PREPARATION, CHARACTERIZATION AND APPLICATION OF NANOSILICA AND NANOSILICA XEROGEL FROM BAMBOO LEAVES

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Abstract

Giant bamboo (Wa-bo-gyi) is the largest member of the grass family in Myanmar. In this research, the preparation and characterization of the nanosilica extracted from bamboo leaves (*Dendrocalamas giganteus* Munro, Wa-bo-gyi) were studied. At first, bamboo leaves ash was prepared from bamboo leaves by calcination at various temperatures with different times. Characterization of bamboo leaves ash samples was carried out by using EDXRF, XRD, SEM and FT IR techniques. The XRD data for all of these synthesized samples showed the silica with nanosize. From EDXRF analysis, the presence of SiO_2, K_2O, P_2O_5, CaO of bamboo leaves was found. The maximum relative abundance of SiO_2 was found to be 52.29 % at the calcination temperature of 1000 °C for 2 h. The physicochemical properties of the selected sample (calcined at 1000 °C for 2 h), were 0.37 % of moisture, 3.08 % of ash, 1.44 g/mL of bulk density and 10.03 pH of Silica xerogel powder was prepared from the selected sample by using 1 M NaOH and 6 M H_2SO_4 solution. The prepared silica powder was characterized by EDXRF, XRD, SEM and FT IR analyses. The particle size of xerogel was slightly increased (30.5 nm) and the relative abundance of silica was significantly promoted to 84.15 %. The selected sample was introduced into the formulation of cement. The quality of cement (soundness, normal consistency, setting time, compressive strength and tensile strength) improved when mixing with the selected ash sample.

Keywords: *Dendrocalamas giganteus* Munro, nanosilica, xerogel

Introduction

Bamboo is probably the fastest-growing and highest yielding natural resource and construction material available to mankind. However, the use of bamboo generates other residues not used as fibers, such as the bamboo leaf. In some countries, significant amounts of bamboo are processed, generating high volumes of solid waste. These wastes are often burnt in open landfills, negatively impacting the environment.

The main objective of the present work is to extract nanosilica from natural sources bamboo leaves. In the present research, the bamboo leaves sample was collected from Mayangone Township, Yangon Region. The preparation to get bamboo leaves ash was worked in muffle furnace from 400 and 900 °C for 1 h and 2 h, respectively. In addition, the ash sample was also prepared at 1000 °C for 1 h, 2 h, 3 h and 4 h.

Bamboo leaves ash is black or grey colour. By controlling the burning conditions like temperature and time, amorphous silica of ultrafine size and reactivity will be produced. Bamboo leaves ash is formed by silica with a completely amorphous nature (Aye Aye Aung, 2014). Silica (SiO_2) is the major component in bamboo ash and other oxides such as CaO, K_2O , Al_2O, SO_3, Fe_2O_3, MgO and MnO are present in it (Cocina, 2010).

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Nanosilica has been prepared through dissolution and precipitation process. The dissolution of silica was carried out by using an alkali leaching process (1 M NaOH) to partially dissolved carbonaceous materials. The resulting sodium silicate was filtered and dried in an oven for 24 h at 100 °C. The precipitation of silica from sodium silicate solution was carried out using 6 M H_2SO_4 at the pH of 7 and left for aging 12 h. The obtained silica gel was centrifuged and washed with hot water and dried at 80 °C for 24 h to get silica powder (Vaibhav, 2014).

Nanosilica is used mainly in concrete mixture for construction industry, nanosilica possess more pozzolanic nature (Itler, 1979). It has the capability to react with the free lime during the cement hydration and forms C-S-H get giving strength, impermeability and durability to concrete. In addition, nanosilca can be widely used porcelain, plaster, batteries, paints, adhesives, cosmetics, glass, steel, chemical fiber and environmental protection to upgrade the quality (Wang, 2004).

Materials and Methods

Sample Collection and Preparation

Bamboo leaves were collected from Mayangone Township, Yangon Region. Bamboo leaves were washed with water several times to remove dirt particles. They were dried at room temperature for two weeks.

Preparation of Bamboo Leaves Ash from Bamboo Leaves

Silicon dioxide reduction was conducted by ashing the bamboo leaves in two stages, first by combusting the dried bamboo leaves and then ashing using a muffle furnace. Dried bamboo leaves were weighed and burned in open space for pre-ash sample. Then this pre-ash was calcined in a muffle furnace at a temperature of 400 °C, 500 °C, 600 °C, 700 °C, 800 °C and 900 °C with holding time of 1 h and 2 h, respectively. The obtained ash samples were designated as BLA-1(1) and BLA-1(2) for calcination temperature of 400 °C and calcination time of 1 h and 2 h, respectively. Similarly, BLA-2(1) and BLA-2(2), BLA-3(1) and BLA-3(2), BLA-4(1) and BLA-4(2), BLA-5(1) and BLA-5(2), BLA-6(1) and BLA-6(2) were prepared at the calcination temperature of 500 °C, 600 °C, 700 °C, 800 °C and 900 °C, respectively. In addition, the char was calcined in a furnace at a temperature of 1000 °C with holding time of 1 h, 2 h, 3 h and 4 h until it became a grey ash and which were respectively denoted as BLA-7(1), BLA-7(2), BLA-7(3) and BLA-7(4). Once calcined, the ash was ground and sieved with 60 mm mesh. The sample was put in sealed plastic bag and this bag was stored in the desiccator at room temperature.

Characterization of the Prepared Bamboo Leaves Ash Samples (BLAs)

The prepared ash samples were characterized by EDXRF, XRD, SEM and FT IR analyses. According to the results of analyses, BLA-7(2) was selected for the preparation of nanosilica xerogel.

Analyses of Physicochemical Properties of BLA-7(2)

Some physicochemical properties such as moisture content, ash content, bulk density and pH of the selected sample BLA-7(2) were determined by conventional methods.

Preparation of Nanosilica Xerogel from Bamboo Leaves Ash BLA-7 (2)

A 2 g of bamboo leaves ash (the selected sample) and 20 mL of 1 M NaOH solution were mixed in a beaker (250 mL, pyrex). The mixed solution was set up for 24 h. Then obtained sodium silicate solution was filtered with Whatmann No. 41 filter paper into a beaker and dried in an oven for 24 h at 100 °C. The precipitation of silica from sodium silicate solution was made by adding 6 M H_2SO_4 to pH 7 and 24 h. The precipitated nanosilica was carefully centrifuged and washed with hot water to avoid the loss of residue. The residue on the filter paper was washed with hot water several times. Finally, the residue was dried in an oven at 80 °C for 24 h to give silica xerogel powder.

Characterization of Silica Xerogel

The prepared silica xerogel powder was characterized by EDXRF, XRD, SEM and FT IR analyses**.**

Results and Discussion

Preparation of Bamboo Leaves Ash

Bamboo leaves were collected from Mayangone Township, Yangon Region. The collected sample was washed with water, air dried in an open vessel and pre-ashed on electrical hot plate for 2 h. Then the pre-ashed sample of bamboo leaf was calcined in muffle furnace at the temperatures of 400 - 900 °C for 1 h and 2 h and 1000 °C for 1 - 4 h respectively to obtain bamboo leaves ash. Totally 16 ash samples were prepared. The percent yields of ash from the Wa-bo-gyi samples were obtained in the range of 1.24 – 46.0 %.

Characterization of Bamboo Leaves Ash

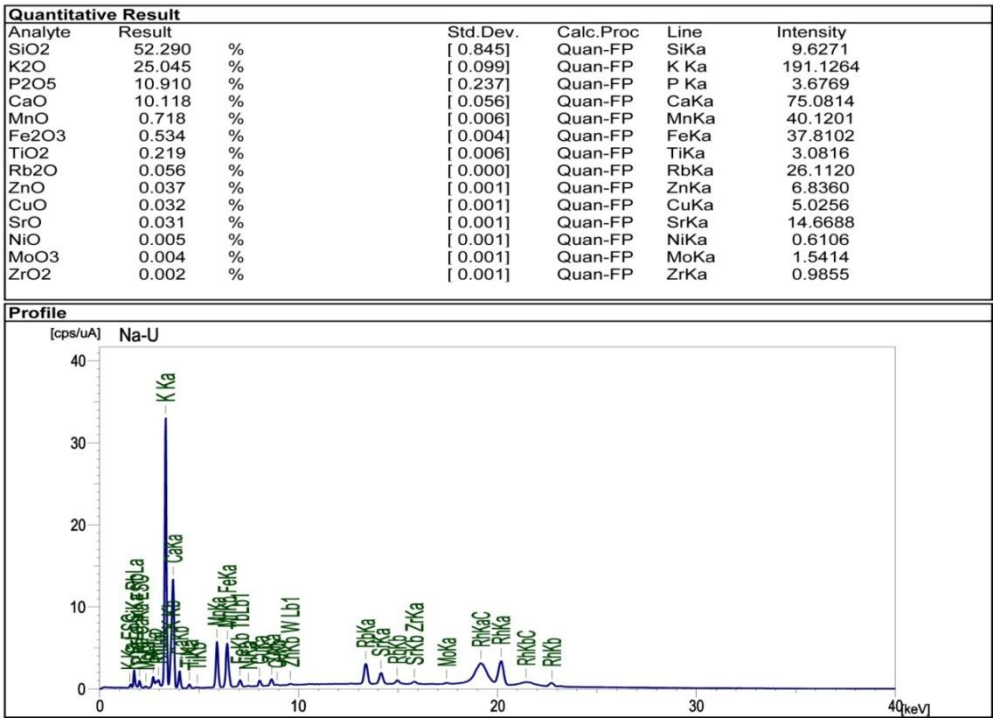
EDXRF analysis

The elemental composition of bamboo leaves ashes were determined by EDXRF analysis. These spectra showed that SiO_2, K_2O, CaO and P_2O_5 are the major constituents in BLA samples. Other oxides such as MnO, Fe_2O_3, CuO, TiO_2 and PbO in BLA were also found below 1 %. The silica percent of different bamboo leaves ashes are shown in Table 1.

Table 1 Relative Abundance of Silica in the Different Bamboo Leaves Ash by EDXRF Analysis

Sr. No.	Sample	Calcination time (h)	Calcination temperature (°C)	SiO_2 (%)
1	BLA-1(1)	1	400	31.279
2	BLA-1(2)	2	400	35.073
3	BLA-2(1)	1	500	33.862
4	BLA-2(2)	2	500	34.703
5	BLA-3(1)	1	600	31.809
6	BLA-3(2)	2	600	32.983
7	BLA-4(1)	1	700	30.597
8	BLA-4(2)	2	700	33.862
9	BLA-5(1)	1	800	37.258
10	BLA-5(2)	2	800	35.388
11	BLA-6(1)	1	900	40.374
12	BLA-6(2)	2	900	45.662
13	BLA-7(1)	1	1000	35.264
14	BLA-7(2)	2	1000	52.290
15	BLA-7(3)	3	1000	41.430
16	BLA-7(4)	4	1000	50.972

Among these 16 ash samples, the maximum relative abundance of SiO_2 was achieved to be 52.29 % at the calcination temperature of 1000 °C for 2 h. So BLA-7(2) is silica rich sample. BLA-7(2) was selected for the preparation of nanosilica xerogel powder.



Quantitative Result

Analyte	Result		Std.Dev.	Calc.Proc	Line	Intensity
SiO2	52.290	%	[0.845]	Quan-FP	SiKa	9.6271
K2O	25.045	%	[0.099]	Quan-FP	K Ka	191.1264
P2O5	10.910	%	[0.237]	Quan-FP	P Ka	3.6769
CaO	10.118	%	[0.056]	Quan-FP	CaKa	75.0814
MnO	0.718	%	[0.006]	Quan-FP	MnKa	40.1201
Fe2O3	0.534	%	[0.004]	Quan-FP	FeKa	37.8102
TiO2	0.219	%	[0.006]	Quan-FP	TiKa	3.0816
Rb2O	0.056	%	[0.000]	Quan-FP	RbKa	26.1120
ZnO	0.037	%	[0.001]	Quan-FP	ZnKa	6.8360
CuO	0.032	%	[0.001]	Quan-FP	CuKa	5.0256
SrO	0.031	%	[0.001]	Quan-FP	SrKa	14.6688
NiO	0.005	%	[0.001]	Quan-FP	NiKa	0.6106
MoO3	0.004	%	[0.001]	Quan-FP	MoKa	1.5414
ZrO2	0.002	%	[0.001]	Quan-FP	ZrKa	0.9855

Figure 1 EDXRF spectrum of BLA-7(2)

XRD analysis

X-ray powder diffraction (XRD) is a analytical technique primarily used for phase identification of a crystalline material and can provide information on unit cell dimensions. X-ray diffraction is a common technique for the study of crystal structures, atomic spacing and crystallite size. Almost all of the diffractograms indicated that the prepared ash samples were crystalline nature and mixture of different types of silica. The diffractograms of BLA samples, the diffraction angles (2q) of all the major peaks were observed around 20° and 30° which are the presence of quartz and tridymite phase. The crystallite sizes of these 16 ash samples were calculated by Scherrer equation. The sizes of all samples were observed within the range of nano scale. The crystallite size of BLA-7(2) was found to be 19.6 nm. It was found to be the smallest crystallite size among 16 ash samples. Therefore BLA-7(2) was selected for the preparation of nanosilica xerogel powder. Figure 2 shows the XRD diffractogram of BLA-7(2) sample.

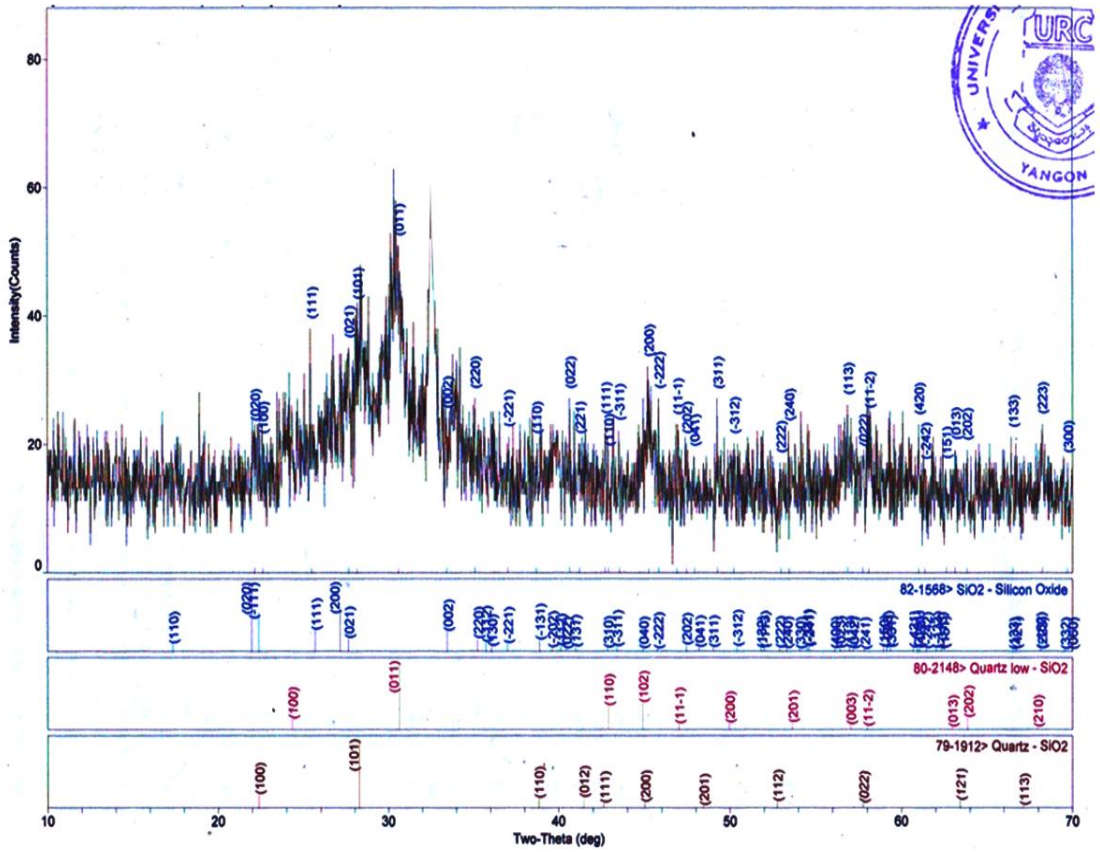


Figure 2 XRD diffractogram of BLA-7(2)

SEM analysis

The SEM micrographs of bamboo leaves ash samples showed porous nature at the calcinations temperature of 400 - 900 °C, however with rising calcinations temperature and calcinations time the ash samples (1000 °C for 1 h, 2 h , 3 h and 4 h) were seen to be more rigid.

At lower temperature of calcinations at 400 °C, 500 °C, the semi-ash or primary char of bamboo leaves did not show well defined pores. At the temperatures of 600 °C, 700 °C, 800 °C and 900 °C the surface morphology of the bamboo leaves ash were observed a distinct appearance of pores. The bamboo leaves ash at these temperatures were used as good sorbent for the removal of heavy metals and degradation of organic dye. At higher calcination temperature (at 1000 °C), the bamboo leaves were found to be completely changed to ash. At the calcination temperature 1000 °C, the samples were more rigid than the previous samples. Figure 3 shows the SEM micrograph of BLA-7(2).

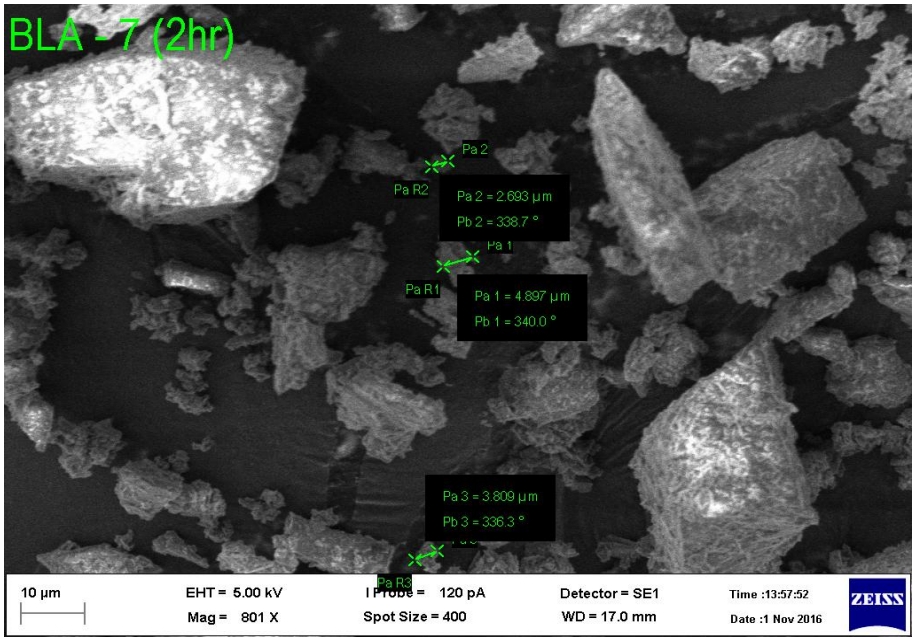


Figure 3 SEM micrograph of BLA-7(2)

FT IR analysis

Figure 4 shows the FT IR spectrum of BLA-7(2). The wave number at 3477 cm^{-1} is related to stretching vibration of Si-OH group. The wave number at 1661 cm^{-1} is related to bending vibration of Si-OH group. The wave number at 1057 cm^{-1} was observed as asymmetric stretching vibration of Si-O-Si group. The wave number at 616 cm^{-1} was observed as bending vibration of Si-O bond. FT IR data assignments are shown in Table 2.

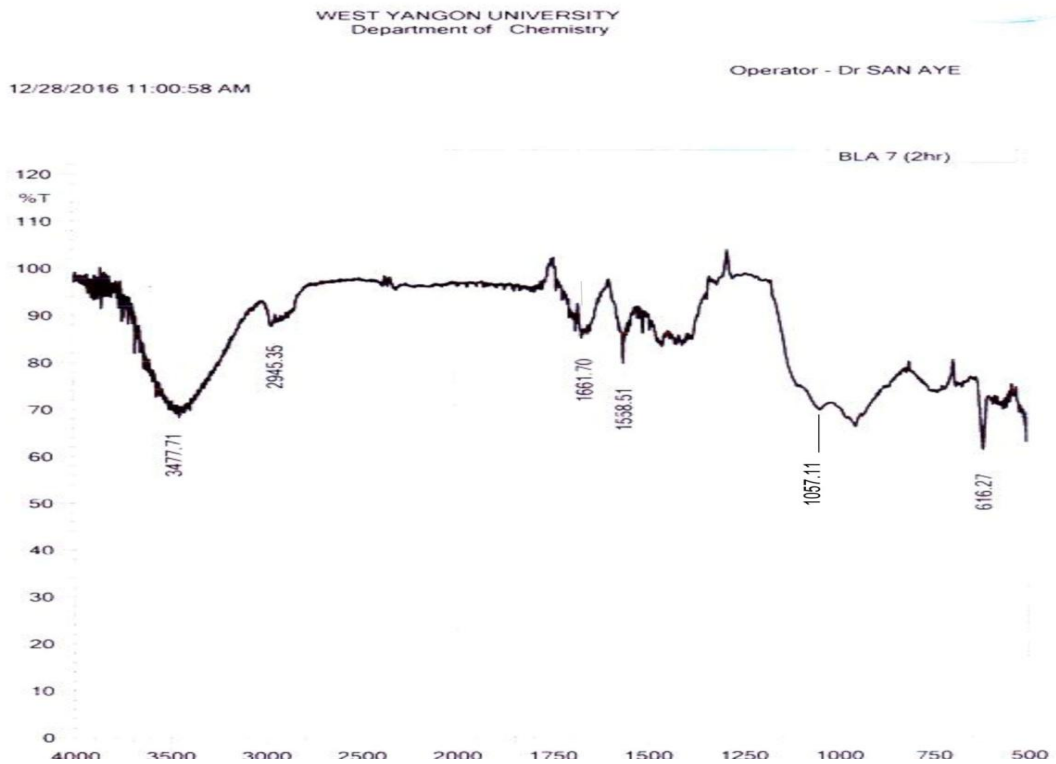


Figure 4 FT IR spectrum of BLA-7(2)

Table 2 Band Assignment for FT IR Spectrum of BLA-7(2)

Band No.	Observed wave no.(cm^{-1})	* Literature wave no.(cm^{-1})	Band Assignment
1.	3477	3700-3200	n_{Si-OH}
2.	1661	1750-1650	d_{Si-OH}
3.	1057	1200-700	$n_{Si-O-Si}$
4.	616	<650	d_{Si-O}

* Nakamoto, 1986; Amutha *et al.*, 2010

Physicochemical Properties of BLA-7(2)

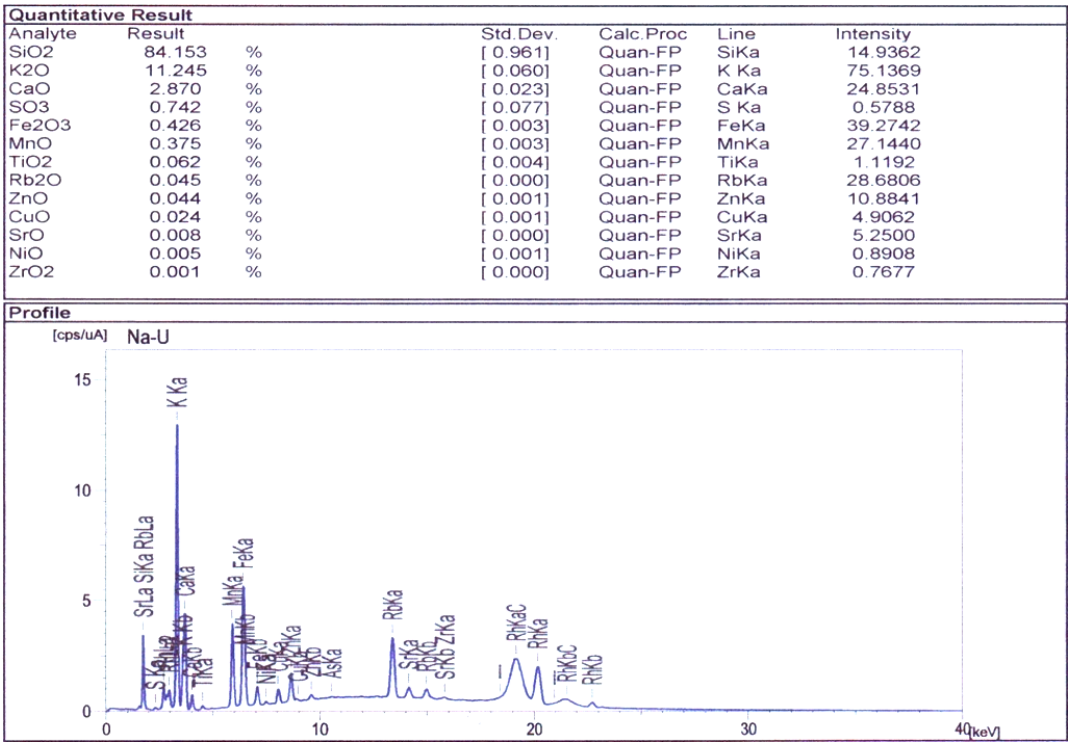
Some physicochemical properties of BLA-7(2) are relevant to moisture content, ash content, bulk density and pH. The moisture content 0.37 %, bulk density 1.44 g mL^{-1} and pH value (10.03) of BLA-7(2) sample (3.08% yield) were obtained. So this sample was found to be in base nature.

Characterization of Nanosilica Xerogel Powder

Nanosilica xerogel powder was prepared from BLA-7(2) by dissolution and precipitation method. The prepared xerogel powder was characterized by EDXRF, XRD, SEM and FT IR techniques.

EDXRF analysis of nanosilica xerogel powder

The EDXRF spectrum of xerogel powder is described in Figure 5. The comparison of the relative abundance of some oxides in BLA-7(2) and nanosilica xerogel powder are shown in Table 3. In the preparation of nanosilica by calcination of bamboo leaves, the maximum silica percent was observed as 52 % in BLA-7(2) but it was promoted to 84 % in nanosilica xerogel powder. Therefore this process (xerogel formation process) gave the silica rich substance.



Quantitative Result

Analyte	Result		Std.Dev.	Calc.Proc	Line	Intensity
SiO2	84.153	%	[0.961]	Quan-FP	SiKa	14.9362
K2O	11.245	%	[0.060]	Quan-FP	K Ka	75.1369
CaO	2.870	%	[0.023]	Quan-FP	CaKa	24.8531
SO3	0.742	%	[0.077]	Quan-FP	S Ka	0.5788
Fe2O3	0.426	%	[0.003]	Quan-FP	FeKa	39.2742
MnO	0.375	%	[0.003]	Quan-FP	MnKa	27.1440
TiO2	0.062	%	[0.004]	Quan-FP	TiKa	1.1192
Rb2O	0.045	%	[0.000]	Quan-FP	RbKa	28.6806
ZnO	0.044	%	[0.001]	Quan-FP	ZnKa	10.8841
CuO	0.024	%	[0.001]	Quan-FP	CuKa	4.9062
SrO	0.008	%	[0.000]	Quan-FP	SrKa	5.2500
NiO	0.005	%	[0.001]	Quan-FP	NiKa	0.8908
ZrO2	0.001	%	[0.000]	Quan-FP	ZrKa	0.7677

Figure 5 EDXRF spectrum of silica xerogel powder

Table 3 Comparison of the Relative Abundance of some Oxides in BLA-7(2) and Silica Xerogel Powder Prepared from BLA-7(2)

Sample	Relative abundance of some oxides (%)			
	SiO_2	K_2O	P_2O_5	CaO
Nanosilica (BLA-7(2))	52.290	25.045	10.118	10.919
Nanosilica Xerogel	84.153	11.245	-	2.870

XRD analysis of nanosilica xerogel powder

Figure 6 is the XRD diffractogram of nanosilica xerogel powder. According to XRD results, nanosilica xerogel consists essentially of tridymite form of silica, which is the crystalline nature. The crystallite size of these sample was calculated by Scherrer equation and the result was observed within the range of nano scale. The average crystallite size of xerogel was found to be 30.5 nm. When the preparation of nanosilica xerogel powder, the crystallite size was larger than that of BLA-7(2) but the percent composition of nanosilica in xerogel powder was very much higher than that of BLA-7(2).

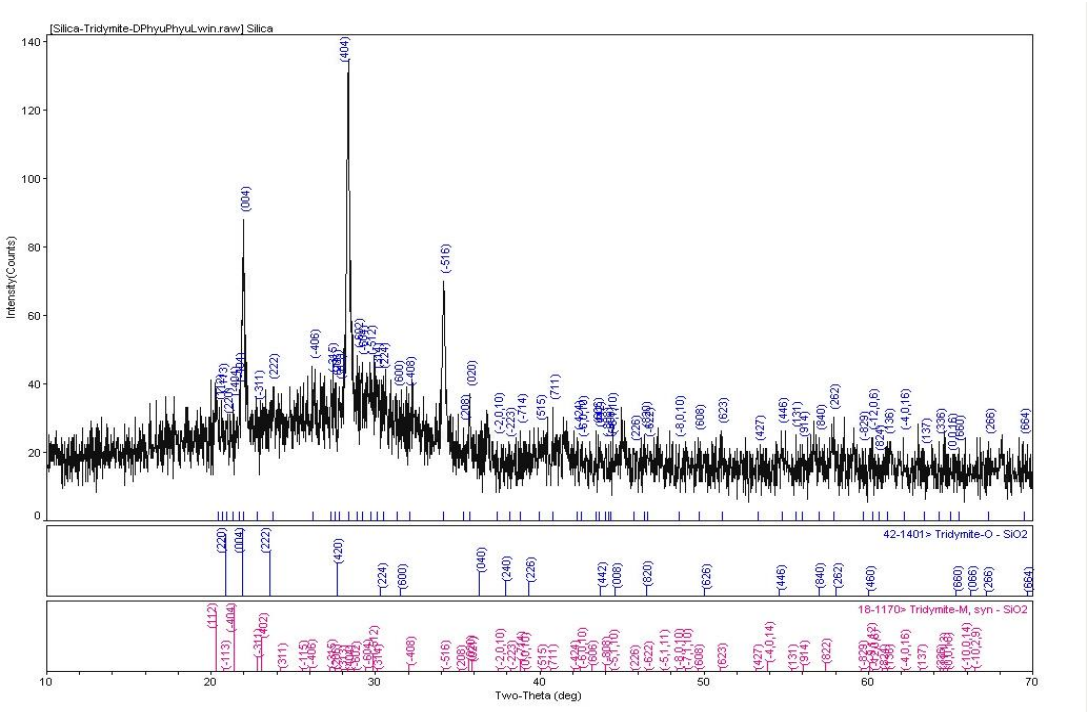


Figure 6 XRD diffractogram of silica xerogel powder

SEM analysis of nanosilica xerogel powder

The SEM micrographs of nanosilica xerogel are presented in Figures 7 (a) and (b). It indicated that xerogel powder has porous nature.

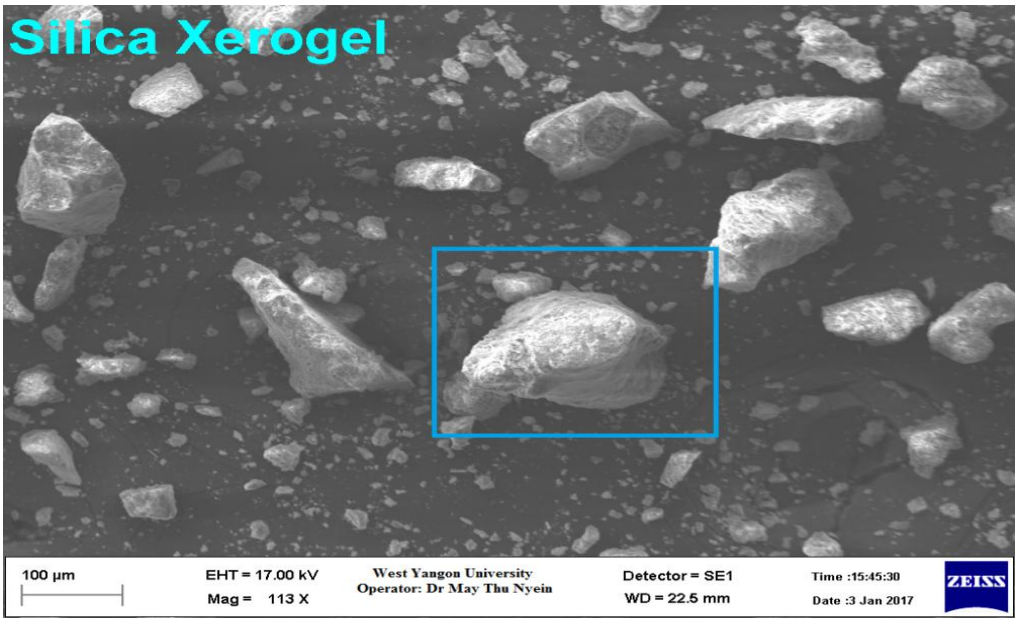


Figure 7(a) SEM micrograph of silica xerogel (Magnification 113 x)

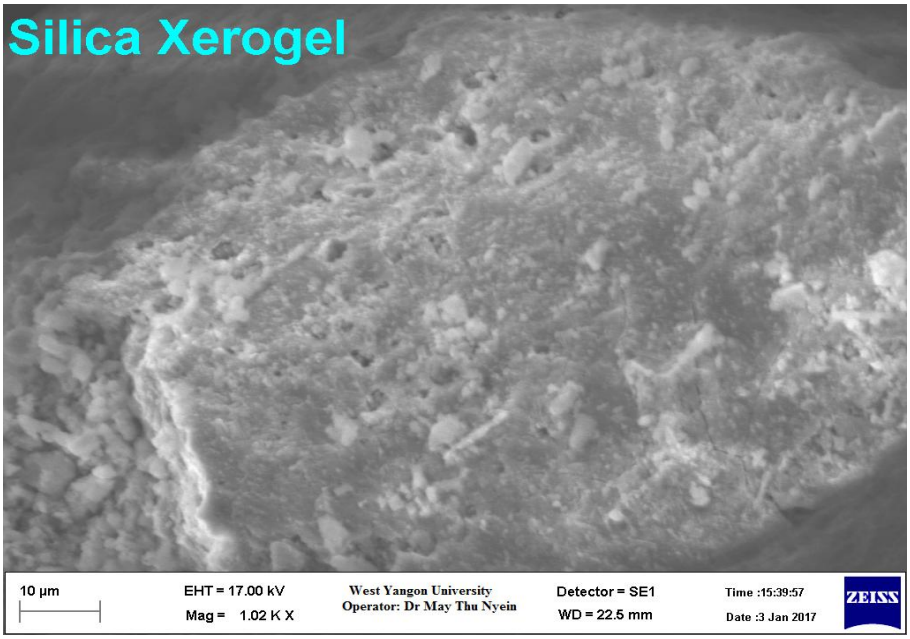


Figure 7(b) SEM micrograph of silica xerogel (Magnification 1020 x)

Table 4 Comparison of the Crystallite Size of BLA-7(2) and Silica Xerogel Powder

No.	Sample	2q (degree)	FWHM (degree)	Calculated (nm)
1	Nanosilica (BLA-7(2))	30.601	0.420	19.6
2	Nanosilica Xerogel	28.353	0.268	30.5

FT IR analysis of nanosilica xerogel powder

The characterization of prepared nanosilica xerogel was studied by FT IR spectrophotometer. Figure 8 and Table 5 show the FT IR spectrum and its band assignment of nanosilica xerogel. The possible assignment for the band at 3357 cm^{-1} is stretching vibration of OH bond in Si-OH group. The band at 1631 cm^{-1} indicates the bending vibration of OH bond in Si-OH group. The band at 1047 cm^{-1} is stretching vibration of Si-O asymmetric bond in Si-O-Si group.

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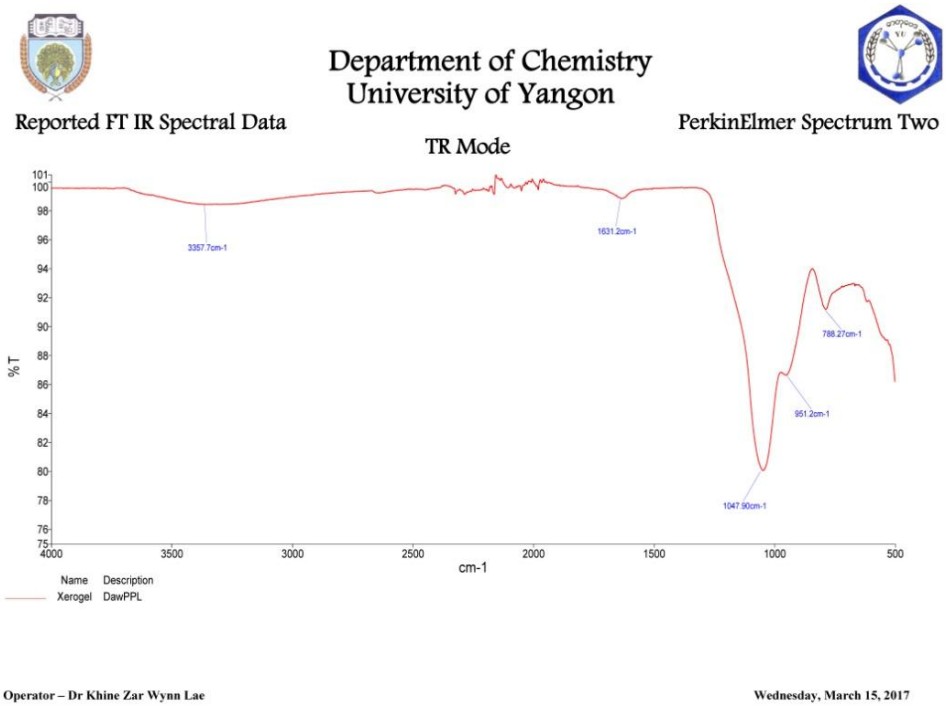

Figure 8 FT IR spectrum of silica xerogel

Table 5 Band Assignment for FTIR Spectrum of Nanosilica Xerogel

Band No.	Observed Wave No. (cm^{-1})	*Literature Wave No. (cm^{-1})	Band Assignment
1.	3357	3700-3200	n_{Si-OH}
2.	1631	1750-1600	d_{Si-OH}
3.	1047	1200-1000	$n_{Si-O-Si}$
4.	951	1200-700	n_{Si-OH}
5.	788		$n_{Si-O-Si}$

*****Nakamoto, 1986; Amutha *et al.*, 2010

Physicomechanical Analysis of Cement Containing 5 % BLA-7 (2) (5 %BLA Cement)

The quality of 5 % BLA cement (soundness, normal consistency, setting time, compressive strength and tensile strength) were improved when mixing with the selected ash sample.

Table 6 Physicomechanical Properties of Cement (Alpha) and BLA Cement

Sample	Normal Consistency (%)	Setting Time (min)		Soundness (mm)	Compressive strength (MPa)		Tensile strength (psi)	
		Initial	Final		7 days	28 days	7 days	28 days
Cement (Alpha)	28.5	150	256	1.6	11.3	15.7	230	243.3
BLA Cement	30.8	171	234	1.0	15.6	16.7	230	270

Conclusion

In this research, an attempt was made to produce nanosilica from *Dendrocalamas giganteus* Munro (wa-bo-gyi) bamboo leaves ash. The formation of nanosilica in bamboo leaves ash depends not only on the calcination temperature but also calcination time. At low calcination temperature, the colour of ashes was observed dark-grey; depending upon the proportion of unburnt carbon present. The ash obtained at 800 °C was almost grey colour. The bamboo leaves was calcined at 1000 °C, the ash was observed white.

This study indicated that the maximum relative abundance of SiO_2 can be produced by maintaining the calcinations temperature at 1000 °C under calcination time for 2 h (BLA-7(2)). The yield percent of this sample was 3.08 %.

BLA-7(2) was characterized by EDXRF, XRD, SEM and FT IR techniques. From EDXRF result, the maximum relative abundance of silica was found to be 52.29 % and small amount of other trace oxides. XRD analysis of this sample was observed crystalline nature and crystallite size was 19.6 nm. From the SEM analysis it was found that this sample has porous nature. FT IR analysis showed the presence of Si-OH, Si-O-Si and Si-O bond.

The production of high purity silica from BLA-7(2) was studied by dissolution and precipitation method. From EDXRF analysis, the relative abundance of silica was distinctly increased from 52.29 to 84.15 % and other constituent oxides were decreased. From the XRD analysis, the silica xerogel powder was found to be crystalline nature and tridymite phase from the ICDD search/match process. Its crystallilte size was 30.5 nm. From the SEM analysis, silica xerogel powder has porous nature. In the FT IR data of silica xerogel sample, the presence of Si-OH and Si-O-Si groups were found in the prepared silica xerogel.

The physical and mechanical properties of 5 % BLA cement and alpha cement such as normal consistency, setting time, compressive strength and tensile strength were also determined. It was found that the normal consistency of 5 % BLA cement was higher than alpha cement. The increase in normal consistency was observed that it needed larger amount of water than the alpha cement. This is because of 5 % BLA mixture with the cement presents more porous nature and hence tends to absorb more water.

The setting time of 5 % BLA cement and alpha cement were compared. It can be observed that the addition of 5 % BLA in the cement speeds up setting time. The soundness test

result of 5 % BLA mixing with cement is 1 mm, which is well within the limiting value of 10 mm specified by BS12 (British Standard).

The compressive strength of 5 % BLA blended cement was increased from 11.3 MPa to 15.6 MPa after 7 days, and from 15.7 MPa to 16.7 MPa after 28 days. The tensile strengths of BLA blended cement and alpha cement were found to be the same value after 7 days. But they were found to be increased from 243.3 psi to 270.0 psi after 28 days.

From the experimental results of compressive strength and tensile strength, it can be inferred that addition of 5 % BLA enhance the quality of cement.

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