CHAPTER 4

Preparation and Characterization of Al_2TiO_5 Filler

4.1 Introduction

To date there has been a lot of attention to produce an inexpensive ceramic material which exhibits desirable properties such as resistance to thermal shock, ability to function as thermal insulating material, high mechanical strength, low thermal expansion coefficient, and the capability of achieving little or no shrinkage upon sintering. Utilization of fillers that includes calcium carbonate, carbon fibers, glass fibers, mica, caolin, TiO_2, SiO_2, Al_2O_3 in polymer electrolytes is with the purpose of improving mechanical and electrical properties [Dissanayake et al., 2003].

The present work, focus on the preparation and characterization of aluminum titanate using sol-gel method. Aluminum titanate has been classified as an important high-temperature ceramic due to its low thermal expansion coefficient, low thermal conduction and excellent thermal shock resistance [Buessum et al., 1952]. The structure of the final product (Al_2TiO_5) was determined using X-Ray diffraction (XRD).

4.2 XRD of aluminum titanate

In this study, synthesis of Al_2TiO_5 was done with certain amounts of aluminum nitrate ($Al(NO_3)_3$) and titanium dioxide (TiO_2). After going through reflux process, the solution has changed into honey-like gel. This honey-like gel was then dried on the hot plate. The mixture powder was then sintered for two hours at selected temperatures. Al-

$_2TiO_5$ can be prepared by sintering stoichiometric mixtures of alumina and titania above 1400 ^oC [Freudenberg and Mocellim, 1987]. However, sol-gel techniques are preferable to prepare this ceramic because lower temperatures can be used. Andrianainarivelo et al., (1997), have shown that aluminum titanate can be prepared via non-hydrolytic synthesis at 600 ^oC. Coury et al., (1994) produced Al_2TiO_5 at 800 ^oC by synthesis with alkoxides stabilized with acetylacetone. The sol-gel process synthesis usually leads to a separate crystallization of TiO_2 and Al_2O_3, and Al_2TiO_5 forms at temperatures higher than the eutectoid temperature (1280 ^oC) [Brugger and Mocellin, 1986]. Table 4.1 lists XRD peaks that correspond to the relevant crystallographic planes of of Al_2TiO_5.

Table 4.1: The formation of Al_2TiO_5 at different sintering temperatures.

2θ (°)	**Sintering temperatures**						**Reference**
	750	**850**	**950**	**1100**	**1300**	**1350**	
18.0				/			Stanciu et al., 2004
19.0		/	/			/	Sobhani et al., 2008, Joe et al., 1997
26.0						/	Yang et al., 2009
26.5				/			Stanciu et al., 2004
27.0	/	/	/				Sobhani et al., 2008
27.5					/	/	Joe et al., 1997
30.6				/			Stanciu et al., 2004
33.0						/	Yang et al., 2009
33.72	/	/	/				Sobhanni et al., 2008
34.0				/	/	/	Stanciu et al., 2004, Joe et al., 1997
34.8		/	/				Sobhani et al., 2008
35.15							Remy et al., 1989
36.15							Remy et al., 1989
37.8		/	/				Sobhani et al., 2008
38.7				/			Stanciu et al., 2004
41.0						/	Yang et al., 2009
41.3						/	Yang et al., 2009
42.0				/			Stanciu et al., 2004
43.0			/	/			Sobhani et al., 2008, Stanciu et al., 2004

Table 4.1 continued…

43.4				/			Stanciu et al., 2004
46.0						/	Yang et al., 2009
48.0				/	/	/	Stanciu et al., 2004, Joe et al., 1997
48.8		/	/				Sobhani et al., 2008
50.2						/	Yang et al., 2009
51.0				/	/	/	Stanciu et al., 2004, Joe et al., 1997
52.6		/	/				Sobhani et al., 2008
52.8				/			Stanciu et al., 2004
54.3				/			Stanciu et al., 2004
54.45			/				Sobhani et al., 2008
55.0					/	/	Joe et a., 1997
57.5				/			Stanciu et al., 2004
57.55			/				Sobhani et al., 2008
57.7					/		Joe et al., 1997
59.0			/				Sobhani et al., 2008
63.5			/				Sobhani et al., 2008
64.05				/			Stanciu et al., 2004
66.55				/			Stanciu et al., 2004
68.3				/			Stanciu et al., 2004
72.8				/			Stanciu et al., 2004
74.0			/				Sobhani et al., 2008

Fig. 4.1 shows the XRD patterns of the Al_2TiO_5 ceramics sample with different moles of aluminum nitrate sintered at 1050 °C. Peaks of Al_2TiO_5 can be observed at 2θ = 35.15°, 36.15° [Remy et al., 1989], 27.5° [Joe et al., 1997], 37.8°, 52.6°, 54.45°, 57.55° [Sobhani et al., 2008], 25.6°, 41.3° [Yang et al., 2008], 43.4°, 64.05°, 66.55°, 68.3° and 77.2° [Stanciu et al., 2004]. With more aluminum nitrate (AN) added, intensity of the peaks sharpens.

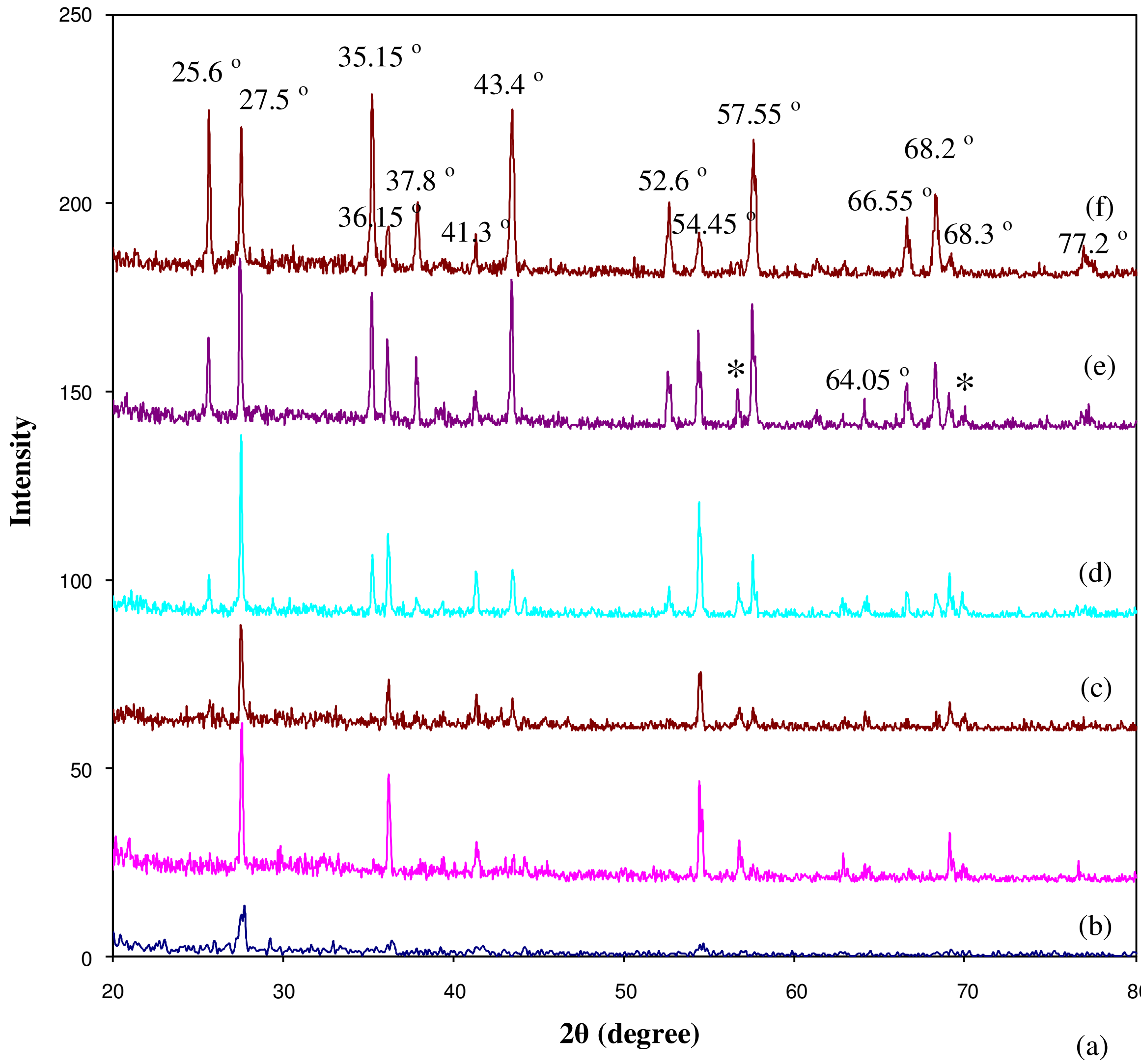

Fig. 4.1: XRD patterns for sample containing (a) 0.01 mole AN (b) 0.02 mole AN (c) 0.04 mole AN (d) 0.06 mole AN (e) 0.08 mole AN (f) 0.09 mole AN sintered at 1050 °C (*) indicate TiO_2 peaks.

From Table 4.1 and Fig. 4.1, only samples prepared using 0.08 and 0.09 moles of aluminum nitrate showed the most number of peaks that correspond to the various crystallographic planes of Al_2TiO_5. In this work, the mixture with 0.08 mole of aluminum nitrate was prepared again in order to determine the lowest sintering temperature for the formation of Al_2TiO_5 and sintering time two hours. This composition was chosen and not the composition using 0.09 mole of aluminum nitrate because in the XRD pattern for precursors sintered at 1050 ^oC the product formed using

0.08 mole aluminum nitrate showed a peak at 2θ = 63.95° near to 64.05° reported by Stanciu et al., 2004 and Sobhani et al., 2008 which is not present in the diffractogram for sample using 0.09 mole of aluminum nitrate. The diffractogram for the product formed using 0.08 and 0.09 mole aluminum nitrate still show traces of TiO_2 as can be implied from peaks at 2θ = 56.7°, 68.35° and 69.25° [Stanciu et al., 2004]. The crystallite size for the prepared samples was calculated using the peak at 2θ = 27.5° since this peak is present in the diffractogram of all samples shown in Fig. 4.1. Table 4.2 lists the crystallite size of Al_2TiO_5 with varied amounts of aluminum nitrate. The addition of aluminum nitrate increased the crystallite size of Al_2TiO_5 structure.

Table 4.2: The crystallite size of Al_2TiO_5.

No. of moles (Al_2O_3:TiO_2)	Crystallite size (nm)
0.01:0.09	46.75
0.02:0.08	116.88
0.04:0.06	95.63
0.06:0.04	161.83
0.08:0.02	123.76
0.09:0.01	161.88

In the study, to determine the lowest sintering temperature, the sintering temperature was varied from 750 °C up to 1050 °C. Fig 4.2 presents the XRD pattern of the 0.08 Al_2O_3-0.02 TiO_2 ceramic sample at different sintering temperatures. The sintering step was done at different temperatures in an attempt to obtain the lowest formation temperature with minimal residual α-alumina and rutile TiO_2 in the Al_2TiO_5 powder due to incomplete reaction of the components [Skala et al., 2009]. The XRD studies show that the product obtained by heating the precursor up to 850 °C is amorphous. The XRD pattern of the precursor heated at 1050 °C shows complete formation of Al_2TiO_5. The diffractograms clearly show that the aluminum titanate lines are much stronger in the XRD spectrum for the sample sintered at 1050 °C. Thus from the study, the sintering temperature cannot be lower than 1050 °C since even at 1050 °C there are still traces of rutile TiO_2 present.

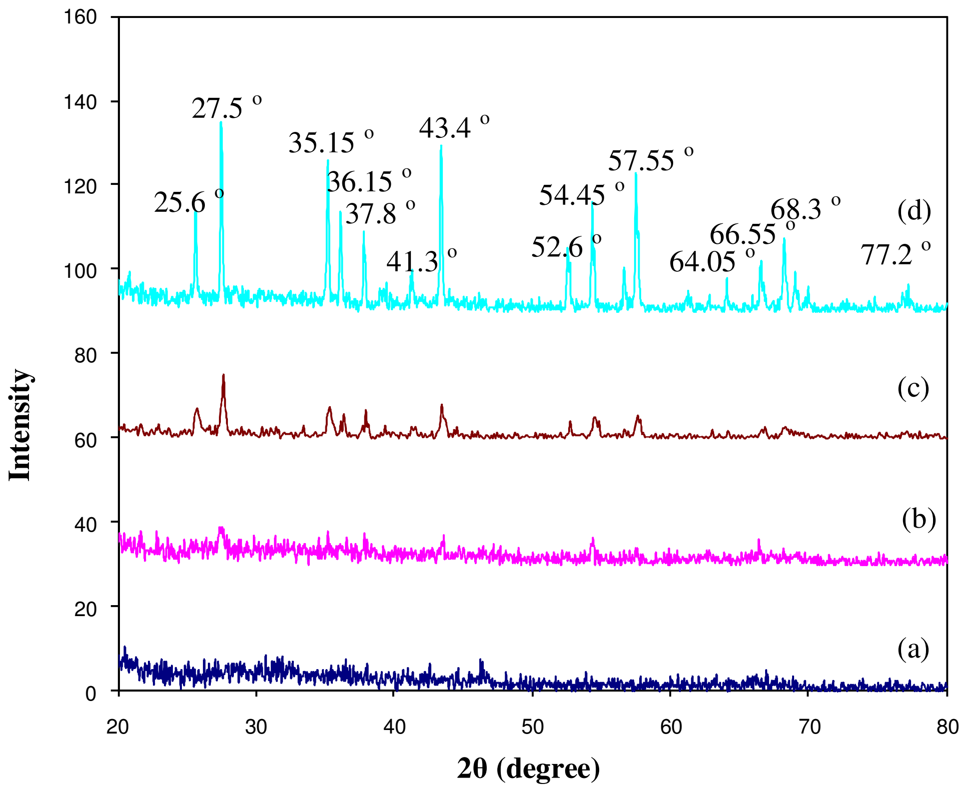

Fig. 4.2: XRD patterns for sample containing 0.08 mole AN with different sintering parameter (a) 750 °C (b) 850 °C (c) 950 °C (d) 1050 °C.

4.3 Summary

Aluminum titanate has been successfully prepared using sol-gel method at precursor sintering temperature of 1050 °C. The best composition for the preparation of Al_2TiO_5 is mixing $Al(NO_3)_3$ and TiO_2 in the ratio of $Al(NO_3)_3$:TiO_2 = 4:1. The diffractogram of the product formed using this composition contains peaks corresponding to that of Al_2TiO_5 with some traces of rutile TiO_2. From the results of this chapter, the first objective in the preparation of Al_2TiO_5 has been achieved.