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Investigation on Second Harmonic Generation (SHG) efficiency of the Grown Semi Organic Crystals – Γ-Glycine with Additives by Aqueous Solution Method.

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Abstract: In the last few decades nonlinear optical materials are getting attention in the field of optical data storage, telecommunication, Second Harmonic Generation (SHG) and optical signal processing, etc. In the present work we are reporting the single crystal growth of γ-Glycine with different complexes like nitrate, oxalates and chloride. The above said crystals were grown in a time span of 3 to 4 weeks in an aqueous solution by adopting slow evaporation solution growth technique at room temperature. The grown specimens were subjected to various characterizations. Its relative second harmonic generation efficiency was tested by using Kurtz and Perry method, keeping KDP as reference. Some of the Nitrates namely Potassium nitrate, Ammonium oxalate, Ammonium chloride have enhanced the SHG efficiency. But at the same time γ-Glycine mixed with Barium nitrate and Calcium nitrate has decreased the SHG efficiency.
Key words: NLO crystal, semi organic, SHG
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1. INTRODUCTION:

Materials answering for high optical nonlinearity are a potential area which has attracted many theoretical and experimental researchers. At present the trend is towards semi organic material compared to inorganic and organic since they possess higher mechanical strength and chemical stability (1)

Pure inorganic NLO material, Potassium di hydrogen phosphate (KDP) and its isomorphs are representative of hydrogen bonded materials which possess important piezoelectric, electro-optic and nonlinear optical properties with excellent mechanical and thermal properties, but possess relatively modest optical nonlinearity (2-4). Hence it is appropriate to grow semi organic crystals which possess the aspects of organic and inorganic materials resulting in promising NLO properties (5).

Many amino acids have already been reported to have NLO property (6). L – Arginine phosphate is the first semi organic material discovered in 1983 (7). For short pulse applications, a second order non linear material ideally must have a large threshold and a large non linear coefficient (8). Amino acids - Glycine, Luicine are well known amino acids for second harmonic generation. These semi organic crystals answer for appreciable high SHG efficiency making the crystals suitable for non linear applications.

These semi organic crystals typically involve the generation of harmonics of Nd based near infrared solid state lasers (9). Typical one, the 1064 nm fundamental of a Nd-YAG laser can be converted to its 532nm, (Second harmonic) or to its 355nm (Third harmonic) or to its 266 nm (Fourth harmonic) using γ-Glycine compound.

We have made an attempt to measure NLO activity in amino acid – γ-Glycine- with additive namely Alkali nitrates such as Potassium, Sodium, Barium, Calcium as well as Ammonium Oxalate and Ammonium Chloride, in definite ratios. All crystals fall in the category of semi organic NLO material. We report here the role or importance of the nitrates, oxalate, and chloride favoring NLO property resulting in higher efficiency of the second harmonic generation (SHG).

2. EXPERIMENTAL

CRYSTAL GROWTH:

Analytical reagent [AR] grade samples were used. Salts were taken in their molar mass, separately and then mixed together. Supersaturated solutions of the mixed salts were kept for slow evaporation in beakers covered with filter paper at room temperature. The selected combinations are:

γ-Glycine+ Potassium Nitrate (GPN Crystal)
γ-Glycine + Sodium Nitrate (GSN Crystal)
γ-Glycine + Barium Nitrate + Potassium Nitrate (GBNPN Crystal)
γ-Glycine + Barium Nitrate + Calcium Nitrate (GBNCN Crystal)
γ-Glycine + Barium Nitrate (GBN Crystal)
γ-Glycine + Ammonium Oxalate (GAO Crystal)
γ-Glycine + Ammonium Chloride (GAC Crystal)

All the grown crystals were colorless. Good transparent crystals were obtained in a matter of 3 to 4 weeks shown in figures 1 to 7.

Fig-1

GSN crystal

Fig-2

GAO crystal

Fig-3

GAC crystal

Fig-4

GBN crystal

Fig-5

GBNPN Crystal

Fig-6

GBNCN crystal

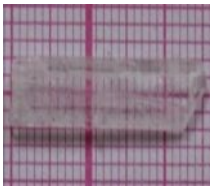

Fig-7

GPN crystal

3. RESULTS AND DISCUSSION:

3.1 STRUCTURAL DATA:

Powder XRD has been taken for some of the grown samples and are shown in figures 8 to 12.

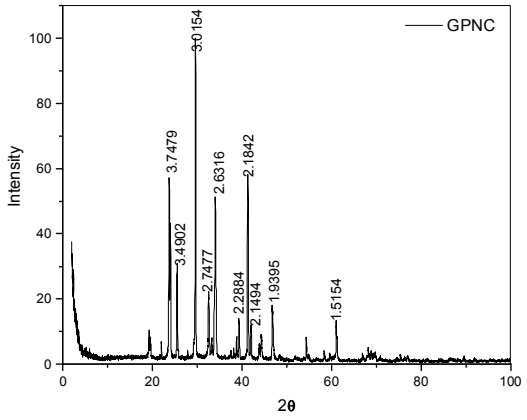

Fig-8: Powder XRD of GPN crystal

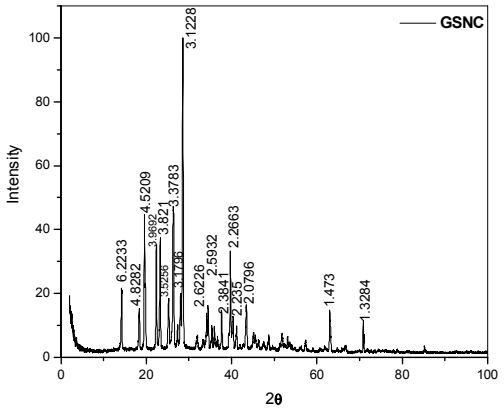

Fig-9 : Powder XRD of GSN crystal

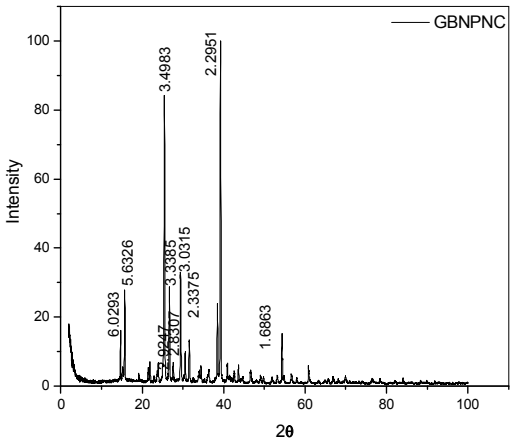

Fig-10 : Powder XRD of GBNPN crystal

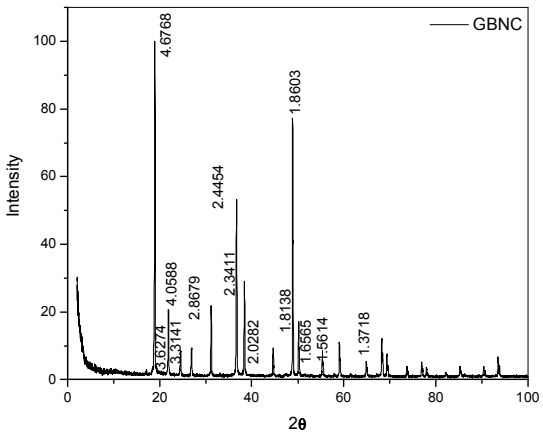

Fig-11; Powder XRD of GAC crystal

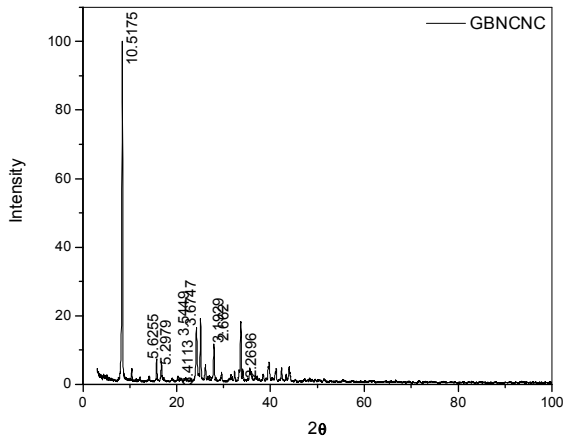

Fig-12: Powder XRD of GAO crystal

3.2 SHG EFFICIENCY:

Kurtz and Perry (10) powder method is an important tool for researchers searching for organic/semi organic/inorganic NLO materials.

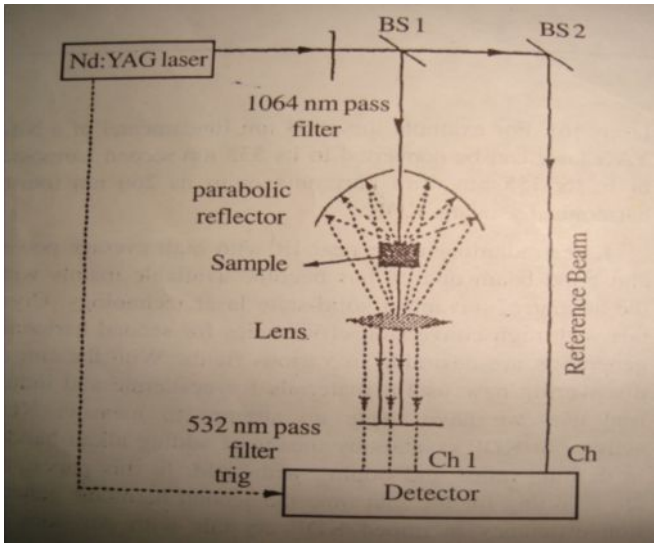

Fig 13

Experimental setup.

The experimental setup used in the present investigation was similar to the generic one devised by Kurtz and is illustrated in Figure 13. It consisted of a Q-switched Nd: YAG laser, the output of which was filtered through 1064nm narrow pass filter.

The power of the fundamental beam was monitored by a split beam technique, in one channel of the power meter. The sample was ground in the form of fine powder of known grain size and pressed between two glass plates. The sample size was kept larger than the beam cross section. The generated harmonic was passed through a 532 nm narrow pass filter and fed to other channel of the power meter. The ratio of the fundamental and harmonic intensities determines the efficiency of the sample. To eliminate the experimental error, urea sample of the same size was also tested in the same setup and the efficiency was evaluated as a ratio. The input power of the laser beam was measured to be 16.5 mJ/ pulse. Pure KDP was used as reference sample. Both the reference and test samples had uniform particle size of 130 to 150microns.The experiment was carried out in pure KDP and later in all the samples. Throughout the experiment the laser power was kept constant. Comparative study of SHG efficiency of all the samples with respect to KDP is shown in **table 1.**

Results show that SHG efficiency of γ-Gylcine containing alkali nitrates answer for higher efficiency compared to KDP. To exhibit NLO activity, the additives present in the host material (γ-Glycine) have to be macroscopically aligned, then only there can be increase in efficiency, justified experimentally (11,12).Among the samples studied, crystals namely Glycine with Sodium nitrate, γ-Glycine with Barium nitrate and Potassium nitrate have answered for higher efficiency. Whereas crystal containing Ammonium oxalate has resulted in excellent efficiency taking KDP as reference. The best additive to organic material works out to be Ammonium oxalate. Next in the series happens to be Sodium Nitrate and (Barium + Potassium Nitrate) combination. These additives are favoring NLO.

Table 1: SHG efficiency:

Semi organic Crystal	SHG signal (mV)	Efficiency with respect to KDP
Glycine+Potassium Nitrate	**6.8**	**0.98**
Glycine + Sodium Nitrate	**9**	**1.35**
Glycine + Barium Nitrate + Potassium Nitrate	**9**	**1.35**
Gycine + Barium Nitrate + Calcium Nitrate	0	0
Gycine + Barium Nitrate	0	0
Gycine + Ammonium oxalate	**10.2**	**1.48**
Gycine + Ammonium chloride	**6**	**0.85**

It is surprising that the additives namely Barium Nitrate, Barium with Calcium Nitrate is acting as poison, not favoring NLO efficiency. In the present study 1 molar % of Barium nitrate and 1 molar % of Barium nitrate + Calcium nitrate has been used. It is interesting to carry out the work at lower values to know exactly at what amount of the above materials starts nullifying the SHG efficiency and making the host crystal lose the NLO property. Definite conclusion regarding the result requires further work on phase matched SHG efficiency of single crystal.

4. CONCLUSIONS:

- Good, transparent crystals of γ-Glycine with Alkali nitrates were grown by slow evaporation method.
- Additives such as Potassium Nitrate, Sodium Nitrate, Ammonium Oxalate, Ammonium chloride, Potassium nitrate + Barium nitrate are favoring NLO efficiency. Hence these crystals are proved to be useful candidature for many applications.
- Barium Nitrate, Barium Nitrate + Calcium Nitrate are acting as poisons to NLO property.
- Enhanced efficiency confirms the better alignment of additives in the crystal matrix.

5. ACKNOWLEDGEMENTS:

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