



## PRESSURE AND FAST NEUTRON IRRADIATION EFFECT ON THE PROPERTIES OF $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{8+\delta}$ SYSTEM

Bushra. A. Aljurani, Ghazala.Y.Hermiz, Amal.K.Jassim

University of Baghdad, College of Science, AL-Haithem

gyhermiz@yahoo.com

Received 09-02-2012,, online 13-02-2012

### Abstract

Solid state reaction method was used to prepare samples with nominal composition  $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{8+\delta}$ . The influence of variation of pressure (0.3, 0.5 and 0.7GPa.) and fast neutrons with average energy of (5MeV) and flux of  $2.8 \times 10^4$  n/cm<sup>2</sup>.sec (from <sup>241</sup>Am/Be source) at room temperature for one week, on the properties of Tl-1223 superconducting samples have been studied

X-ray diffraction (XRD) analysis showed a polycrystalline structure with a high  $T_c$  phases (1223) as a dominant phase and low  $T_c$  phase (1212 and 1112) addition to existing of some impurity phases for all samples before and after irradiation. It has been found that transformation from orthorhombic to tetragonal phase occurred when exposed samples to fast neutron. In addition, structural defect (point defect, clusters or columnar tracks) produce some time disordering of microstructure and reduce the critical temperature of HTSC material. The highest transition temperature 135K obtained for unirradiated sample pressed under 0.5GPa.

### I. INTRODUCTION

After the discovery of high temperature superconductivity in BSCCO system [1] Herman et al [2] have reported superconducting transition in a Tl-Ba-Ca-Cu oxide composite with zero resistance below 107K. The Tl system has many superconducting phases with different transition temperatures and stacking structures containing various numbers of Cu-O layers. In addition, transmission electron microscopy studies show that various types of intergrowths can be observed in sintered samples of the Tl system [3]

Doping with various elements was found to be useful and effective in improving the properties of HTSc. For example, Pb doping of Bi-Sr -Ca-Cu -O and Tl-Ba-Ca-Cu-O superconducting systems induces a partially melted liquid phase that eases the diffusion of the element to form the high- $T_c$  phases and to increase their critical temperature. [4]

The physical perfection of superconducting materials has a large influence on the superconducting properties. In general, it has been found that the current carrying capacity is proportional to the defect density, and also to the degree of difference between the defect and the matrix. Physical heterogeneities have been introduced into the materials by mechanical deformation, inclusion of second phases, voids, and impurities, and, finally, by irradiation. The inhomogeneities are viewed as pinning centers which restrict the motion of magnetic flux through the material.

Various investigations [5, 6] have been carried out on the effect of fast neutron irradiation and gamma-ray on the properties of polycrystalline and single crystal superconductors. Cooksey et al. [5] show that gamma-rays and neutrons had little to no effect on the critical temperature and critical current density of thin films of YBCO and

Tl-2212 and Tl-2223. Since these properties of superconductors are highly sensitive to atomic displacement effects in the material. They conclude that very little damage occurred in the unbiased superconductors at room temperature as a result of the irradiation performed.

Aoki et al. [7] carried out a feasibility study on the application of HTS coils in medical accelerators, which are used for particle cancer therapy. In this application, HTS are exposed to radiation. As a result, the HTS conductors become radioactive, thereby making the operation and maintenance of the reactors or accelerators difficult. Further, radiation affects the superconducting characteristics and causes an increase in the amount of nuclear waste generated. They carried out irradiation experiments on Bi-2223 and YBCO tapes at room temperature by using 14-MeV neutrons.

In this paper the effect of the pressure and fast neutron irradiation on the structure and transition temperature of TBCCO system were studied.

## II. EXPERIMENTAL PART

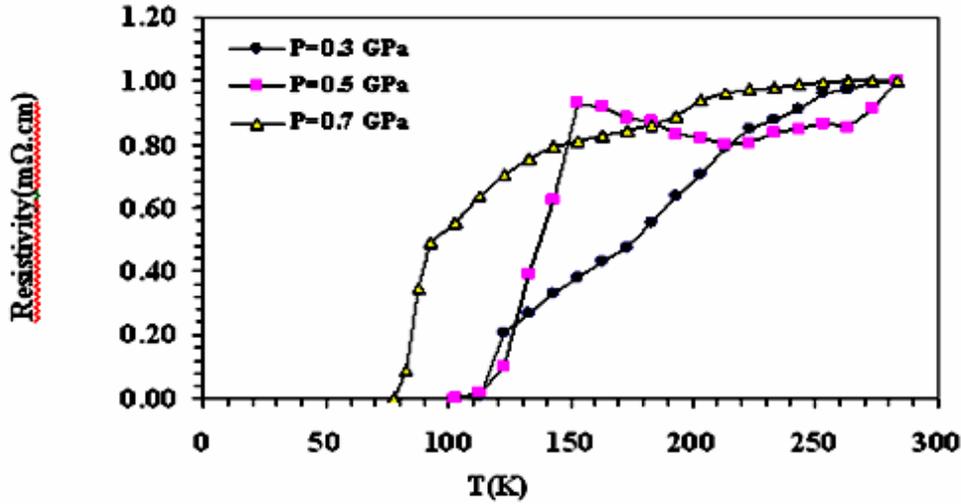
Two step solid –state reactions was used to prepare the samples. Mixing appropriate amounts of the oxide, carbonates of Ca, Cu, and Ba develop  $Ba_2Ca_2Cu_3O_8$  precursor. The mixture homogenization takes place by adding a sufficient quantity of 2-propanol to form paste, during the process of grinding for about (50-60) min. Measuring the weight of the dried mixture (w1), and put it in an alumina crucible, calcined in a tube furnace in air that has programmable controller type (Eurptherm818) for 24 hours at 900 °C with a rate of 2°C / min. The second step, the  $Ba_2Ca_2Cu_3O_8$  precursor was reground again and mixed with  $Tl_2O_3$  and  $Pb_3O_4$  to obtain the nominal compositions  $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{8+\delta}$ . This mixture was then pressed into pellets 1.3 cm in diameter and (0.2 – 0.3) cm thick, using hydraulic type (SPECAC) under different pressure of 0.3, 0.5 and 0.7 GPa. Then these samples placed in a tube programmable furnace. Temperature of the furnace was raised at a rate 200 °C/h up to 600 °C for one hour and there after at a rate 100 °C/h up to 860 °C and held at this temperature for 3h. Finally, the furnace was cooled to room temperature by the same rate of heating.

Four probe dc methods at temperature range (80-300) K were used to measure the resistivity ( $\rho$ ) and to determine the critical temperature ( $T_c$ ). The structure of the prepared samples was obtained by using x-ray diffractometer (XRD) (Philips). A computer program was established to calculate the lattice parameters a, b, and c. The program is based on Cohen's least square method [8].

Irradiation of the samples have been done at room temperature by using fast neutron source  $^{241}Am/Be$  with energy 5MeV and neutron flux  $2.8 \times 10^{14}$  n/cm<sup>2</sup>.sec at room temperature for one week .

## III. RESULT AND DISCUSSIONS

The temperature dependence of the electrical resistivity for samples of  $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{8+\delta}$  under different pressures 0.3, 0.5 and 0.7 GPa. is shown in

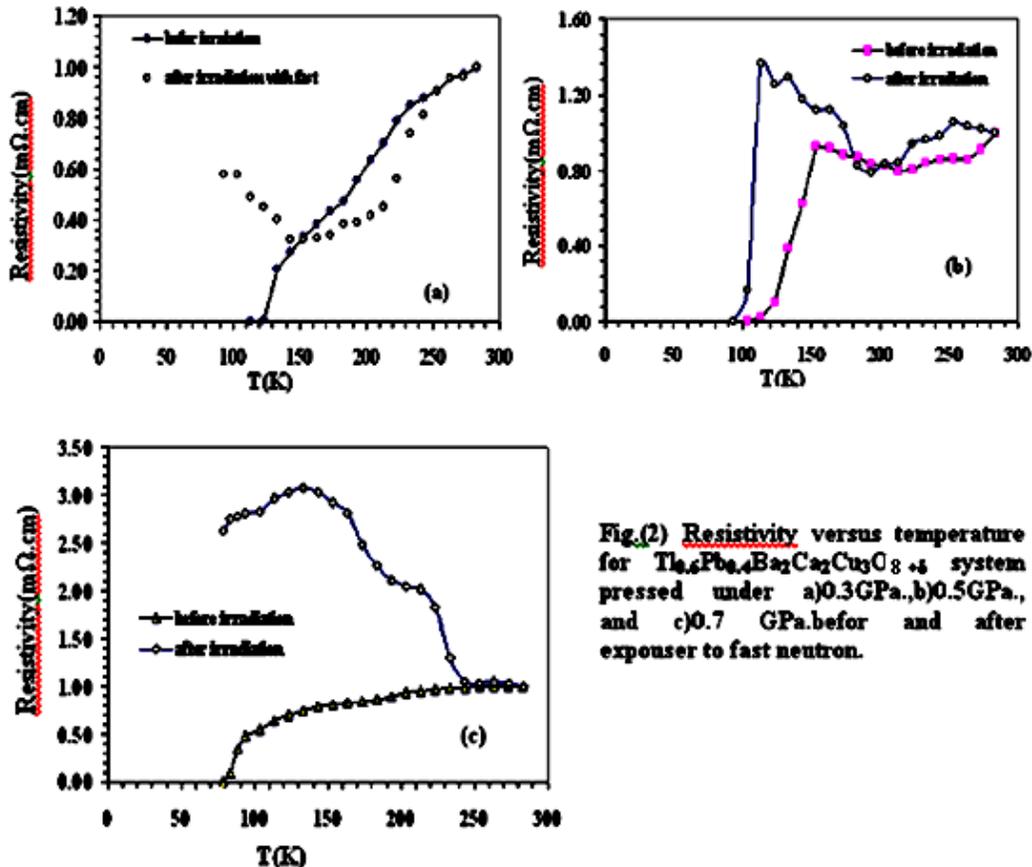


**Fig(1): Resistivity versus temperature for  $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{8+\delta}$  system pressed under different pressure**

Fig. (1). It is found from this Fig. that the resistivity of the samples increases with the enhancement of pressure from 0.3 to 0.7 GPa.

The sample pressed under 0.5GPa. shows a semiconductor behavior from 270K to 150K and have zero resistivity at 103 K ,while the other samples show a superconductor behavior from 270K to the boiling point of liquid nitrogen and the transition temperature is at 123K ,135K and 85K for the specimen pressed under 0.3,0.5 and 0.7 respectively.

Fast neutrons irradiation reduces the superconducting performance; the resistivity of the irradiated samples was increased. Samples pressed under 0.3 and 0.7GPa. loses their superconductivity,they behave as semiconductor in range extend from (140-90)K



**Fig.(2) Resistivity versus temperature for  $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{8+\delta}$  system pressed under a)0.3GPa.,b)0.5GPa., and c)0.7 GPa.before and after exposure to fast neutron.**

and from(300-140)K respectively as shown in Figs.(2a, 2c).On the other side the transition temperature decreases to 108K for samples pressed under 0.5GPa.after exposure to fast neutron as shown in Fig.(2b).The reason may attributed to the change of oxygen stoichiometry which controls the hole concentration in conducting  $\text{CuO}_2$  planes .Thus, irradiation induced change in oxygen content as expected to bring change in carrier concentration resulting in the value of  $T_c$  and resistivity. The defects which leads to weak links between grains and consequently it renders largely unconnected after irradiation as indicated by Sauerzopf et.al.[9]

The samples pressed under different pressure were subjected to gross structural characterization employing the x-ray diffraction technique. The XRD data collected from various samples showed a polycrystalline structure with a high  $T_c$  phases (1223) as a dominant phase and low  $T_c$  phase (1212 and 1112) addition to existing of some impurity phases for all samples before and after irradiation. It has been found that transformation

**Table (1):** Values of lattice constants,  $c/a$  and transition temperature for  $\text{Tl}_{0.6}\text{Pb}_{0.4}\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$

Pressure GPa.	Before irradiation					After irradiation				
	a(A°)	b (A°)	c (A°)	c/a	$T_c$ (K)	a(A°)	b (A°)	c (A°)	c/a	$T_c$ (K)
0.3	3.8399	3.8237	15.7051	4.0899	123	semi	semi	semi	semi	Semi.
0.5	3.8639	3.8965	15.9167	4.1193	135	3.8295	3.8292	15.8667	4.1432	108
0.7	3.9035	3.8695	15.8063	4.0492	85	3.8525	3.8526	15.8100	4.1038	<77

from orthorhombic to tetragonal phase occurred when exposure samples to fast neutron as shown in Table (1).Indeed neutron irradiation generally causes knock of oxygen deficiency and metallic to semiconducting phase transition occurs as confirmed by electrical properties. According of the structural properties, the irradiating samples with fast neutron confirmed by both a decrease of the lattice parameter a and increase of the  $c/a$  of the samples in comparison with that unirradiated samples. The reprehensive XRD patterns are shown in Fig.(3-5).

The results of XRD of all irradiated samples revealed lower intensities and disappear of most high  $T_c$ -phase, if compared with the samples before irradiation as shown in Figs (4-5b). This was attributed to displacement of some atoms in the crystal structure as a result of sequence striking due to the cluster defects and a condensation of this defect overlapping is proposed as the mechanism to explain the deterioration of the superconducting properties when irradiated with fast neutrons.

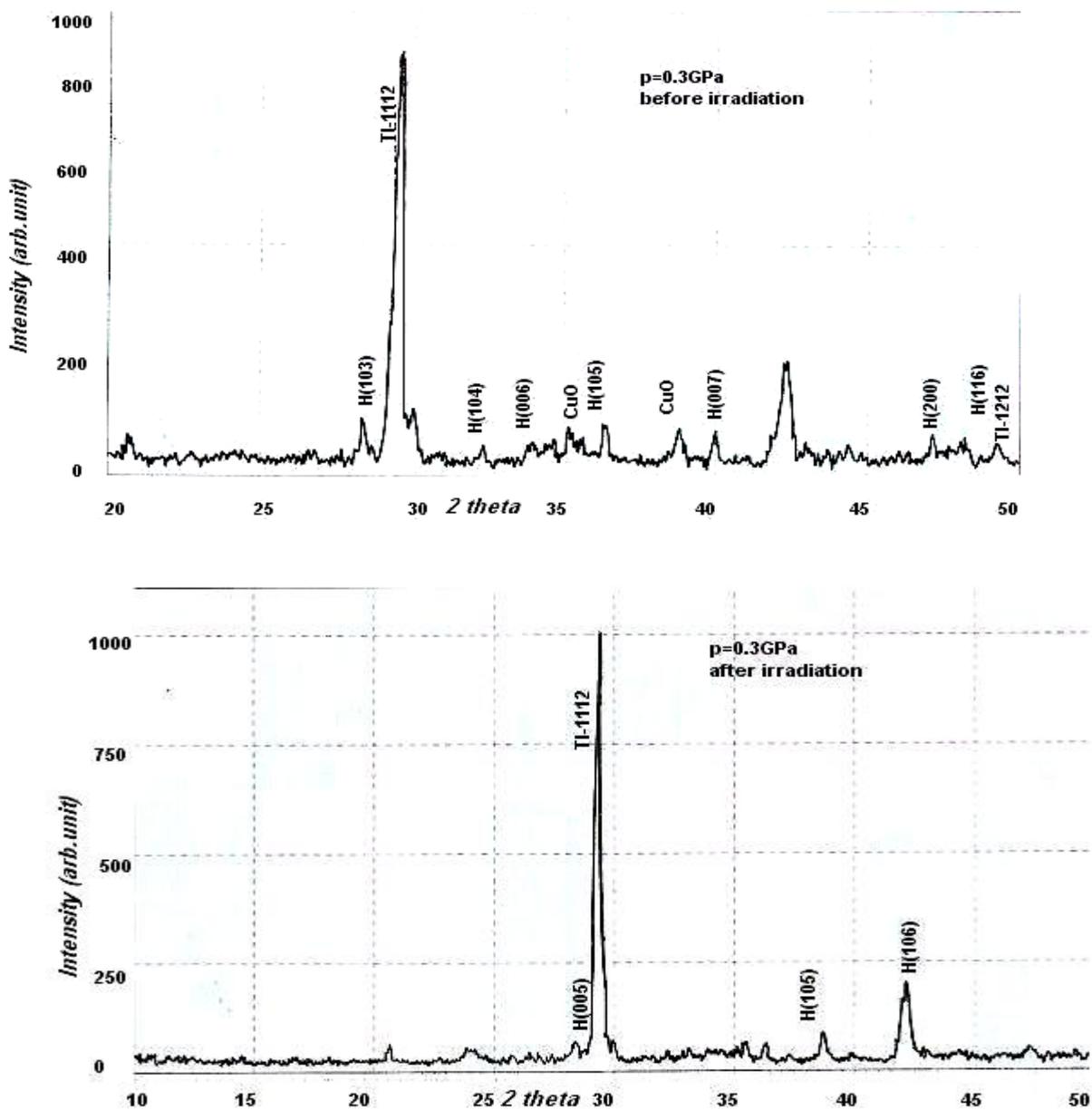


Figure (3) XRD pattern for  $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{8+\delta}$  pressed under 0.3GPa.

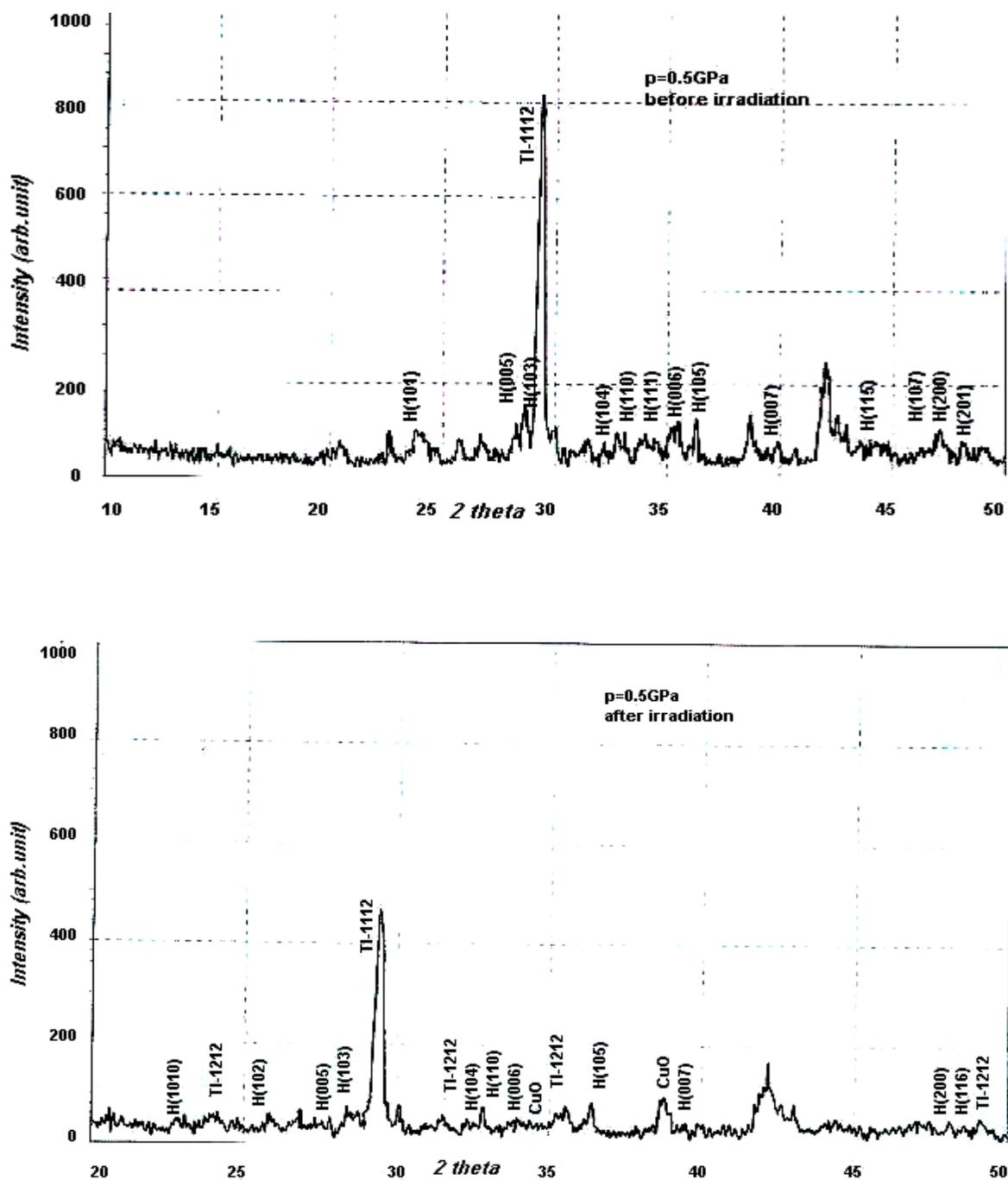


Figure (4): XRD pattern for  $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{8+\delta}$  pressed under 0.5 GPa.

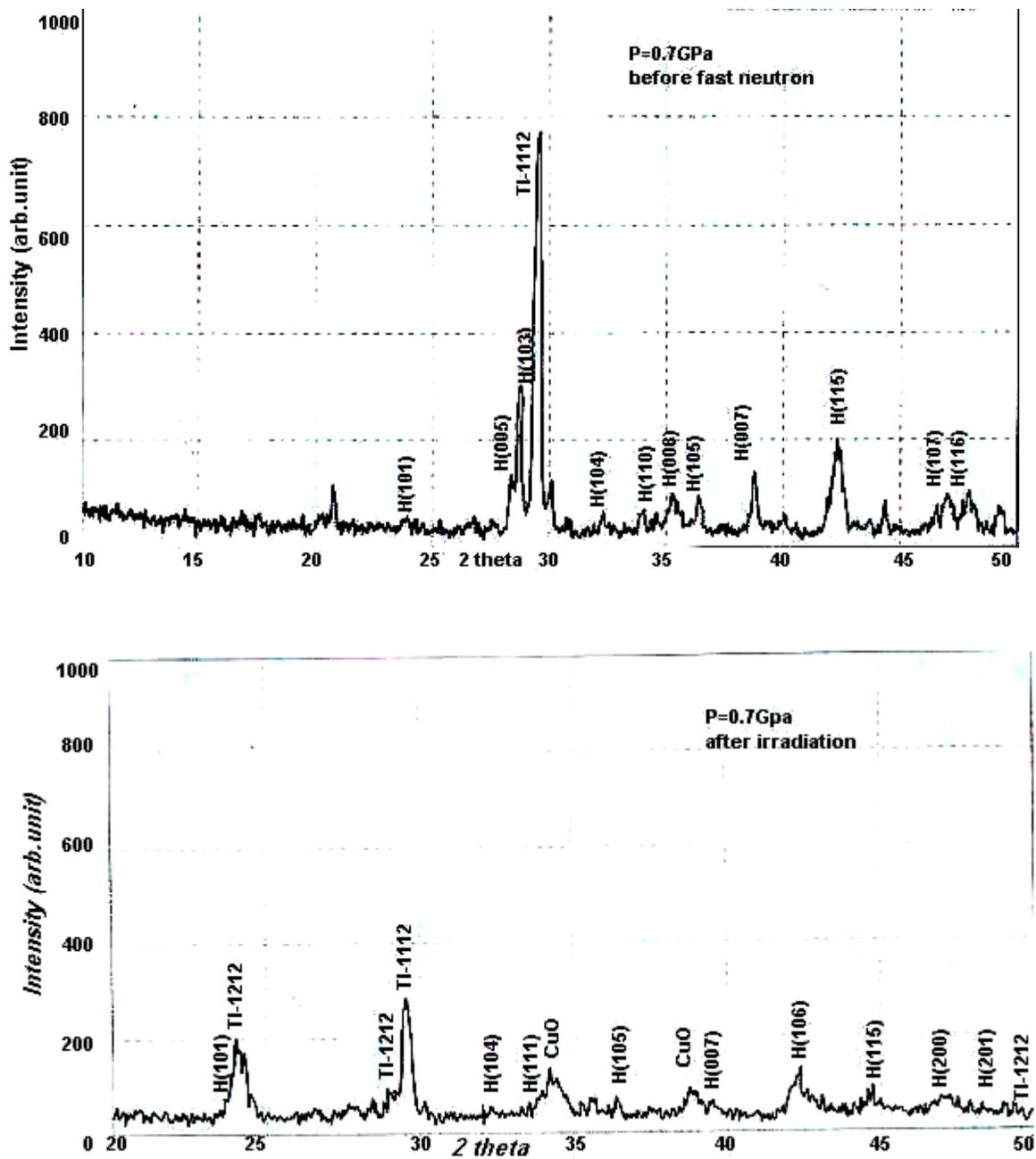


Figure (5): XRD pattern for  $Tl_{0.6}Pb_{0.4}Ba_2Ca_2Cu_3O_{10+\delta}$  pressed under 0.7 GPa.

## IV. CONCLUSIONS

Conclusions from the results can be summarized as follows:

- The highest transition temperature 135K obtained for unirradiated sample pressed under 0.5GPa.
- The XRD data show that phase transition occurs from orthorhombic to the tetragonal structure when expose Tl-1223 to the fast neutron for one week.
- Fast neutron irradiation reduces the superconducting performance by addition structural defect (point defect, clusters or columnar tracks) produced some time disordering of microstructure and reduced the critical temperature.

**Acknowledgments:** We thanks Dr.Kalid Hadi (University of Baghdad, College of Education, Ibn AL-Haithem) for his efforts concerning the neutron irradiation work

## References

- [1] H. Maeda, Y. Tanaka, M. Fukutomi, T. Asano," A New High- $T_c$  Oxide Superconductor without a Rare Earth Element" *Jpn.J.Appl.Phys.***27**, L209 (1988).
- [2] Sheng Z Z and.Herman A M."Bulk superconductivity at 120 K in the Tl–Ca/Ba–Cu–O system " *Nature* **332**, 138 (1988).
- [3] S.Iijima,T.Ichihashi,Y.Shimakawa,T.Manako and Y.Kubo," Intergrowth Structures in Superconductor Tl-Ba-Ca-Cu-O Oxides" *Jpn. J. Appl. Phys.* **27**, 1054 (1988).
- [4] M.Wang,G.Xiong,X.Tang and Z.Hong"The formation mechanism of  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  superconducting phase" *Physica C* **210**, 413 (1993).
- [5] J.W.Cooksey,W.D.Brown,S.S.Ang,H.A.Naseem and R.K.Ulrich,"Gamma-ray and fast neutron radiation effects on thin film superconductors" *IEEE transactions on nuclear science* **41**, 2521 (1994).
- [6] H. Ashour, S. A. Fayek, M. B. S. Osman and H.A. Mohammed"Effect of radiation on some superconducting ceramic materials" *Superconductor Science and Technology*, **17**, 227 (2004).
- [7] Aoki, T. Ueda, H. Ishiyama, A. Miyahara, N. Kashima, N. Nagaya, S.," Effect of Neutron Irradiation on High-Temperature Superconductors" *Applied Superconductivity, IEEE Transactions on*, Issue **3**, 3200 (2011).
- [8] I.F. Ferguson and A.H. Rogerson,"A program for the derivation of crystal unit cell parameters from x-ray powder diffraction measurements" *Computer Phys. Communications* **32**, 95 (1984).
- [9] F.M.Sauerzopf and H.P.Wiesinger,"Fast neutron irradiation and flux pinning in single crystalline high temperature superconductors" *Cryogenics*, **33**, 8 (1995).