4.8 C.

Growth of Sapphire thin films by pulsed laser deposition

submitted for ULF-LENS annual report 1994/95

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1. Introduction

Formation of sapphire thin films, and most particularly sapphire waveguides, is of great importance in the context of optical and opto-electronic devices. Sapphire is the hardest of all oxide crystals, and has a range of physical properties such as thermal conductivity, electrical conductivity and resistance to chemical attack, that make it a superb choice for environments where extended UV transmission, reliability and strength are required.

In the optics arena, titanium-doped sapphire has had a radical impact on the field of tunable lasers. $Ti:Al_2O_3$ is an optically pumpable crystal, producing a laser capable of operating between 650 nm and 1100 nm. Typical dopant levels for Ti^{3+} ions are at the $\sim 0.1\%$ level in commercial samples of $Ti:Al_2O_3$, which yields an absorption coefficient of $\lambda = 2.0\ cm^{-1}$ at 514.5 nm,[1] a suitable wavelength region for optically pumping into the peak of the absorption band at 490 nm.

To date, the fabrication of sapphire waveguides has had mixed success. Ion-beam implantation studies have been carried out using He ions,[2] but the formation of the index barriers required for guidance has necessitated doses exceeding 10^{17} ions/cm^2, which causes surface damage, fracture, and formation of new compounds within the layer. Chemically active ions such as carbon have also been implanted, but the resultant waveguide losses are still high.[2]

An alternative route involving RF sputtering has produced amorphous films, but these are not of the same refractive index as crystalline sapphire, and were susceptible to chemical attack.[3] Further work is in progress on in-diffusion of titanium into sapphire to make waveguiding layers.[4] In this work, we describe our first attempts to grow crystalline films of α - Al_2O_3, sapphire, via PLD.

2. Experimental results

Films were grown in two different vacuum chambers using pure alumina ceramics as the target material. Target rotation only was used here, as it is technically difficult to rotate the substrate, while simultaneously heating it to the $\simeq 700°C$ temperature required for epitaxial crystalline growth as experimentally determined for a range of crystalline materials. For any new material growth study, the most important parameters to vary are substrate temperature, and partial pressure of any background gas used, often oxygen. With this in mind, the distance from target to substrate, laser energy and energy density, and rep-rate were all kept fixed at 4 cm, $\simeq 500$ mJ, 3.5 Jcm^{-2}, and 25 Hz. The substrate temperature was varied between 400°C and 750°C, and oxygen was used as an ambient gas on several occasions.

The most surprising aspect of Al_2O_3 growth was the deposition rate. For a 15 minute run, thicknesses of between 5.0 μm and 7.5 μm were produced, using single crystal (0001) "c"-plane oriented polished sapphire as substrates. For a 60 minute deposition, under identical conditions, the measured thickness was 26 μm. This is a very pleasing result, and represents one of the thickest films produced by us so far.

Under $\sim 10^{-4}$ mbar base pressure, ablation of pure alumina ceramic produced exceptionally clear films on the sapphire substrate material. Even at 400°C, there seemed no evidence of film cracking, separation from the substrate, or other deleterious effects. Preliminary measurements of refractive index using Brewster angle methods revealed the refractive index of the film to be 1.76 at λ = 633 nm. This is very close to the literature value of 1.766.[1] Furthermore there was no evidence of interference rings or fringes with these films which would occur if the index of the deposited layer differed appreciably from the substrate index.

With a background pressure of $\sim 10^{-1}$ mbar of O_2 present in the chamber at a temperature of ~700°C, a film was produced which did not yield a value of 1.76 for refractive index, and distinct interference patterns were observed. Further depositions therefore were performed in vacuum. This result is something of a surprise, as many of our previous growth studies have shown that stoichiometry is not preserved unless O_2 is present during growth.

Figure 1 shows the result of an x-ray texture camera picture obtained for a virgin Al_2O_3 substrate. Interpretation of the diffracted spots reveals the surface to be (0001), but slightly off-axis. The corresponding picture for the 26 μm thick film as grown, and without any annealing treatment is shown in figure 2. It is evident from this figure that in addition to the diffraction spots generated from the substrate, several broad bands appear, indicating a textured film, with small grain sizes of order 50-200Å. Figure 3 shows the diffractometer trace for this film, again revealing that the peaks are very broad, and are not a close match to the desired α-Al_2O_3 phase, i.e. sapphire.

Annealing of this film was subsequently carried out a temperature of 1200°C for 24 hours, in flowing oxygen. The post-anneal texture camera picture, shown in figure 4 now reveals in addition to the diffraction spots, a series of bands, at the same 2θ values, confirming the presence of the α-Al_2O_3 phase. Further confirmation is shown in figure 5, which is a diffractometer trace of the post-anneal film. When compared with figure 6, which is a simulation of a pure random powder sample of α-Al_2O_3, good agreement is found for the d-spacings of the two sets of peaks. In other words, the post-anneal sample corresponds to α-Al_2O_3.

3. Discussion and conclusion

These results were obtained from a very limited number of sample depositions. To achieve a 26 μm thick film which has been post-processed into sapphire is very encouraging. The next step is clearly to repeat the deposition parameters, but at higher substrate temperatures. To this end, a purpose designed CO_2 laser heater is being developed, which should be capable of heating the sapphire substrates to temperatures approaching 1000°C. Additionally, we are about to further anneal this sample to temperatures of 1300-1400°C, to investigate improved crystalline growth conditions.

In the longer term our aims are two-fold. Firstly, sapphire-on-sapphire will not lead to a waveguide. We intend to grow sapphire on quartz substrates, which have the requisite lower index needed for guidance. Secondly, we must dope the sapphire film during growth with the correct percentage ($\sim 0.1\%$) of titanium ions, in the appropriate Ti^{3+} valence state. This will be attempted using a double-target system, using separate targets of Al_2O_3 ceramic, and titanium metal, or ideally a single $Ti:Al_2O_3$ single crystal target. This option however is likely to prove expensive, so perhaps $Ti:Al_2O_3$ ceramic would also be used if available.

Acknowledgements

The authors are pleased to acknowledge Professor Costas Fotakis and all the staff at FORTH-IELS, especially Gianna Zergioti and Roula Klini.

References

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2. P.D. Townsend, P.J. Chandler, R.A. Wood, L. Zhang, Elec. Letts **26** p.1193 (1990).

3. K.K. Shih, D.B. Dove, J.Vac.Sci. Technol. **A12,** p321, (1994)

4. L.M. Hickey, private communication.

Figure captions

Figure 1. Texture camera picture of (0001) oriented sapphire substrates used.

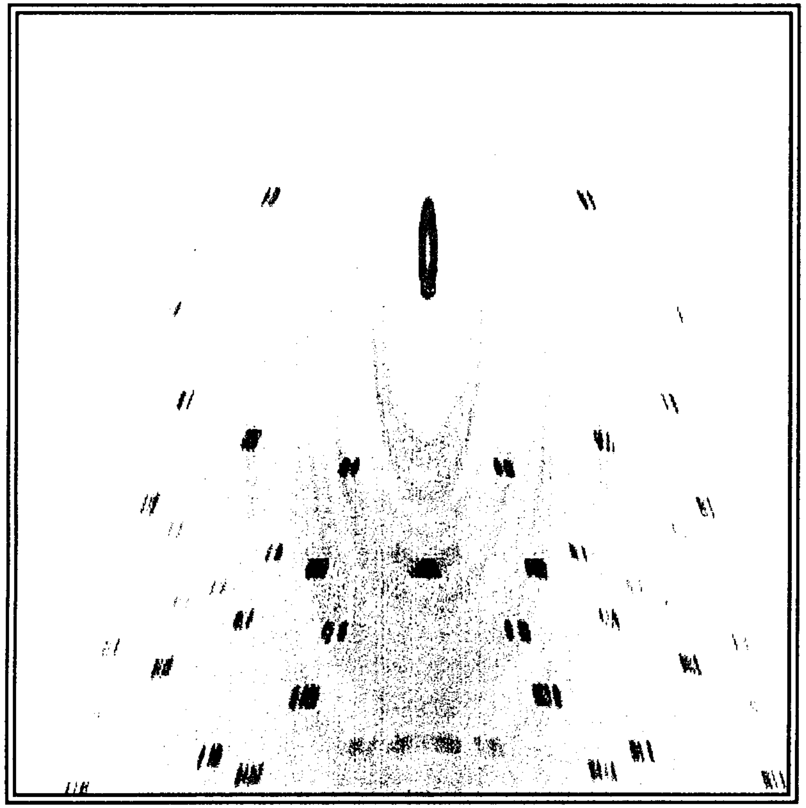
Figure 2. Texture camera picture of film #61 as grown. Broad bands indicate film is not Al_2O_3.

Figure 3. Diffractometer trace of film #61.

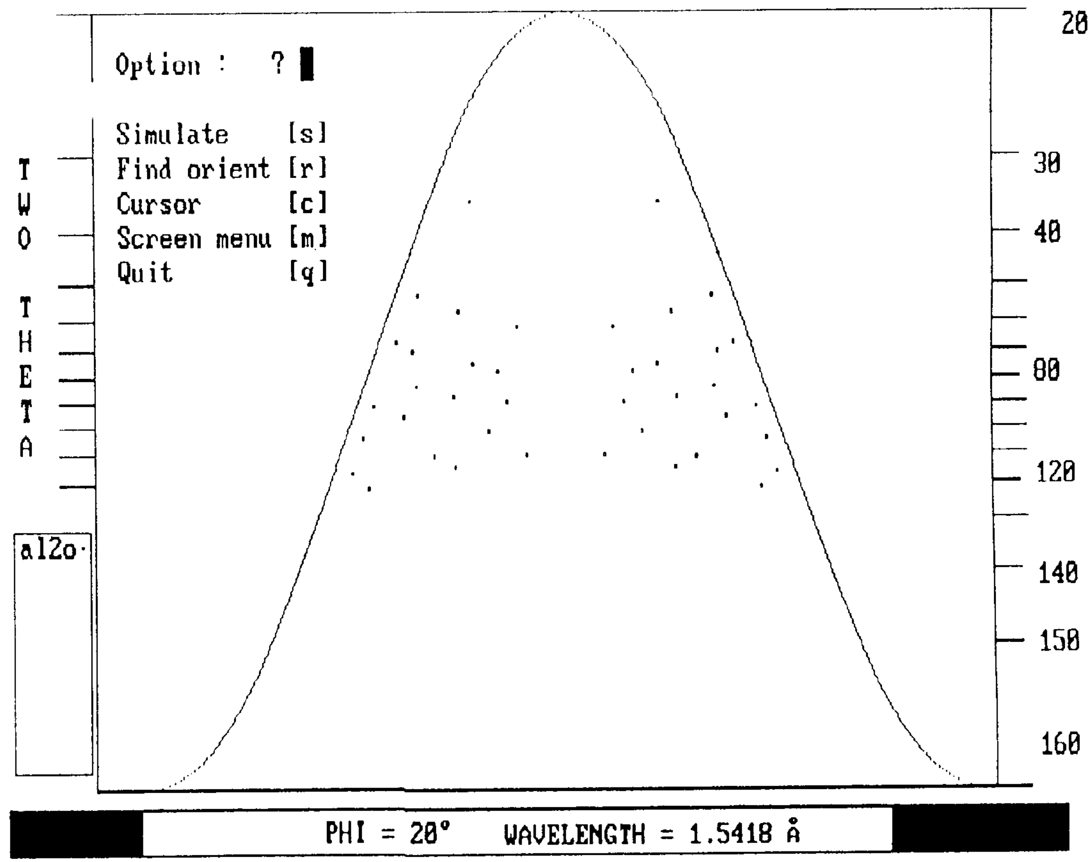
Figure 4. Texture camera picture of film #61 after annealing at 1200°C. Horizontal bands are at same 2θ values as (0001) oriented sapphire substrates.

Figure 5. Diffractometer trace of post anneal film.

Figure 6. Diffractometer trace of α-Al_2O_3 simulation.



sample



simulation – (00.1) Al_2O_3 substrate

Fig 1

Don't use

#61 as received

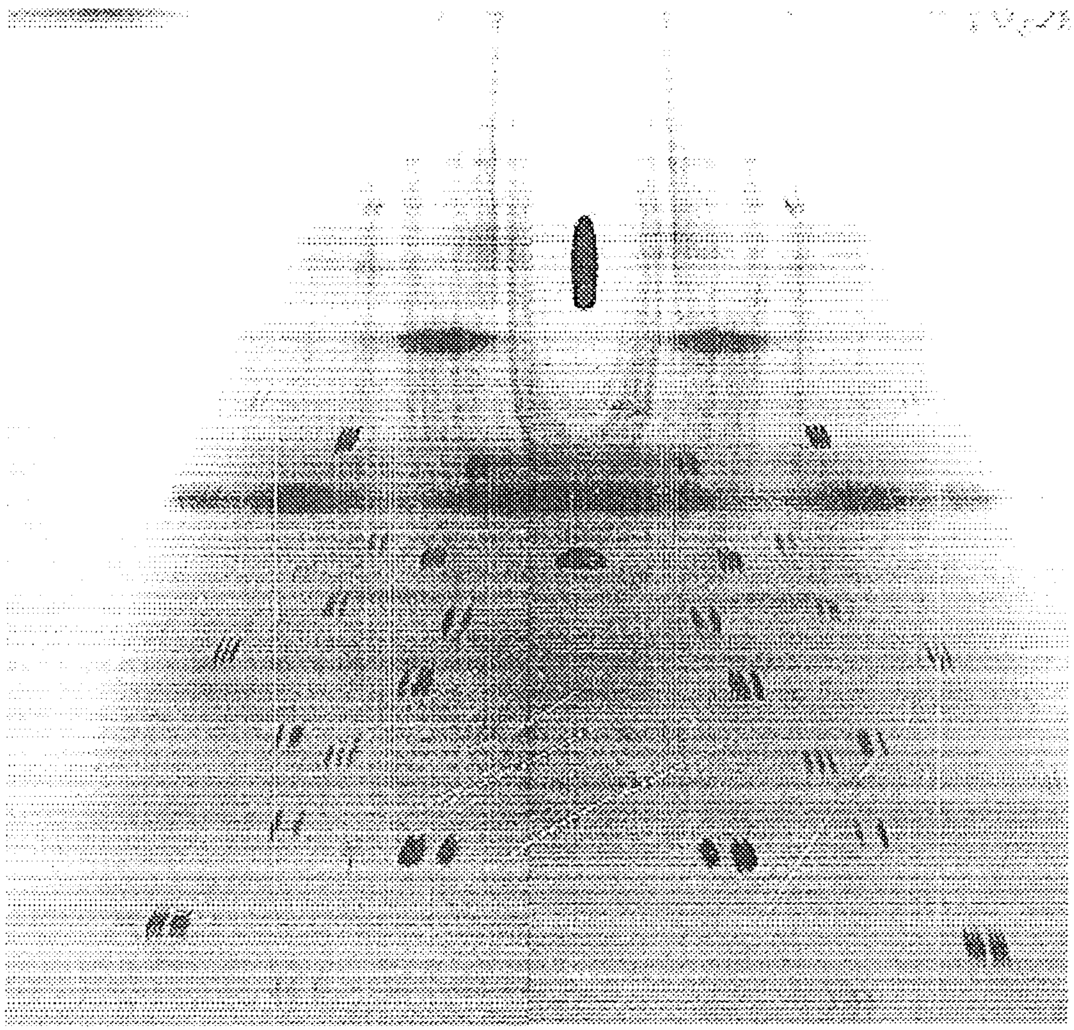


Fig 2

#61 - Al_2O_3(?) on Al_2O_3 [pre- anneal]

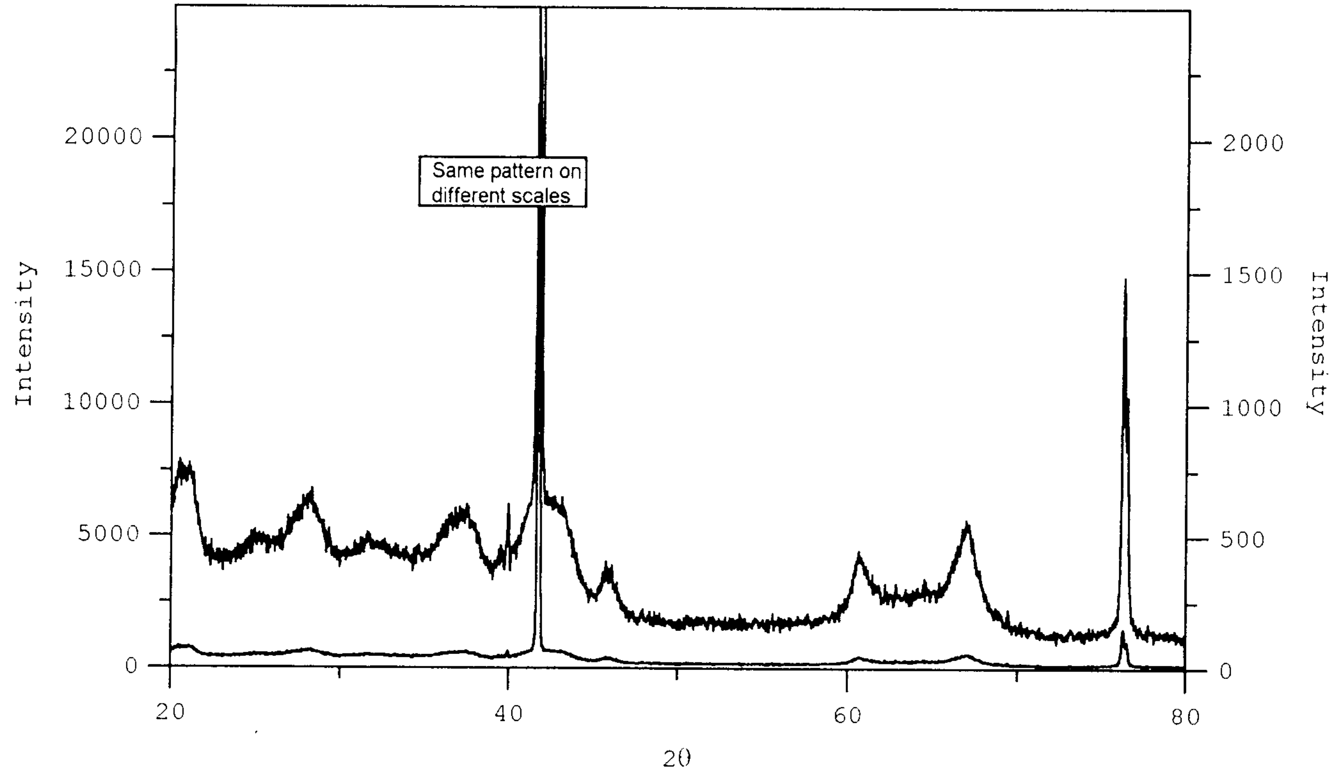


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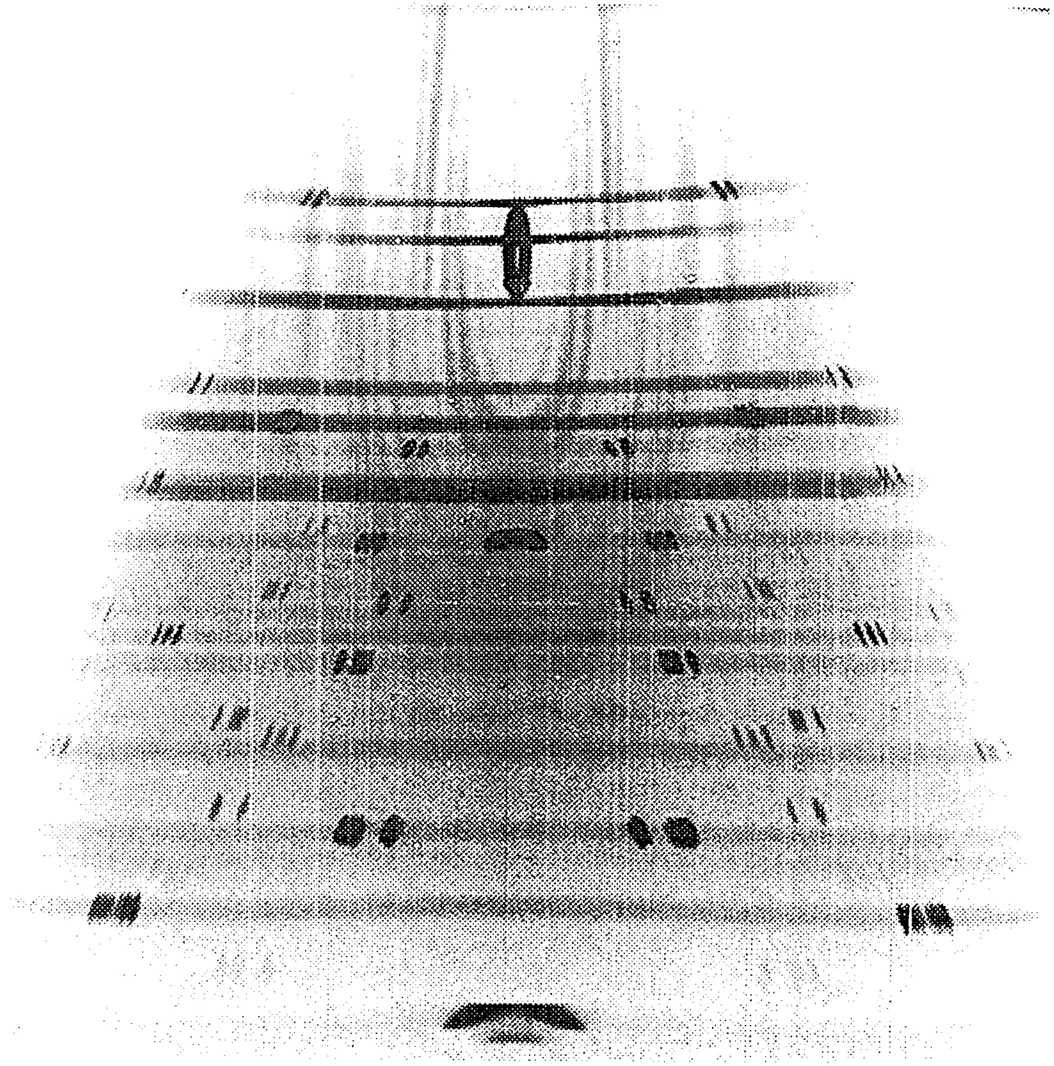
Fig 3



#61
post anneal
(1200°C - 24hs)

Fig 4

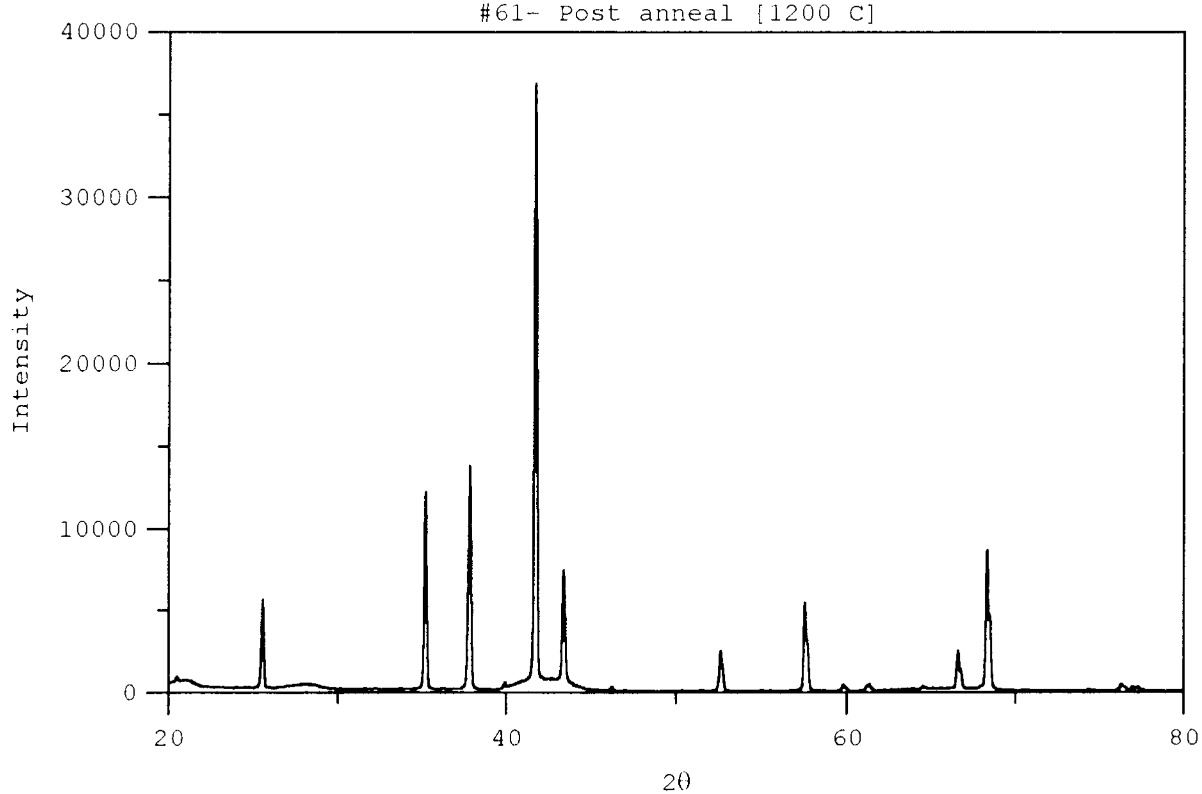


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fig 5

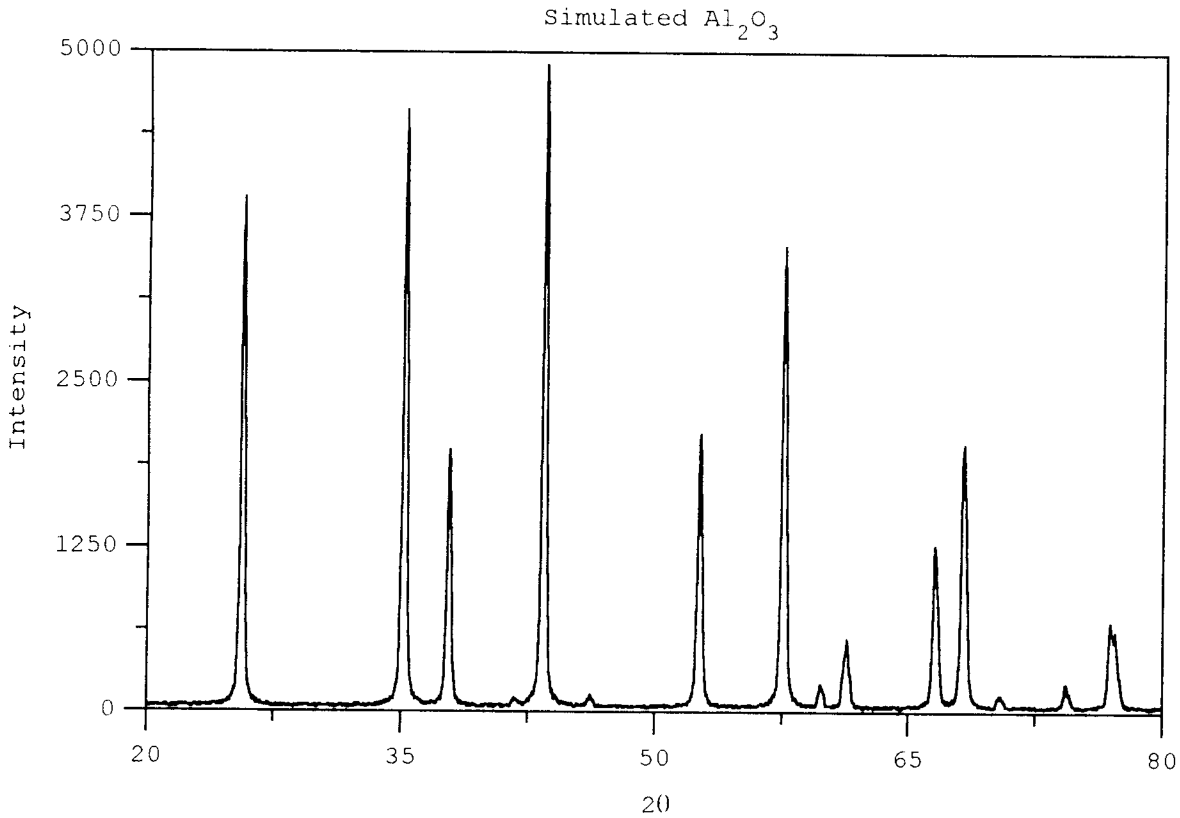

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Fig 6