

Non-destructive compositional analysis of sol–gel synthesized lithium titanate (Li_2TiO_3) by particle induced gamma-ray emission and instrumental neutron activation analysis

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Abstract Lithium titanate, one of the important tritium breeding materials in D–T based fusion reactor under ITER programme, was synthesized through sol–gel route. For chemical quality control of finished product, it was necessary to quantify the lithium and titanium contents. As this ceramic sample is difficult to dissolve, non-destructive analytical methods are preferred for compositional analysis. In the present work, two non-destructive nuclear analytical methods namely particle induced gamma-ray emission (PIGE) using proton beam and instrumental neutron activation analysis (INAA) using reactor neutrons were standardized for the determination of lithium and titanium concentrations, respectively and applied to eleven samples of lithium titanate. To the best of our knowledge, Li quantification in lithium titanate sample is being reported for the first time using PIGE. For quantifications of Li and Ti, 478 keV prompt gamma-ray from ${}^7\text{Li}$ ($p, p'\gamma$) ${}^7\text{Li}$ and 320 keV gamma-ray from ${}^{50}\text{Ti}$ (n, γ) ${}^{51}\text{Ti}$ were measured, respectively, by high resolution gamma-ray spectrometry. The PIGE and INAA methods were validated using several synthetic samples containing lithium and titanium, respectively. Concentrations of lithium and titanium and Li/Ti mole ratios were evaluated and compared with the stoichiometric concentration of Li_2TiO_3 .

Keywords Fusion reactor · ITER · Lithium titanate · Sol–gel method · Lithium · PIGE · Titanium · INAA · Gamma-ray spectrometry

Introduction

Currently, there is a great interest in studies on D–T reaction based fusion reactor under International Thermonuclear Experimental Reactor (ITER) programme. This reactor needs regular supply of deuterium and tritium fuels. Due to very low abundance of tritium (T), it is necessary to produce tritium artificially in order to meet the fuel requirements. For production of tritium, lithium based ceramics are proposed as the blanket materials in fusion reactor. The material for blanket in the reactor should have good chemical, thermo-physical and mechanical stability at higher temperature, low Li vaporization rate, good tritium permeability and low neutron activation characteristics [1, 2]. Presently, studies are carried out using natural lithium based ceramic compounds. The lithium based tritium breeders include Li_2O , Li_2ZrO_3 , Li_2TiO_3 , LiAlO_2 and Li_4SiO_4 [3]. Li_2O is a good tritium breeder but not considered because of high sensitivity towards moisture [4]. Out of these ceramics, lithium titanate (Li_2TiO_3) is considered as one of the suitable candidates for blanket material because of tritium recovery at low temperature and good chemical stability [4, 5]. Different synthetic routes namely solid state reaction [6, 7], sol–gel route [8] and in situ hydrolysis method [9] are proposed for synthesis of lithium titanate. Among these, sol–gel based synthesis of lithium titanate has many advantages like mass scale production, few processing steps, shorter heating cycle, and low rejection rates. In the present work, Li_2TiO_3 used for compositional analysis study was prepared by sol–gel route [8], a brief description of synthesis

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of which is given in the experimental section. Compositional characterization of Li_2TiO_3 finished product is essential for optimization of synthesis procedure as well as for chemical quality control exercise.

Wet chemical methods like ICP-AES and ICP-MS techniques are sensitive for Li estimation but not suitable in the present case as titanium based ceramic materials are difficult to dissolve due to refractory nature of titania (TiO_2). Among the solid sample analysis techniques, laser ablation ICP-MS [10] and laser induced breakdown spectroscopy (LIBS) can determine Li and Ti directly. However, matrix interference is one of the major problems in LIBS technique for solid samples. Other non-destructive techniques such as instrumental neutron activation analysis (INAA), prompt gamma neutron activation analysis (PGNAA) and X-ray based techniques like X-ray fluorescence (XRF) and particle induced X-ray emission (PIXE) are also not suitable for quantification of Li. Whereas Ti can be easily quantified by INAA and PGNAA compared to XRF and PIXE. In view of these, there is a need for simple and non-destructive analytical method for quantification of Li in the solid samples of lithium titanate. We have previously determined lithium concentrations non-destructively in a ceramic sample by particle induced gamma-ray emission (PIGE) using proton beam [11]. PIGE is a complementary technique to PIXE and it is capable of determining low Z elements like Li, Be, B, C, N, F, Na, Mg, Al and Si [11–24] in diverse matrices. This method makes use of on-line measurement of characteristic prompt gamma-rays emitted during nuclear processes like inelastic scattering ($p, p'\gamma$) and nuclear reactions ($p, n\gamma$), ($p, \alpha\gamma$), (p, γ). Lithium can be quantified by measuring 429 and 478 keV gamma-rays from ${}^7\text{Li}$ ($p, n\gamma$) ${}^7\text{Be}$ and ${}^7\text{Li}$ ($p, p'\gamma$) ${}^7\text{Li}$ reactions, respectively [11, 13]. PIGE is a sensitive method for lithium determination [24] as the gamma-ray yields [12, 15] for both 429 and 478 keV peaks of ${}^7\text{Li}$ are higher at higher energy (e.g., 4 MeV) of proton beam. PIGE has been applied to various samples for quantification of low Z elements in various samples like ancient roman glasses for quantification of Na, Mg, Al and Si which are useful for archeological studies [16], aluminum pillared montmorillonite clays for their compositional characterization [17], granite rocks for low Z elements [18], environmental reference material (NIST RM-8414 Bovine Muscle), biological samples (bone, teeth, breath and rabbit bone) [19–21] and lithium in lithium doped neodymium dititanate [11]. This technique also finds interesting applications in designing Li-ion based batteries by quantifying Li concentration [22, 23]. PIGE is a preferred method for non-destructive quantification of Li due to its advantageous properties like isotope specificity and high sensitivity. In addition, absence of gamma-ray interference, negligible matrix effect and high sensitivity makes PIGE an attractive technique for Li estimation.

In the present work, a PIGE method using 4 MeV proton beam from folded tandem ion accelerator (FOTIA) at BARC was used for non-destructive quantification of Li in sol-gel synthesized lithium titanate samples. An in situ current normalization method using F (as CaF_2) was followed to obtain current normalized count rate at 478 keV of Li. Titanium was quantified by relative method of INAA in lithium titanate samples using high flux neutrons from pneumatic carrier facility (PCF) of Dhruva reactor, BARC. The paper presents quantitative results of Li, Ti and Li to Ti mole ratios in eleven selective samples as well as Li and Ti results of synthetic samples.

Experimental

Sol-gel method for preparation of Li_2TiO_3

Lithium titanate microspheres were prepared by dispersing feed solution which is prepared by mixing 3 M HMTA and urea solution with mixture of 3 M TiOCl_2 and 3 M LiCl/LiNO_3 solution in the required ratio (Li:Ti = 2:1 mol ratio). Feed solution was dispersed through a stainless steel capillary of 0.8 mm diameter in hot silicone oil which is circulated into a glass column. The feed droplets become hard as they travel down the column with counter current flow of silicone oil. The microspheres are then separated from the oil and washed with CCl_4 to degrease the surface followed by digestion in 1.55 M LiOH at 60 °C and then washed with 1.55 M LiOH to remove unreacted chemicals and reaction products from the spheres to prevent them from cracking during drying and further heat treatment (100 °C). The spheres were dried and sintered at different temperatures (500–1250 °C) for 4 h. XRD pattern of a lithium titanate sample sintered at 1000 °C is given in Fig. 1. In order to achieve desired density [8], sintering temperature of 1000–1250 °C was maintained. An optical photo micrograph of lithium titanate sintered at 1000 °C is given in Fig. 2. Relevant details on the synthesis are given in Table 1 for eleven selected samples prepared by sol-gel route. Other details can be found elsewhere [8].

Particle induced gamma-ray emission (PIGE)

Sample preparation, irradiation and measurements

Standards of lithium were prepared by homogeneously mixing varying amounts of lithium sulphate with constant amount of CaF_2 in cellulose matrix using agate mortar and pestle. For method validation, eight synthetic samples of Li were prepared. One synthetic sample pellet was prepared by mixing constant amount of CaF_2 and Li_2CO_3 (~50 mg) in cellulose matrix and rest seven pellets were prepared by

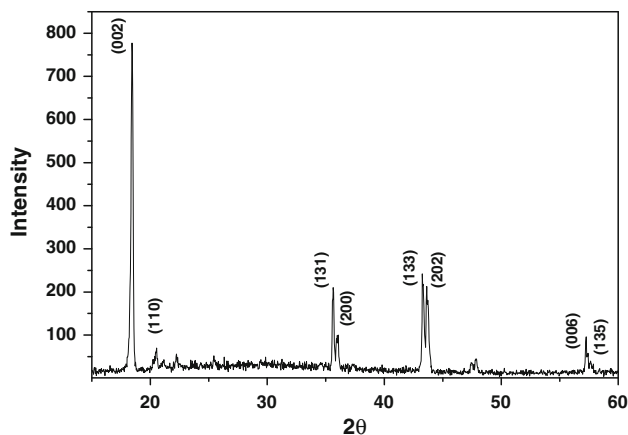


Fig. 1 XRD pattern of 1000 °C sintered Li_2TiO_3 pebbles

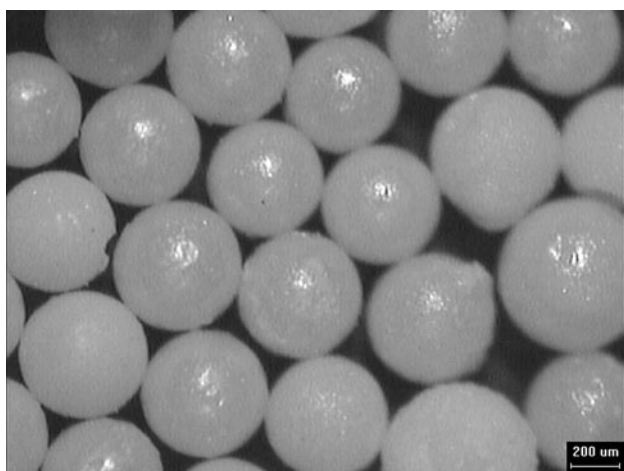


Fig. 2 Optical photomicrograph of 1000 °C sintered Li_2TiO_3 pebbles

mixing cellulose, constant amount of CaF_2 and titanium dioxide (TiO_2 , ~50 mg) with known lithium content (3–19 wt% of Li). The sample pellets of lithium titanate (~100 mg) (Table 1) were homogeneously mixed with fixed amount of CaF_2 in cellulose matrix. The total mass of each of the sample, synthetic sample, and standard pellets was kept constant (~1 g). Target pellets were made from the mixture using KBr die set (2 cm dia \times 0.2 cm thickness) with a pressure of 2 ton.

Sample and standard targets were mounted on a stainless steel ladder and irradiated with 4 MeV proton beam (current ~10–15 nA) from FOTIA, BARC, Mumbai. The γ -rays of 478 and 197 keV, respectively, from ${}^7\text{Li}$ (p, p' γ) ${}^7\text{Li}$ and ${}^{19}\text{F}$ (p, p' γ) ${}^{19}\text{F}$ were measured using a 30 % relative efficiency HPGe detector, placed perpendicularly to the proton beam. The HPGe detector was coupled with 4 k—channel based MCA that has Pulse Height Analysis Software (PHAST) [25]. Targets were irradiated with proton beam and gamma-rays were counted in reproducible sample to detector geometry. A PIGE spectrum of a irradiated

Table 1 Sol–gel synthesis details of lithium titanate samples

Sample no.	Starting material with TiOCl_2	Digestion (LiOH)	Washing (LiOH)	Sintering Temp ($^\circ\text{C}$)
Sample 1	LiCl	✓	✓	500
Sample 2	LiCl	✓	✓	600
Sample 3	LiCl	✓	✓	700
Sample 4	LiCl	✓	✓	800
Sample 5	LiNO_3	✓	✓	1000
Sample 6	LiNO_3	✓	✓	1000
Sample 7	LiNO_3	✓	×	1250
Sample 8	LiNO_3	✓	×	1250
Sample 9	LiCl (50 %) + LiNO_3 (50 %)	✓	×	1000
Sample 10	LiCl (50 %) + LiNO_3 (50 %)	✓	✓	1000
Sample 11	Recycled batch	✓	×	1250

sample of lithium titanate containing F is shown in Fig. 3. Lithium concentration determination in Li_2TiO_3 samples and synthetic samples were carried out by using peak areas under 478 and 197 keV corresponding to lithium and fluorine, respectively. Concentrations of Li in samples were calculated using relative method as given below.

Instrumental neutron activation analysis (INAA)

Sample preparation, irradiation and measurement

Titanium standards were prepared by homogeneously mixing known amount of TiO_2 (30–100 mg) with cellulose matrix. Similarly, synthetic samples for titanium were prepared by mixing varying amount of TiO_2 and lithium sulphate in cellulose matrix. Out of 1 g of homogeneously mixed powder, three samples of ~10 mg were sealed in polyethylene sheet for irradiation. Sample, standard and synthetic samples, sealed in polyethylene, enclosed in polypropylene capsule were irradiated for 1 min at pneumatic carrier facility (PCF) of Dhruva reactor, BARC at a neutron flux of $5 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$. The gamma-ray of 320 keV from ${}^{51}\text{Ti}$ ($t_{1/2} = 5.76 \text{ min}$) was measured using high resolution gamma-ray spectrometry using a 40 % relative efficiency HPGe detector coupled to MCA. A typical gamma-ray spectrum of lithium titanate sample by INAA is shown in Fig. 4.

Calculations

Lithium concentration determination by PIGE

The current normalized count rate (CPS) of Li was obtained by dividing the CPS of 478 keV peak (CPS_{Li}) with the sensitivity ($\text{CPS}/(\text{mg kg}^{-1}) = S_{\text{F}}$) of F at 197 keV

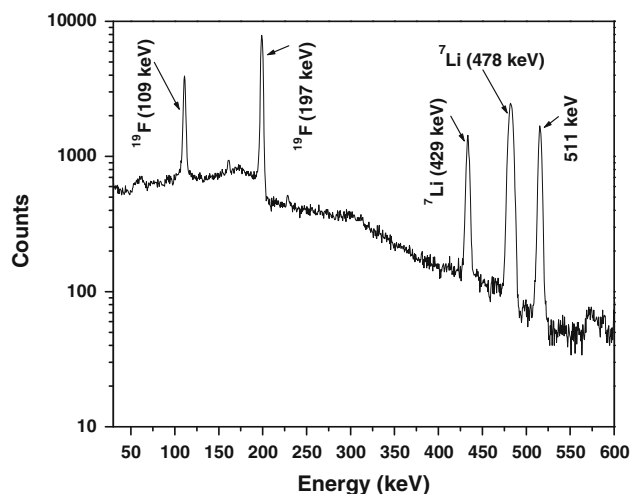


Fig. 3 Gamma-ray spectrum of lithium titanate sample in p-PIGE showing 478 keV peak of ${}^7\text{Li}$ and 197 keV peak of ${}^{19}\text{F}$ used as in situ current normalizer

peak. The normalized count rate of Li ($\text{CPS}_{\text{Li, F}}\text{N}$) is given by,

$$(\text{CPS}_{\text{Li, F}}\text{N}) = \text{CPS}_{\text{Li}} / S_{\text{F}} \quad (1)$$

Concentrations of Li in synthetic and actual samples were obtained by relative method using current normalized CPS of Li as given below.

$$\text{Concentration of Li (mg kg}^{-1}\text{)} = (\text{CPS}_{\text{Li, F}}\text{N}) / (S_{\text{Li, F}}\text{N}) \quad (2)$$

Where $(S_{\text{Li, F}}\text{N})$ is the relative sensitivity of Li with respect to F for the chosen pair of γ -rays (478 keV of Li and 197 keV of F).

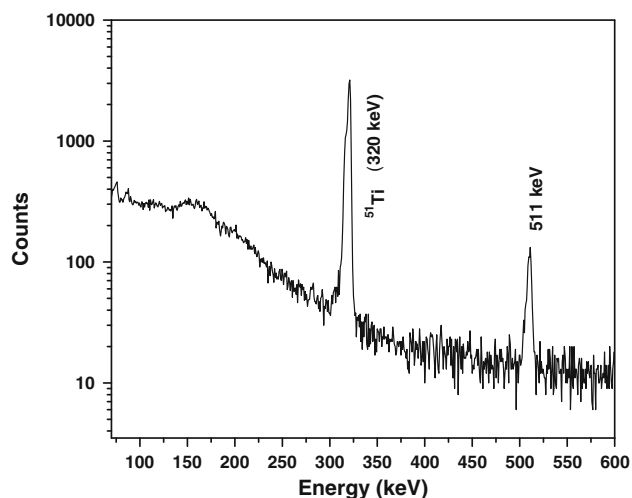


Fig. 4 Gamma-ray spectrum of neutron irradiated lithium titanate sample showing 320 keV peak from ${}^{51}\text{Ti}$ in INAA

Titanium concentration determination by INAA

Titanium concentrations in samples as well as in synthetic samples were calculated by relative method of INAA using the peak area under 320 keV of ${}^{51}\text{Ti}$ as per the following Eq. (3):

$$m_{\text{sam}} = \frac{m_{\text{std}} * \text{CPS}_{\text{sam}} * D_{\text{sam}}}{\text{CPS}_{\text{std}} * D_{\text{std}}} \quad (3)$$

where m , CPS and D are mass of the element, counts per second of activation product of interest and decay factor respectively. Subscripts ‘sam’ and ‘std’ represent for sample and standard respectively. The elemental concentration in wt% is obtained by dividing experimentally determined mass (m_{sam}) of the element with mass of the sample.

Results and discussion

Current normalized count rate of Li at 478 keV peak was obtained using sensitivity of F (S_{F}). *In situ* current normalizer (F) amount was kept constant in all the samples, synthetic samples as well as standards. Sensitivity of Li was obtained using Eq. 1. Concentrations of Li in eight synthetic samples and eleven samples of lithium titanate were determined by relative method using Eq. 2. The lithium concentration values determined in synthetic samples were in the range of 3.2–19 wt%. The plot of determined values versus expected values of Li in synthetic samples is given Fig. 5, which is found to be in good agreement (within $\pm 3\%$) with the expected Li concentrations. The amount of titanium obtained (39.5–50 wt%) by relative method of INAA in three synthetic samples are also in good agreement (within $\pm 2.5\%$) with that of the expected values of titanium as seen in Fig. 6. The uncertainties on determined values of Li and Ti are about 2%.

The results of Li and Ti concentrations (in wt%) obtained for eleven samples by PIGE and INAA, respectively, are given in Table 2 along with Li to Ti mole ratios. The theoretical values of Li and Ti in stoichiometric Li_2TiO_3 are 12.67 and 43.67 wt%, respectively and the Li to Ti wt% ratio and mole ratio are 0.29 and 2.0, respectively. Li concentrations obtained for first four samples (Samples 1–4, sintering temperatures 500–800 °C) with LiCl as the starting material are in the range of 11.71–12.68 wt% which are close to Li concentrations in stoichiometric lithium titanate (12.67 wt%). The determined Ti concentrations by INAA are in the range of 42.7–44.6 wt%. The mole ratios of Li to Ti in these four samples are in the range of 1.9–2.0, indicating products are closer to stoichiometric ones. For samples prepared with LiNO_3 as starting material (Samples 5–8, sintering

temperature 1000–1250 °C), the Li concentrations (wt%) are in the range of 11.2–12.4 wt % and the Ti values are in the range of 44–44.7 wt%. However, the mole ratios of Li/Ti are in the range of 1.75–1.95, which are lower than 2.0 in these four samples. The higher values of mole ratios for samples 5 and 6 are reasonable as these two samples were washed with LiOH after digestion and sintering temperature was 1000 °C. On the other hand, samples 7 and 8 showed lower mole ratio as no washing was done after digestion in addition to the sintering temperature being high (1250 °C). Two mixture samples (samples 9 and 10) were synthesized using 50 % each of LiCl and LiNO₃ in similar way to that of other samples. Sample 9 was prepared without washing after digestion, whereas sample 10 was prepared with washing after digestion and in both cases the sintering temperature was kept 1000 °C. The Li contents were found to be 11.5 and 12.13 wt% for samples 9 and 10 respectively. The Li/Ti mole ratio for sample 10 was found to be higher (1.92) than sample 9 (1.8), as washing was carried out for sample 10. In the recycled batch (sample 11) Li concentration was found to be lower i.e., 11.0 wt% which is expected as recycled Li solution was used for lithium titanate production. This process will help in reducing Li concentration in the wash effluent, which is one of the important aspects of sol–gel based synthesis. Mass balance was checked assuming lithium titanate as combination of Li₂O and TiO₂, and it was found that except for the recycled sample (sample 11), the accounting of mass was found to be in the range of 96–100 %.

The combined percentage uncertainties are about 2 % for Li and 2–3 % for Ti concentrations. The uncertainty components for Li concentrations are due to mass of

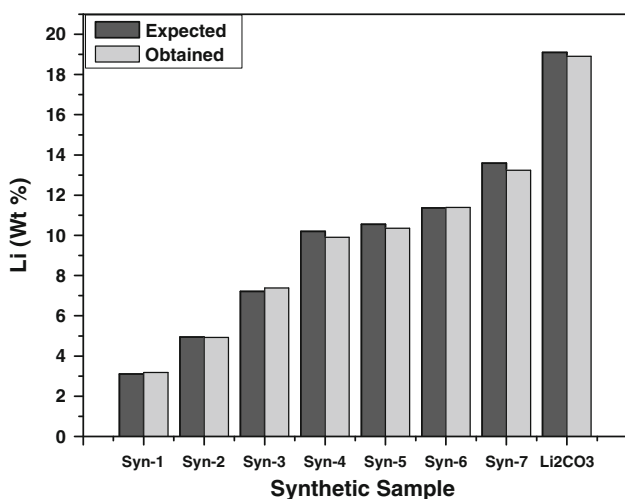


Fig. 5 Comparison of experimentally determined and expected concentration values of Li in synthetic samples and Li₂CO₃: validation of PIGE method

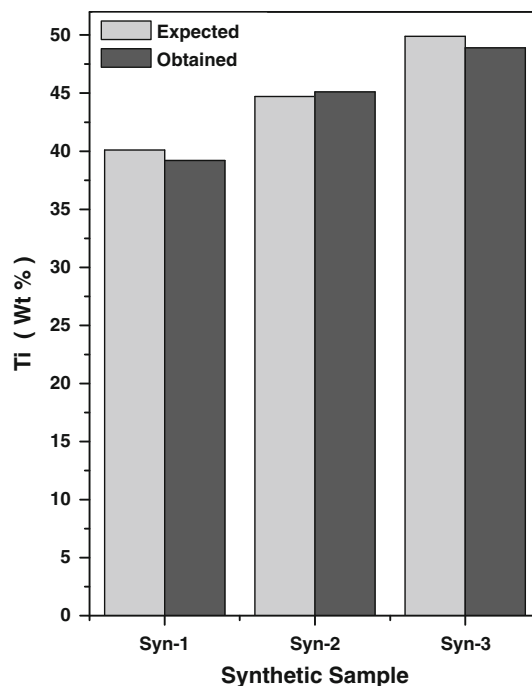


Fig. 6 Comparison of experimentally determined concentration values of Ti with expected values in synthetic samples: validation of INAA method

Li₂TiO₃ (0.1–0.2 %), mass of Li standard (0.1–0.2 %), mass of F as internal standard (0.5–1.0 %), peak areas of corresponding γ -rays of ⁷Li (478 keV) and ¹⁹F (197 keV) in sample and standard (~0.5 %). Similarly in the case of Ti, the uncertainty due to mass of Ti in sample/standard is ~1.0 % and the uncertainties due to peak areas at 320 keV of standard and samples are 1.0 % and 1.0–2.0 %, respectively. Uncertainties on calculated concentrations of Li and Ti (used as standards) have been considered negligible, as stoichiometric compounds were used.

The X-ray diffraction patterns of Li₂TiO₃ samples were recorded to identify the phases present. The XRD pattern was recorded on a STOE (Germany) X-ray powder diffractometer using Cu-K α radiation ($\lambda = 1.5406\text{\AA}$) with a graphite monochromator and nickel filter. The XRD pattern (Fig. 1) matches with the XRD pattern reported for Li₂TiO₃ (PCPDF file No 33-0831) indicating formation of phase-pure compound. It is to be noted that Li₂TiO₃ is not a line compound and has a small range of non-stoichiometry between 1.9 and 2.1 for Li/Ti when the compound is Li₂ ± xTiO₃ ± y [26]. Therefore, for a sample containing only Li, Ti and O mass balance may not come to 100 %. In the case when Li/Ti mole ratio is deviating from the range mentioned above, then the sample may not be monophasic and may contain additional phases like Li₂O or TiO₂ or other possible ternary phases like Li₄Ti₅O₁₂ depending upon Li/Ti ratio. Samples 7, 8 and 10 (having low mole ratios) may contain more than one phase i.e., Li₂TiO₃ with

Table 2 Determined concentration values of Li by PIGE and Ti by INAA in Li_2TiO_3 samples and Li/Ti mole ratios

S. no.	Sample no.	Li (wt%)	Ti (wt%)	Li/Ti mol ratio
1	Sample 1	12.00 ± 0.23	42.7 ± 1.3	1.94
2	Sample 2	11.71 ± 0.22	42.7 ± 1.2	1.90
3	Sample 3	12.68 ± 0.24	43.6 ± 0.9	2.01
4	Sample 4	12.42 ± 0.23	44.6 ± 1.0	1.92
5	Sample 5	12.38 ± 0.23	44.0 ± 0.8	1.94
6	Sample 6	12.31 ± 0.21	44.7 ± 1.1	1.95
7	Sample 7	11.21 ± 0.21	44.0 ± 0.8	1.76
8	Sample 8	11.20 ± 0.21	44.2 ± 1.1	1.75
9	Sample 9	11.50 ± 0.22	44.0 ± 1.4	1.80
10	Sample 10	12.13 ± 0.23	43.5 ± 1.2	1.92
11	Sample 11	11.00 ± 0.22	43.1 ± 0.9	1.76

$\text{Li}_4\text{Ti}_5\text{O}_{12}$. We would like to mention that the main focus of the work was to optimize a simple and sensitive non-destructive method for Li quantification in this important ceramic fusion reactor material.

When LiCl was used as starting material and washing with LiOH was not done, lithium loss was observed in the final product (not shown in Table 2) leading to non-stoichiometric compound. This is because, LiCl melts at 615 °C and then reacts with TiO_2 to give the final product i.e., Li_2TiO_3 . The microspheres thus formed were found to be crack free. When LiNO_3 was used as starting material for Li_2TiO_3 there was no loss of Li observed in the final product up to sintering temperature of 1000 °C. This is because of melting of LiNO_3 at 270 °C and reacting subsequently with TiO_2 to give Li_2TiO_3 at lower temperature of 500 °C, but the disadvantage is the microspheres crack due to exothermic reaction between HMTA and nitrate ion. Thus, attempts were made to prepare the said compound using mixture of LiCl + LiNO_3 (50:50) sintering of 1000 °C. By this modification crack free Li_2TiO_3 microspheres were obtained with 11.5 and 12.13 wt% Li concentrations for without and with washing after digestion, respectively. In some cases, washing was intentionally avoided to reduce Li in the wash effluent. To maintain desired Li/Ti mole ratio in finished product, the sintering temperature should be maintained at 1000 °C. Efforts are being made to optimize the production of crack free lithium titanate by use of mixture of LiCl and LiNO_3 and also it is aimed to use recycled Li from wash effluent.

The in situ current normalization approach for normalized count rate takes care of current fluctuation of proton beam, if any, during the experiment [11]. *In situ* current normalization method is a relatively newer approach and is found to be simple to get current normalized count rate of the analyte of interest compared to the conventional RBS method. Since Li contents are in percentage concentration level, the turn around time for experiment was less by PIGE. Since ^{51}Ti is short-lived, and again INAA method

was faster and the turn around time for analysis was less like that of Li by PIGE. The methods used are appropriate for composition analysis of lithium titanate ceramics as dissolution of sample is not required which is very difficult and invariably gives erroneous results. PIGE can easily be used for quantification of Li in other tritium breeders and simultaneous quantification of Li and Si, Li and Al in Li_4SiO_4 and LiAlO_2 samples, respectively.

Conclusions

A PIGE method using proton beam has been optimized for Li quantification in lithium titanate ceramic samples. The method is simple and non-destructive, appropriate for Li determination in the finished lithium titanate samples providing the advantage of not following wet chemical dissolution. INAA was used for Ti determination in this sample which is simple and non-destructive in nature. There were no gamma-ray interferences for characteristic peaks of Li and Ti in the experiments in PIGE as well as INAA, respectively. To conclude, we provide a comprehensive methodology for the determination of lithium and titanium in sol-gel synthesised lithium titanate which is essential for process optimization as well as quality control work.

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