Chapter 4

Results and Discussion

4.1 Experimental results of pellet specimens

4.1.1 Density and water absorption

Specific gravity, water absorption and relative density were measured as shown in Appendix 1. The relationship between relative density and sintering temperature is shown in Fig.4.1.1 (a). Relative density increased with sintering temperature. Density reached to almost full at 1600 ^{o}C because AKP-30 has very small grain size, which is a good property to enhance sintering. Fig.4.1.1 (b) shows the relationship between water absorption and sintering temperature.

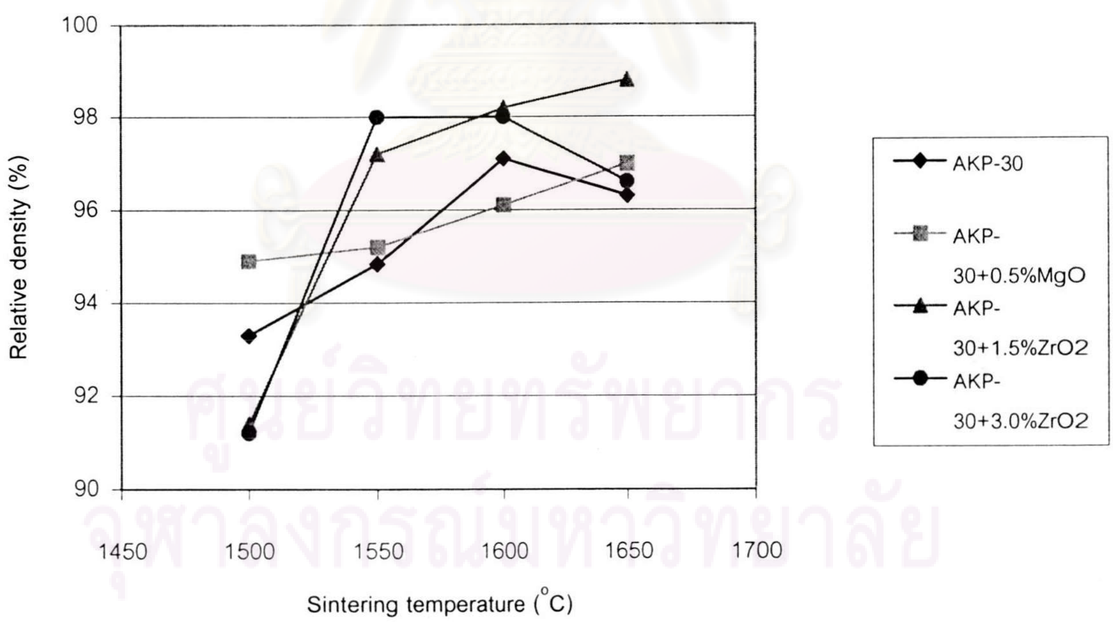

Fig.4.1.1 (a) Relationship between relative density and sintering temperature

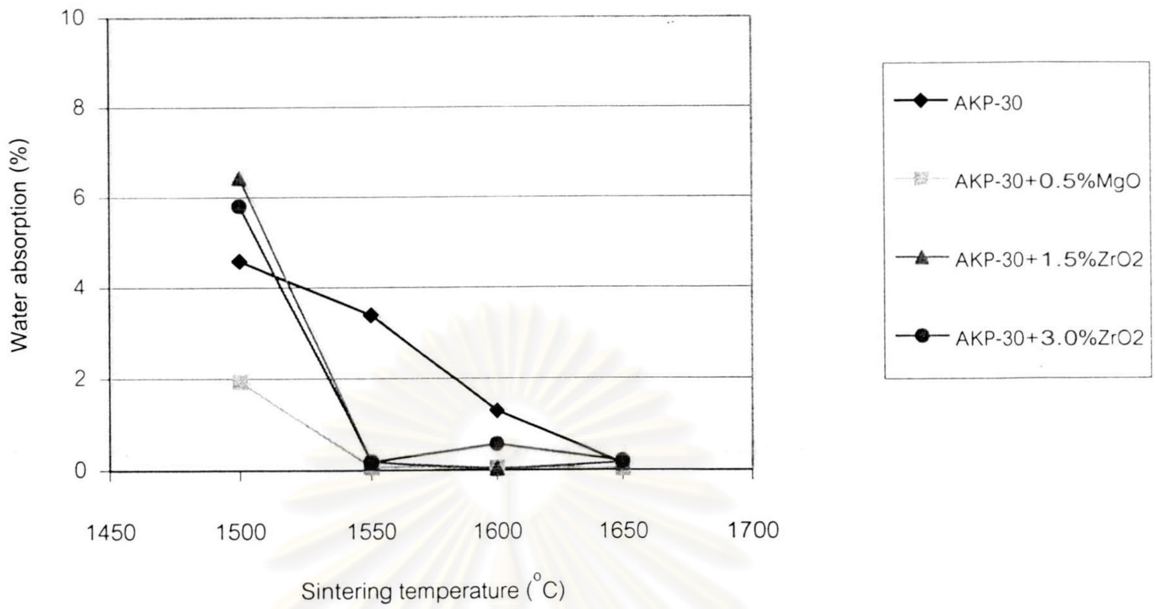

Fig.4.1.1 (b) Relationship between water absorption and sintering temperature

Fig.4.1.1 (a) and Fig.4.1.1 (b) show that the relationship between the relative density and water absorption are normal. The doped compositions show a little higher relative density than the undoped composition. Pure AKP-30 (undoped composition) also shows a little higher water absorption than doped composition. However, as shown in Fig. 4.1.3.3, water absorption of pure AKP-30 is almost zero at the sintering temperature over 1600 ^{o}C. Tape specimen shows very low water absorption at the sintering temperature of 1550 ^{o}C as shown in Fig.4.2.2.2.

4.1.2 Thermal conductivity

In section 4.1.1, AKP-30 can be sintered to almost full density at 1600 ^{o}C. To know the effect of bulk density on the thermal conductivity, the specimens for measuring thermal conductivity were sintered at 1450 - 1650 ^{o}C. For this experiment, 12 mm diameter die was used because the diameter of specimen, which will be measured for thermal conductivity should be in the range of 9.5-10.5 mm diameter. Three to five pieces of specimens for each composition were sintered. The results of specific gravity, water absorption, relative density of this set of specimens are shown in Appendix 2 A.

The thermal conductivity of the material (λ) is dependent on the density (ρ), thermal diffusivity (α) and the specific heat of material (C_p) from the relation

$$\lambda = \alpha\, c_p \rho$$

Thermal conductivity of pure AKP-30 and doped AKP-30 sintered at various temperatures are shown in Table 4.1. Those were calculated from Thermal diffusivity, specific heat and bulk density values are as shown in Appendix 2 B.

Table 4.1 Thermal conductivity of doped and pure AKP-30 alumina specimens

Composition	Sintering Temp.(^{o}C)	Bulk density (g/cm^3)	Thermal conductivity (W/m.K)
AKP-30	1450	3.901	32.8
	1500	3.958	35.6
	1550	3.915	37.4
AKP-30 +0.5% MgO	1500	3.697	24.2
	1550	3.879	34.2
	1600	3.885	35.9
	1650	3.945	36.4
AKP-30 +1.5 ZrO_2	1550	3.867	34.1
	1600	3.852	36.2
	1650	3.942	33.8
AKP-30 +3.0 ZrO_2	1550	3.822	34.2
	1600	3.893	30.0
	1650	3.949	33.9

Thermal diffusivity, which means transient heat flow conditions is an important factor required for getting good thermal conductivity. The density is also an important factor as shown in Fig.4.1.2.1.

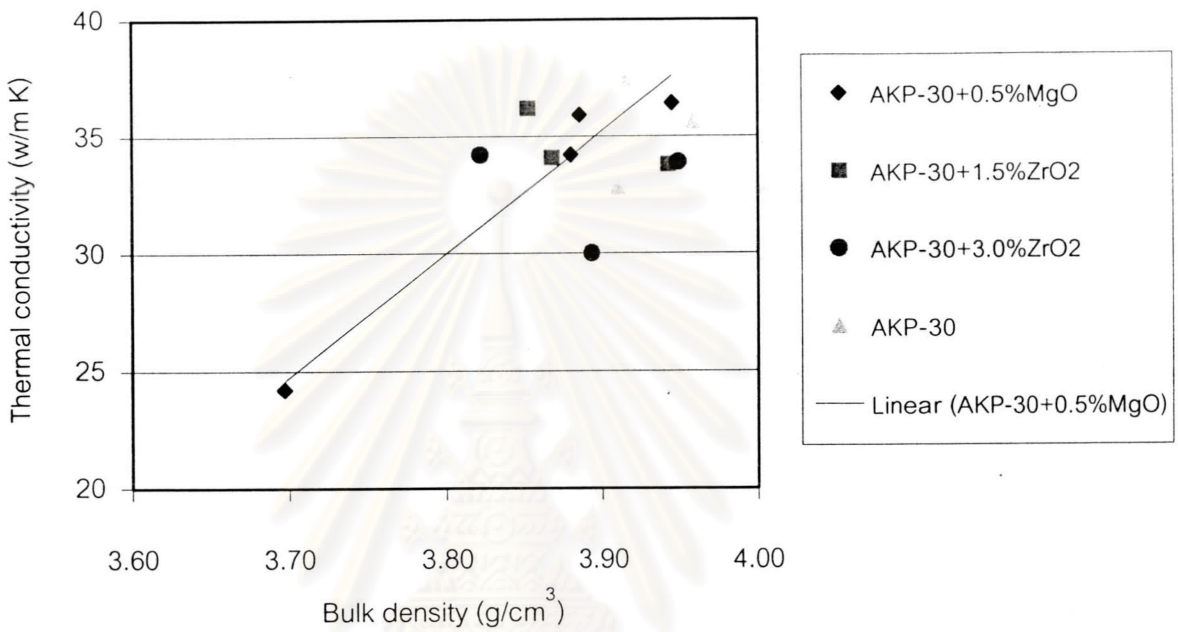

Fig.4.1.2.1 Relationship between bulk density and measured thermal conductivity

The thermal conductivity of a composite depends on the microstructure when the second phase disperse in a matrix, the thermal conductivity of the composite (K_m) is shown as in the equation

$$K_m = \frac{K_c \bullet 1+2\mathbf{V}_d(1-K_c/K_d)/(2K_c/K_d+1)}{1-\mathbf{V}_d(1-K_c/K_d)/(2K_c/K_d+1)} \qquad (5.1)$$

K_c is the thermal conductivity of matrix, K_d is the thermal conductivity of dispersed phase and ν_d is the volume fraction of dispersed phase. When $K_c >> K_d$ the following equation is derived.

$$K_m = K_c\left[(1-\nu_d)/(1+(\nu_d/2))\right] \quad (5.2)$$

In this experiment, ZrO_2 and pore are second phase. The thermal conductivity of ZrO_2 is 3 W/m•K that is lower than that of Al_2O_3, 40 W/m•K. The experimental results can be calculated by the equation 5.2. The volume of ν_d is the sum of pore volume and ZrO_2 volume, From the relative density and ZrO_2 content, ν_d is calculated. All data used for calculation and results are shown in Table 4.2.

Table 4.2.Thermal conductivity of doped and pure AKP-30 alumina based on equation 5.2

Composition	Sintering temperature (°C)	Relative density	1- Relative density	Vol. of ZrO_2	ν_d	K_m (W/m. K)
AKP-30	1450	0.99	0.01	0.00	0.01	39.4
	1500	1.00	0.00	0.00	0.00	40.0
	1550	0.99	0.01	0.00	0.01	39.4
AKP-30 +0.5% MgO	1500	0.93	0.07	0.00	0.07	35.9
	1550	0.98	0.02	0.00	0.02	38.8
	1600	0.98	0.02	0.00	0.02	38.8
	1650	0.99	0.01	0.00	0.01	39.4
AKP-30 +1.5 ZrO_2	1550	0.97	0.03	0.01	0.04	37.7
	1600	0.97	0.03	0.01	0.04	37.7
	1650	0.99	0.01	0.01	0.02	38.8
AKP-30 +3.0 ZrO_2	1550	0.95	0.05	0.02	0.07	35.9
	1600	0.97	0.03	0.02	0.05	37.0
	1650	0.99	0.01	0.02	0.03	38.2

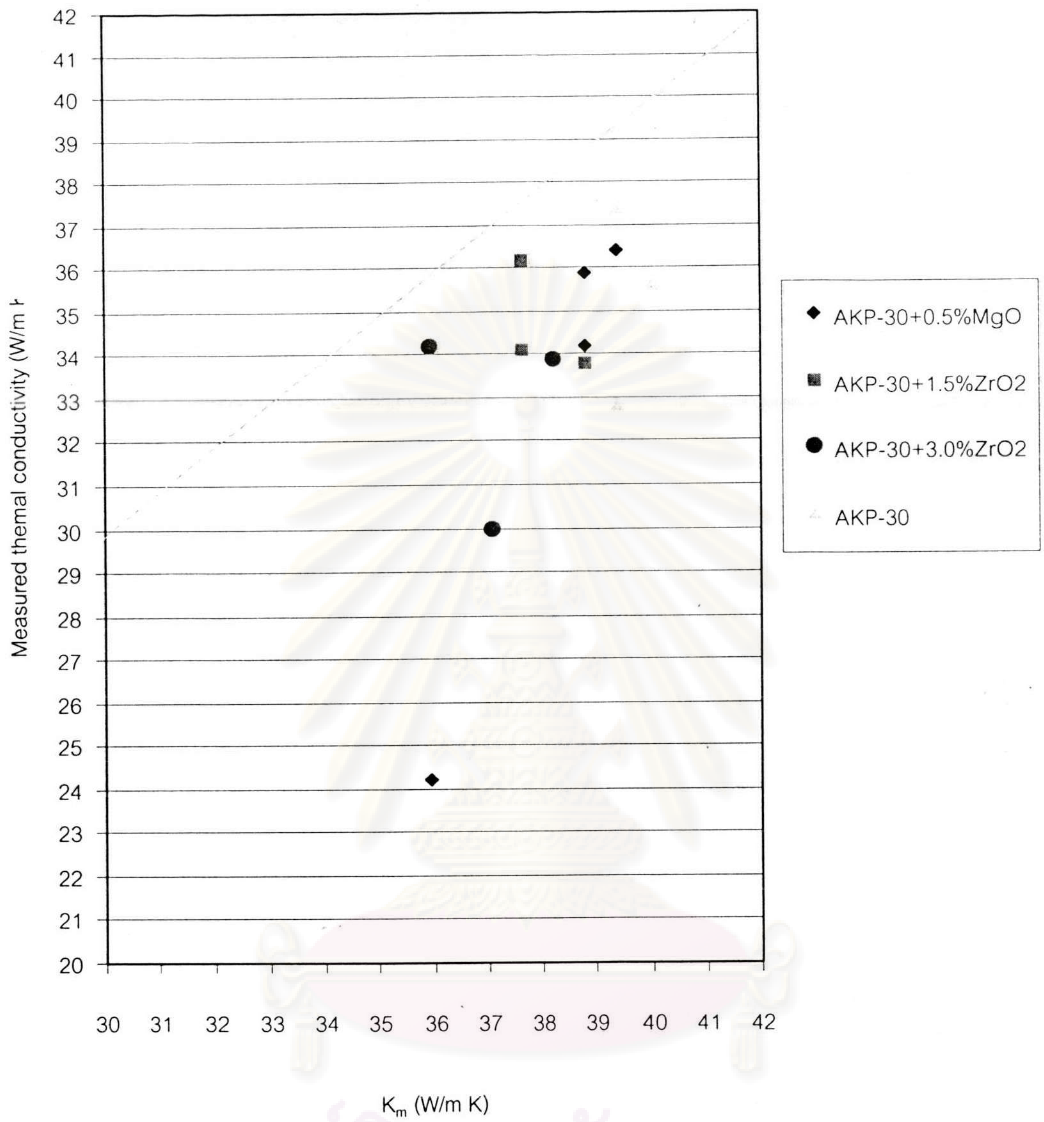

Fig.4.1.2.2. Relationship between measured thermal conductivity and K_m

Fig.4.1.2.2 shows the relationship between measured thermal conductivity and K_m. When pores and ZrO_2 grains are dispersed in the alumina matrix like the model for equation 5.2, measured thermal conductivity and K_m should be equal. The data should be plotted on the dashed line in Fig.4.1.2.2. However, measured values are always a little smaller than K_m as seen in Fig.4.1.2.2.

Thermal conductivity of AKP-30 are mostly higher than 30 W/m•K for all compositions except AKP-30 +0.5 MgO composition, which was sintered at 1500 ^{o}C that showed very low thermal conductivity related to its low density. Except, this pure AKP-30 sintered at 1550 ^{o}C shows the highest thermal conductivity.

4.1.3 Mechanical strength

Relative density, water absorption of specimens which are used for strength measurement are shown in Appendix 3. Thickness and radius of each specimen were used to calculate mechanical strength by equation (3.5). The relationship between mechanical strength and sintering temperature is shown in Fig.4.1.3.1 and Appendix 4. As shown in Appendix 3, Fig.4.1.3.2 and 4.1.3.3, specimens sintered at 1500 ^{o}C are low in relative density and high in porosity. Presence of the pores is thought to be the cause of lower strength in every doped composition except in pure AKP-30 composition, which shows very high mechanical strength at sintering temperature of 1500 ^{o}C with the low relative density. Specimens usually break due to presence of pores, inclusions and surface cracks. Fracture surface was observed to detect fracture origin using an optical microscope. However, we could not find the obvious fracture origin due to the white color of specimens.

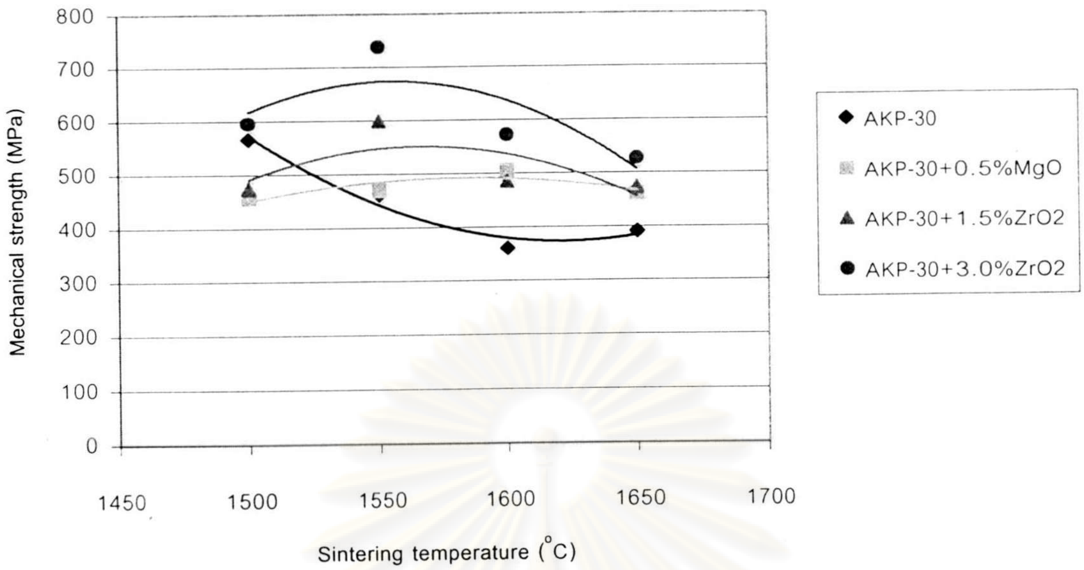

Fig.4.1.3.1 Relationship between strength and sintering temperature of AKP-30

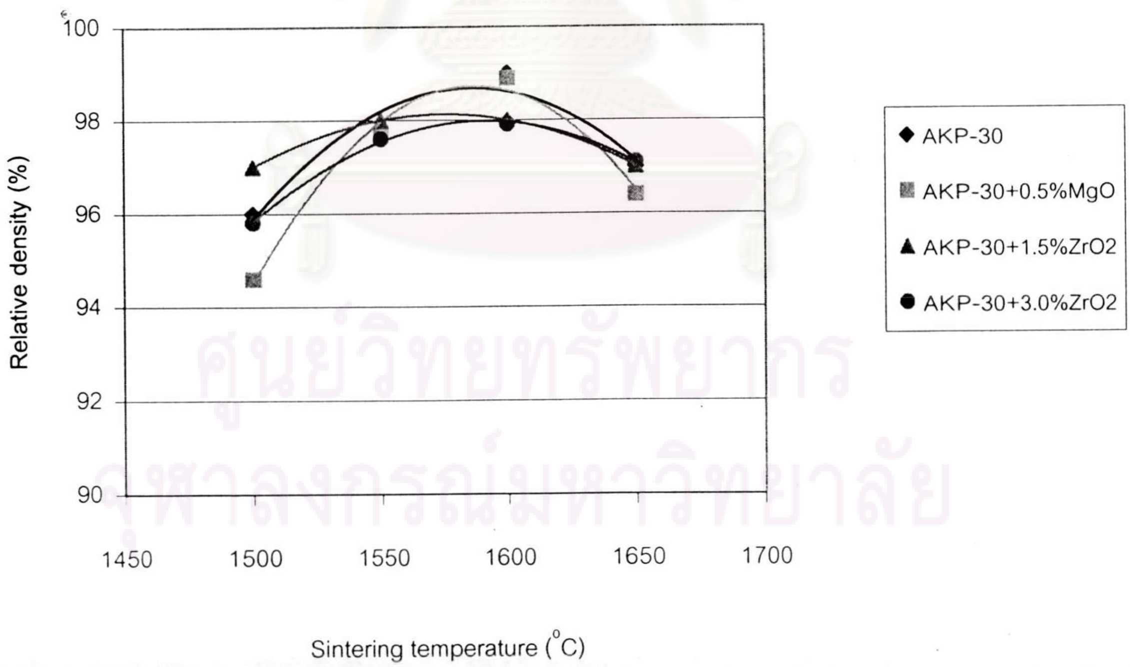

Fig.4.1.3.2 Relationship between relative density and sintering temperature of AKP-30.

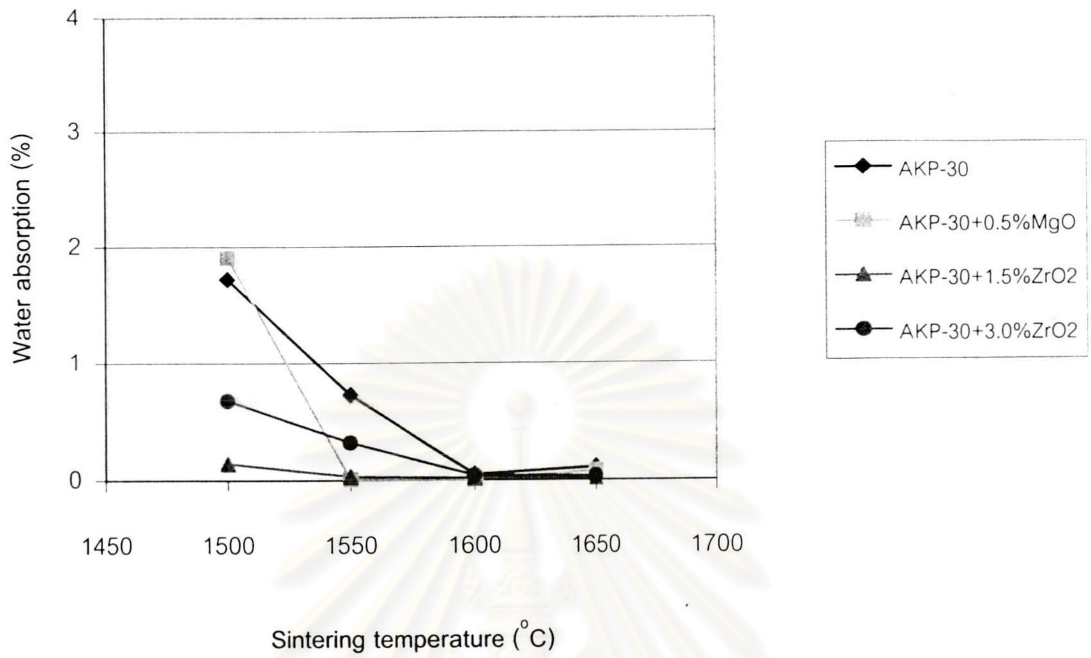

Fig.4.1.3.3 Relationship between water absorption and sintering temperature of AKP-30

The mechanical strength was measured in conformity with ASTM. The increase of mechanical strength in most compositions is in the same tendency of density. AKP-30 + 3.0% ZrO_2 composition shows the highest value related to its grain size and density as shown in Fig.4.1.3.5. The pure AKP-30 shows the abnormal behavior that the density increases with sintering temperature, however its mechanical strength decreases with temperature. The causes of high mechanical strength in pure AKP-30 composition may come from the very small grain size as shown in Fig.4.1.3.4 and Fig 4.1.4.1.

The mechanical strength of the specimen sintered at 1550 ^{o}C is the highest in average. Most of specimens attained over the target value, 400 MPa and the best strength is in the range of 700 –800 MPa.

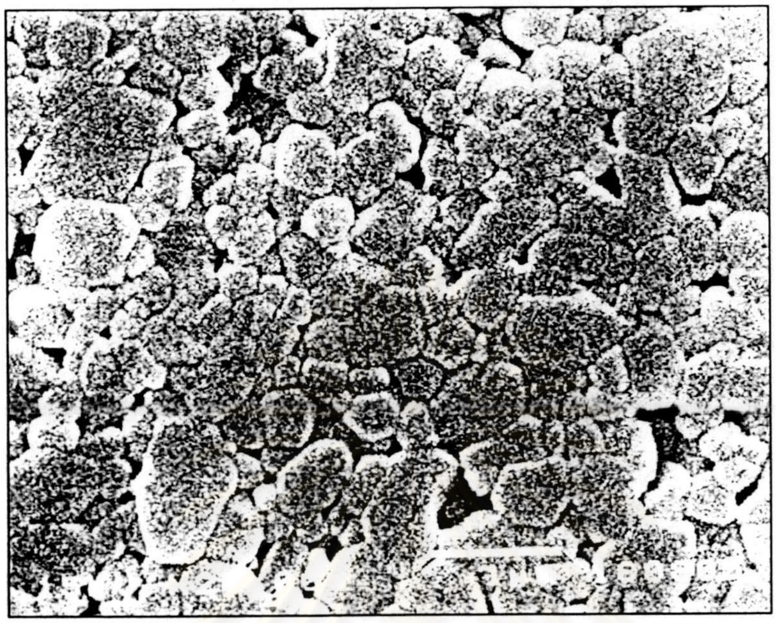

Fig.4.1.3.4 SEM micrograph of pure AKP-30 sintered at 1500 °C

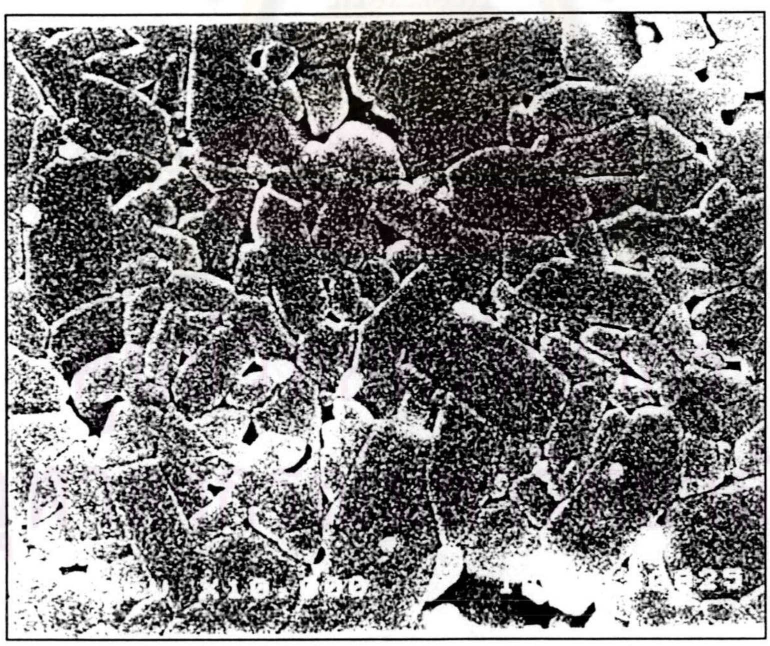

Fig.4.1.3.5 SEM micrograph of AKP-30 +3.0%ZrO_2 sintered at 1550 °C

4.1.4. Microstructure

Microstructure of Pure AKP-30 and doped AKP-30 which were sintered at various temperatures were examined using a scanning electron microscope (SEM) and the average grain size was determined by a line intercepted method following ASTM standard (Designation E 112-96). Results are shown in Table 4.3.

Table 4.3 The average grain size of AKP-30 at various sintering temperature

Composition	Sintering temperature (°C)	Average grain size (micron)
AKP-30	1500	0.33
	1550	0.48
	1600	0.54
	1650	1.25
AKP-30+0.5 MgO	1500	0.80
	1550	0.87
	1600	1.71
	1650	1.85
AKP-30+1.5 ZrO2	1500	1.03
	1550	1.01
	1600	1.80
	1650	2.05
AKP-30+3.0 ZrO2	1500	0.54
	1550	0.92
	1600	1.56
	1650	1.33
AKP-30 (tape)	1450	0.26
	1500	0.30
	1550	0.38

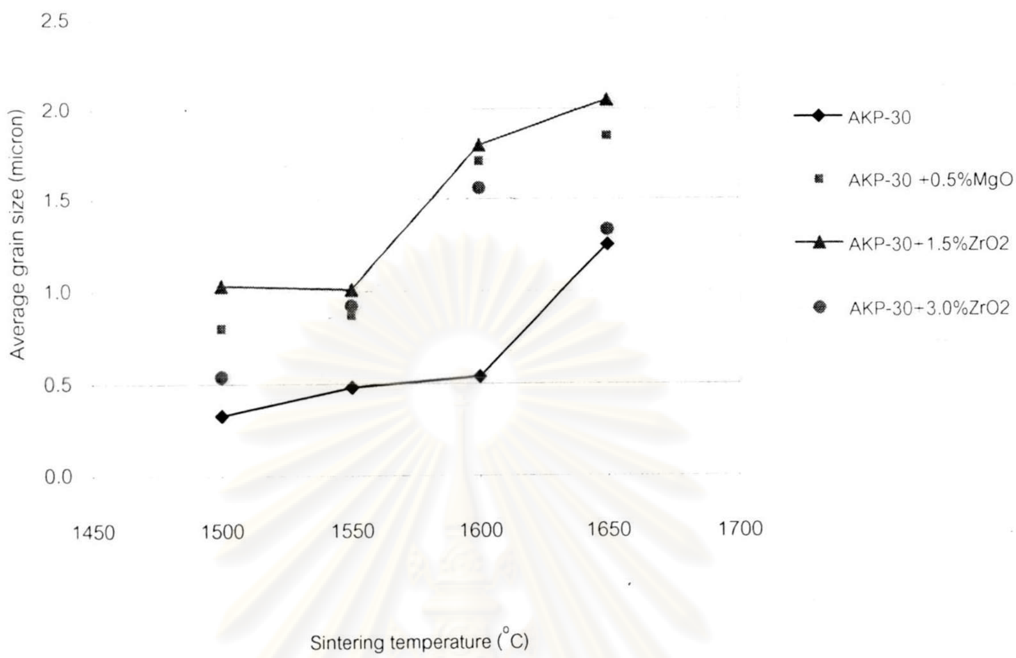

Fig.4.1.4.1 Relationship between average grain size of sintered alumina and sintering temperature

The SEM micrographs of specimens are shown in Appendix 6. The grain size of every composition increases by the increasing of sintering temperature as shown in Fig.4.1.4.1. Pure AKP-30 shows the smallest grain size compared to other compositions. The average grain size of pure AKP-30 thin tape in Table 4.3 also shows smaller grain that should be the good characteristic to achieve high mechanical strength.

MgO and ZrO_2 were used as additives in this experiment. Generally, the effect of MgO is to eliminate the discontinuous grain growth during the late stage of sintering and also affect to decrease the average grain size and that of ZrO_2 is to inhibit grain growth and also decrease the average grain size. From the experimental results, MgO and ZrO_2 could not act to decrease the average grain size completely. This may come from the incomplete dispersion of additives.

4.2 Experimental results of tape specimens

4.2.1 Thermal analysis (DTA/TGA)

Thermal analysis curves by DTA/TGA of Yuken tape and Miyazaki tape are shown in Fig.4.2.1.1 and Fig.4.2.1.2, respectively.

Fig 4.2.1.1 describes the thermal behavior of Yuken binder tape. Three endothermic peaks at 150.5 °C, 211 °C and 269.8 °C show that Yuken binder is composed of at least three kinds of organic materials. The weight loss is about 8 percent.

Fig 4.2.1.2 describes the thermal behavior of Miyazaki binder tape. Two endothermic peaks at 182 °C and 287.9 °C showed that the Miyazaki binder is composed of 2 types of chemicals and the weight loss is about 9.1 percent.

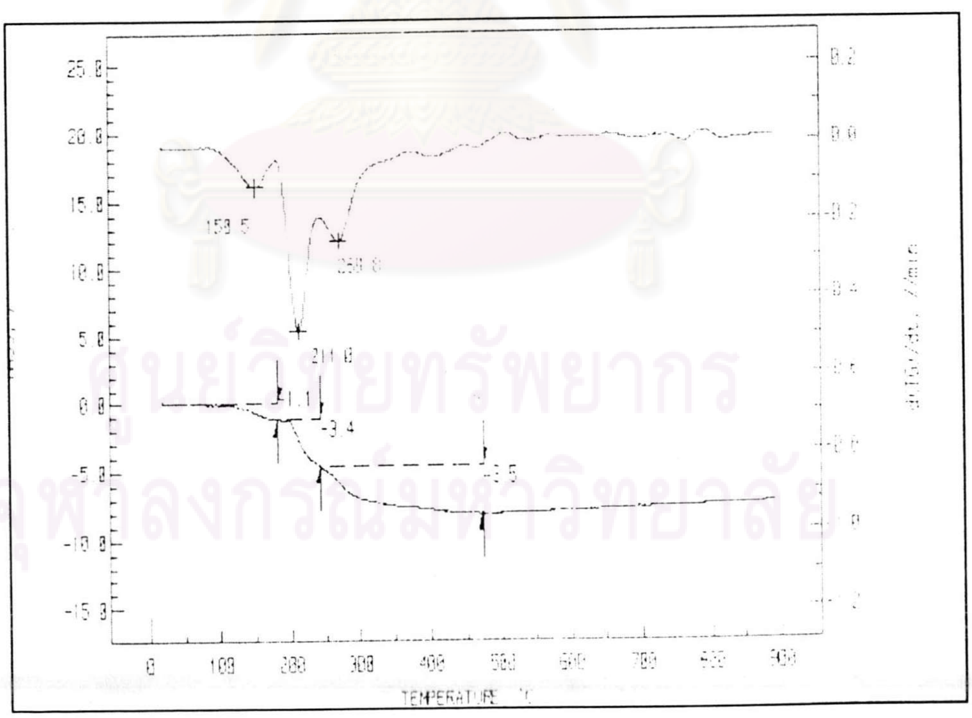

Fig 4.2.1.1 DTA/TGA curve of Yuken tape

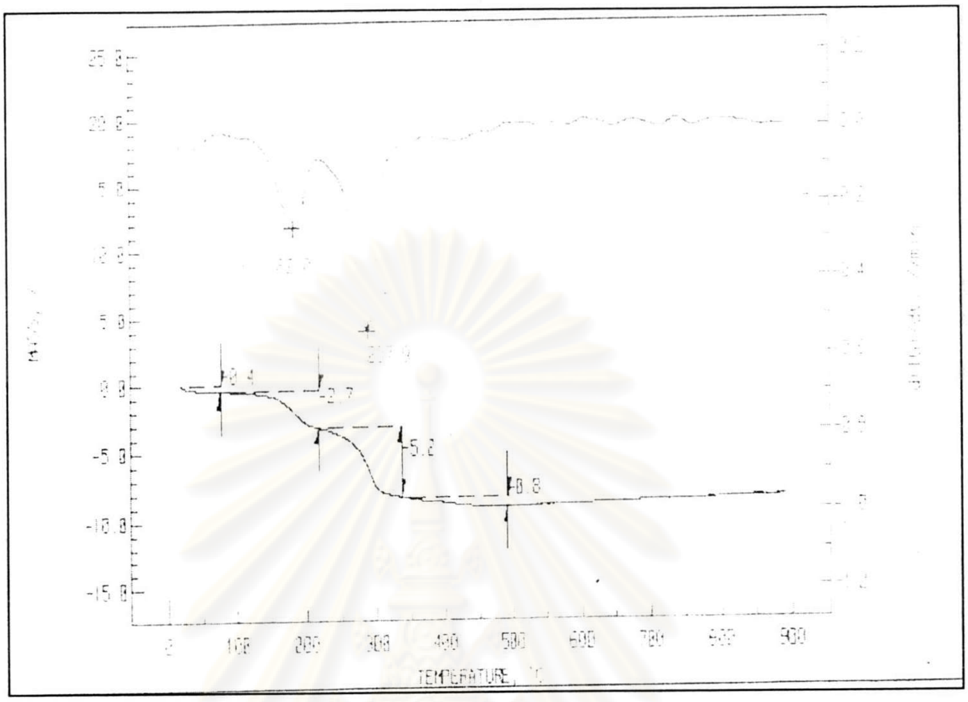

Fig 4.2.1.2 DTA/TGA curves of Miyazaki tape

Thermal analysis data is very important in the design of sintering schedule. In both tape specimens, high weight loss temperature is around 300 °C that the binder should be removed at this temperature and then it becomes constant over 500 °C

The binder removal is a very important step in sintering process because it affects the cracks and the defects in the sintered tape. So that the temperature at 300 °C is selected for binder removal.

4.2.2 Sintering of tape specimens

In the sintering process, flatness of sintered tape is very important. Some load was applied on the top of stacked tapes for preventing the warping. The stack of 5 tape specimens were prepared by sprinkling with Zirconia powder in between each tape. The specimens were heated at 300 ^{O}C for 1 h with a heating rate of 3 ^{O}C/min for binder removal. Further the temperature was raised with a heating rate of 3 ^{O}C/min to 1500, 1550, 1600 and 1650 ^{O}C and soaked for 2 h. The sintered samples were cooled to 35 ^{O}C in the furnace.

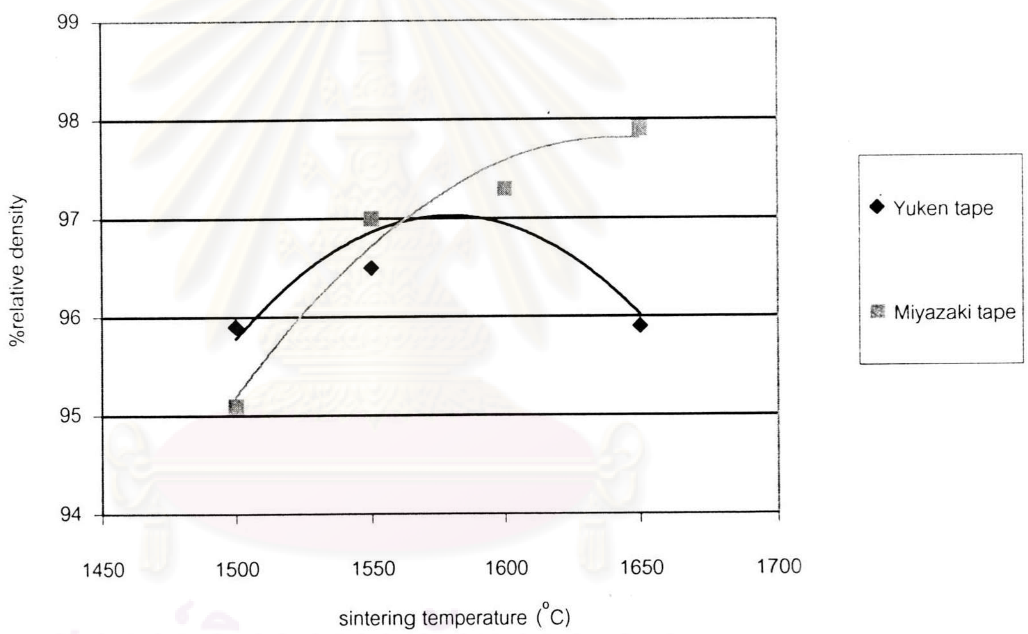

Fig.4.2.2.1 Relationship between relative density and sintering temperature of alumina tape

Relative density and water absorption of sintered tape are shown in Appendix 8. The relationship between relative density and sintering temperature is shown in Fig. 4.2.2.1. The relative density of Yuken tape increases at the temperature range of 1500 – 1600 ^{O}C and then decreases at 1650 ^{O}C. On the other hand, the relative density of Miyazaki tape increases with sintering temperature and becomes the highest density at 1650 ^{O}C.

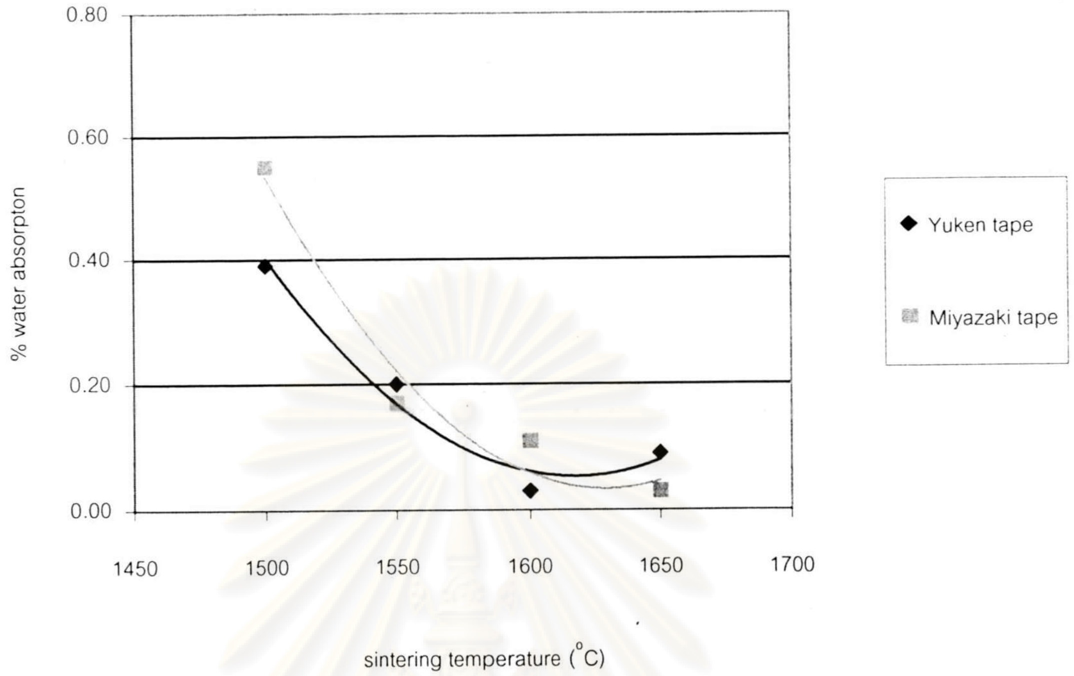

Fig.4.2.2.2 Relationship between water absorption and sintering temperature of alumina tape

The relationship between water absorption and sintering temperature of tapes were shown in Fig.4.2.2.2. The tendency of water absorption shown in the Fig 4.2.2.2 conforms inversely with the tendency of density in Fig. 4.2.2.1 that the water absorption decease when the sintering temperature increases.

Weight loss of sintered tapes was determined after sintering as shown in Appendix 9. The relationship between weight loss and sintering temperature are shown in Fig.4.2.2.3. Weight loss after sintering is rather constant, around10 –11 %. The weight loss of Miyazaki tape is a little higher than that of Yuken tape and is of the same tendency with thermal analysis.

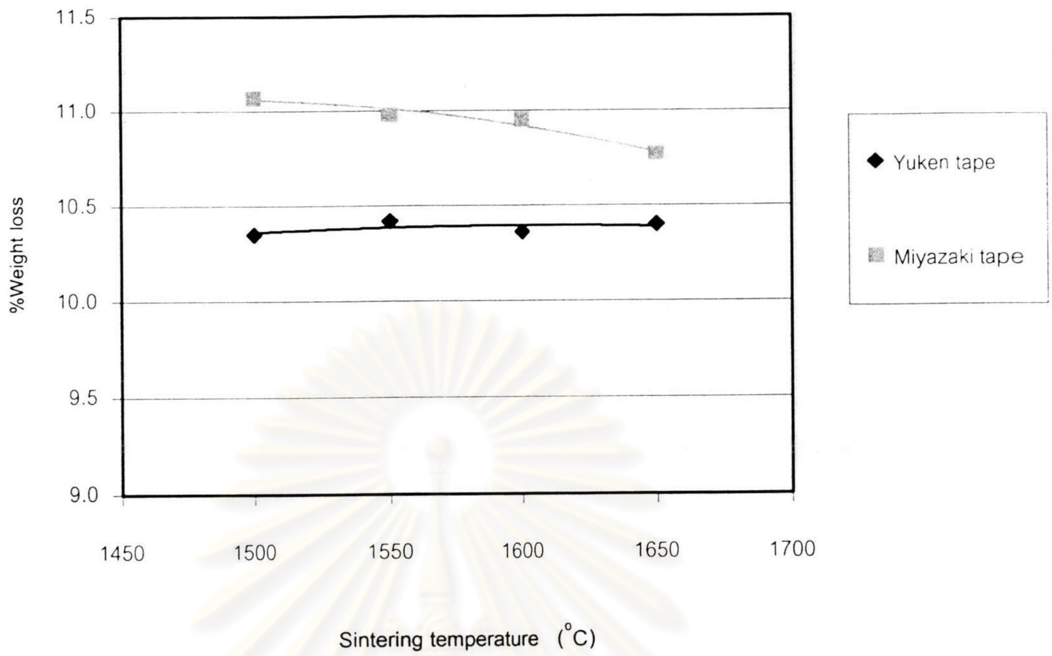

Fig.4.2.2.3. Relationship between weight loss and sintering temperature of alumina tape

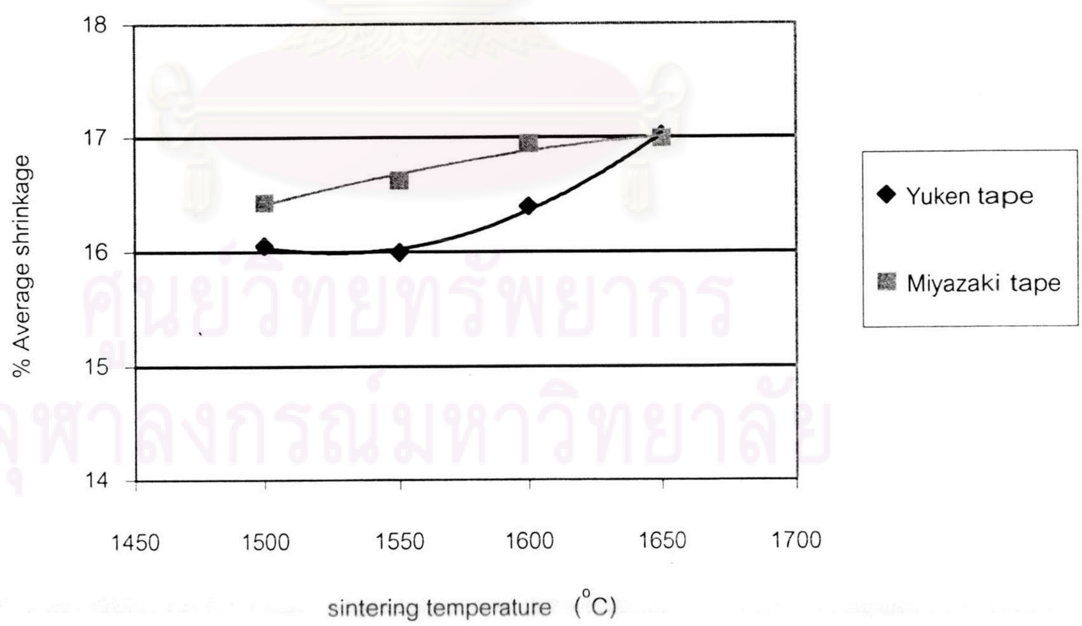

Fig.4.2.2.4. Relationship between the average shrinkage and sintering temperature of alumina tape

Shrinkage of sintered tape was determined as shown in Appendix 10. The relationship between shrinkage and sintering temperature is shown in Fig. 4.2.2.4. The sintered tape has symmetrical shrinkage to vertical and parallel axis around 16 –17 %. The shrinkage of Miyazaki tape is a little higher than that of Yuken.

4.2.3 Mechanical strength of sintered tape specimens

The mechanical strength of tape specimens were measured using biaxial - flexure strength–test fixture (Appendix 13) in conformity with ASTM F 394-78.

Table 4.4 Mechanical strength of sintered tape specimens

Composition	Sintering temperature (°C)	% Relative density	Average mechanical strength (MPa)
Yuken	1550	98.4	438
Yuken	1600	98.4	427
Miyazaki	1550	98.2	465
Miyazaki	1600	99.0	430

Relative density and water absorption of test specimens are shown in Appendix 11 A. Applied load, thickness and mechanical strength are shown in Appendix 12 A.

The average mechanical strength of sintered tapes is shown in Table 4.4 and Fig.4.2.3.1. Most of the sintered tapes have mechanical strength values higher than 400 MPa. The size of specimen is around 2.5 cm in both width and length.

Data of the mechanical strength of sintered tape is shown in Fig.4.2.3.1.and the range is 300 – 600 MPa.

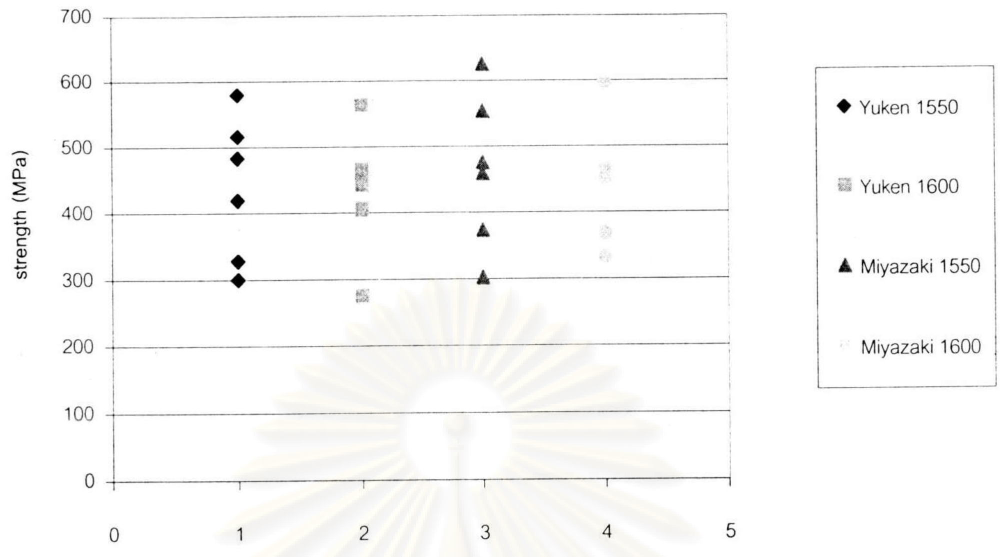

Fig.4.2.3.1 Mechanical strength of sintered tape specimens

The mechanical strength is almost same with that of AISIN samples, 491 MPa, as shown in Table 4.5 The size of AISIN specimens are 3×3 cm.

In this biaxial test, it was supposed that the size of specimen might affect the strength value. Therefore, new specimens with the same size as AISIN's were prepared in Japan (OTAKE CERAM Co., Ltd.)

Table 4.5 Mechanical strength of sintered tape specimens (controlled size)

Composition	Sintering temperature (°C)	% Relative density	Average mechanical strength (MPa.)
Yuken	1450	96.8	633
Yuken	1500	99.0	715
Yuken	1550	99.2	645
Miyazaki	1450	96.7	849
Miyazaki	1500	98.9	845
Miyazaki	1550	99.2	848
Aisin	unknown	93.4	491

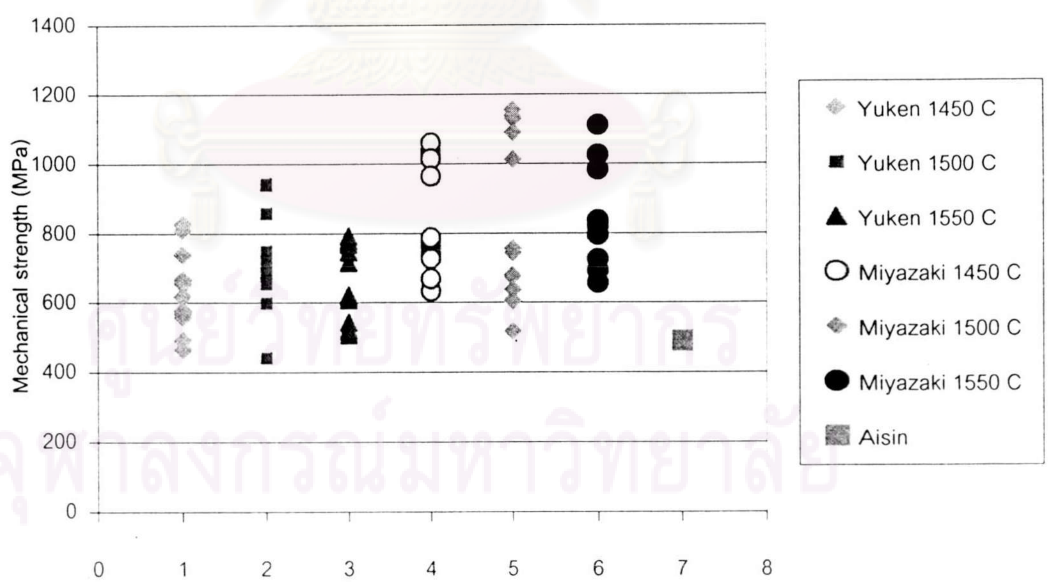

Fig.4.2.3.2 Mechanical strength of sintered tape (controlled sized)

Relative density and water absorption of test specimens are shown in Appendix 11 B. Applied load, thickness and mechanical strength are shown in Appendix 12 B.

The average mechanical strength of sintered tapes (controlled size) is shown in Table 4.5 and Fig.4.2.3.2. The mechanical strength value of most sintered tapes is higher than 500 MPa. They are also related with their densities. The strength of Miyazaki specimens is the highest obviously following the tendency in Table 4.2.3.2 and Fig.4.2.3.2. The bigger specimens can stand a higher load because the outer area supporting specimen from the transformation.

The chemicals in each binder influence the mechanical strength of sintered tape. Miyazaki binder enhances the mechanical strength better than Yuken binder as shown in the results.