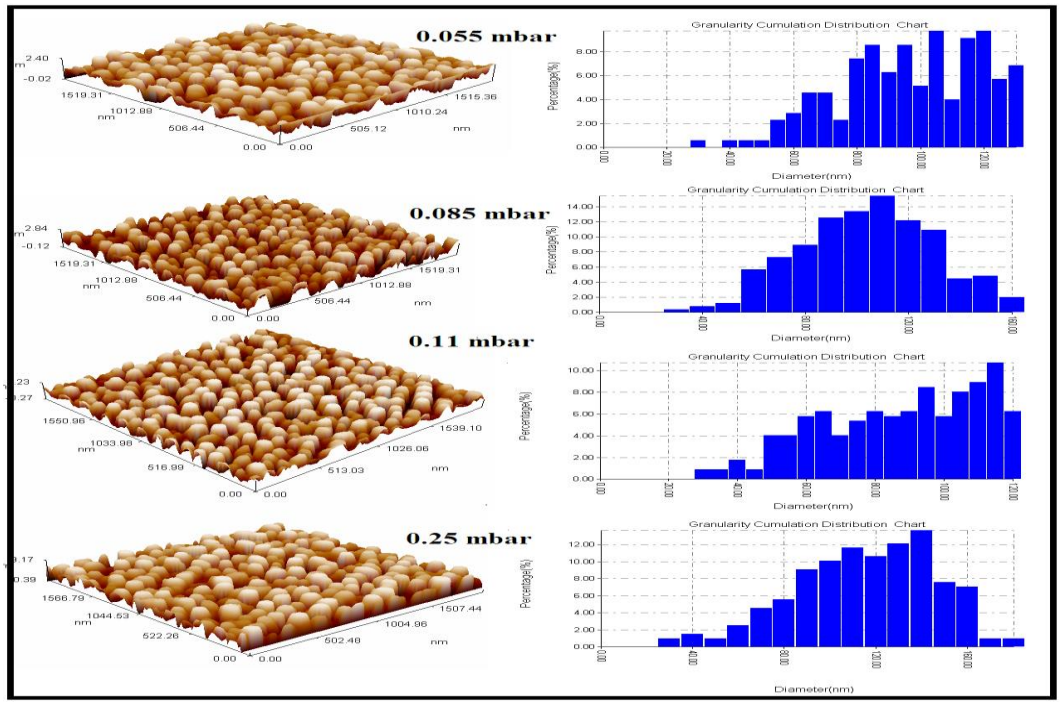

Fig. (3): AFM images and granulation distribution chart of Ag films deposited on glass substrate at different pressure (0.55, 0.85, 1.1, 2.5) ×10^{-1} mbar.

Table(3) data of AFM image of Au nanoparticles for different pressures

Pressure(mbar)	Average Roughness (nm)	Average Grain Size (nm)
0.055	0.468	9.55
0.085	0.617	11.5
0.11	2.35	14.4
0.25	2.03	12.4

Conclusion

Au nanoparticles thin films was successfully synthesized using the dc magnetron sputtering plasma technique at a constant sputtering voltage 2 kV, a deposition time of (90) mins, and pressures of [(0.55, 0.85, 1.1, 2.5)10^{-1} mbar] for the target-substrate distance of (5) cm. XRD analysis confirmed the FCC structure of the Au nanoparticles and peaks at (111) and (220), indicating a pure Au phase. Particles sizes were measured using the SEM, determined to be ~(31.25 to 97.65) nm in the case of all of the samples. The SEM images showed excellent adhesion, the lack of splits, and great consistency. The AFM image confirmed the presence of (spherical) Au nanostructures due to pressure changes in the vacuum chamber, with varied surface morphology and surface roughness (0.468 to 2.35) nm.

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