Improvement of Yield and Phase Purity on Hydrothermal Synthesis Smectite using Taguchi Method

***Yi-Yu Liu, Bing-Sheng Yu**
National Taipei University of Technology, Taiwan
Institute of Mineral Resources Engineering

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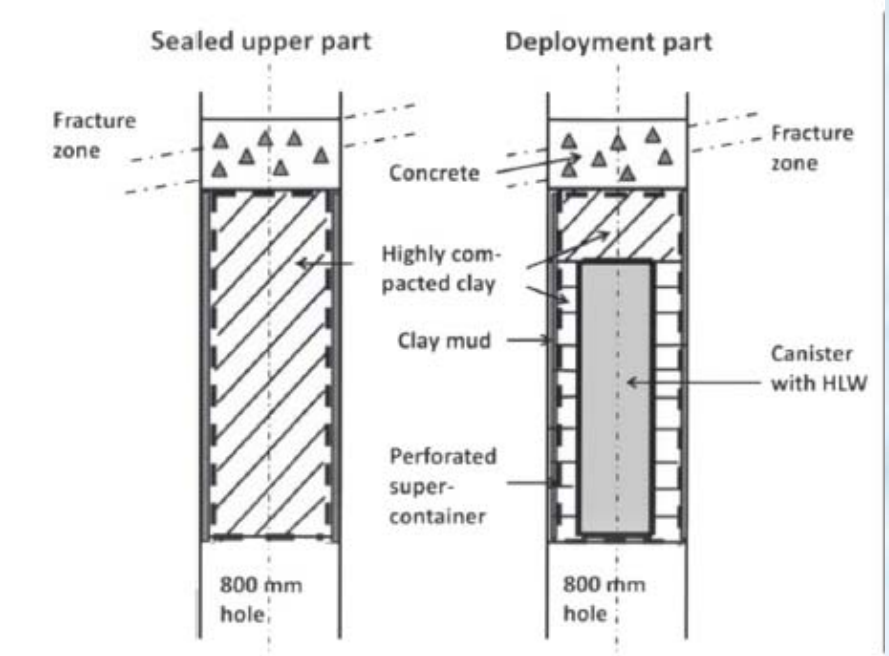
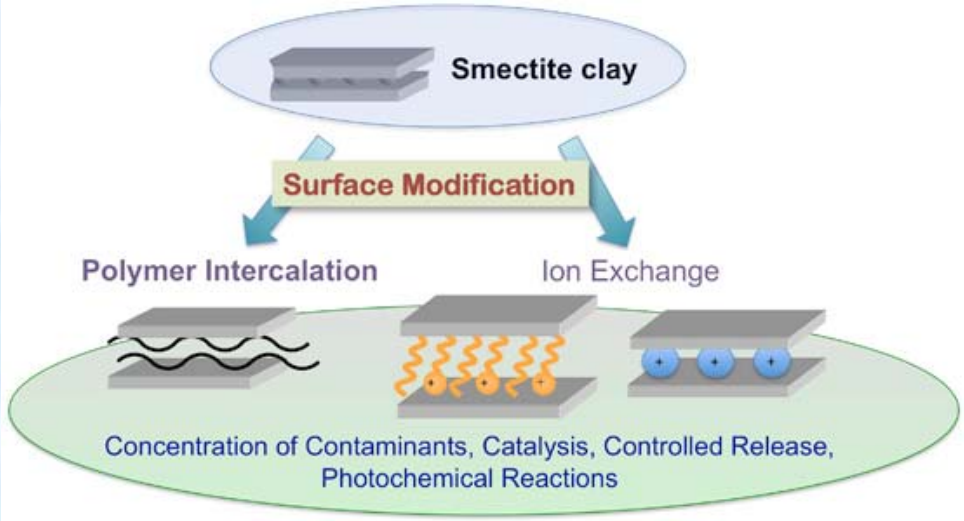
Introduction

- The smectite mineral is widely used in many field of industry, material application and waste barrier.



Pusch et al., 2015



Okada et al., 2014

Introduction

- The pursuit of **phase purity** and **high yield** on synthetic smectite is getting more and more important.

- In recent years, many works have been research about obtain different kinds of synthetic smectite.

- But only a few works discussed the phase purity and percentage of their final products.

Research Purpose

- Establish a quick and easy process of quantitative phase analysis to estimate the phase purity and yield of the synthetic smectite product.

- Optimize the phase purity and yield of synthetic smectite, the contribution between experimental factor and result was calculated by Taguchi method.

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Experimental factors will affect the properties of synthetic smectite products

Si source	SiO_3，TEOS，Na_2SiO_3
Al source	$Al(NO_3)_3$，$Al(OH)(CH_3COO)_2$，$Al(OH)_3$
Mg source	$Mg(NO_3)_2 \cdot 6H_2O$，$Mg(CH_3COO)_2 \cdot 4H_2O$，$Mg(OH)_2$
Temperature (℃)	180，200，220
Reaction time (hours)	48，72，96
Initial pH	7，9，11
Mineralizer	HF，NaF，NH_4F
Mineralizer amount (ml)	0.02，0.025，0.03
Mg : Al	7：3，8：2，9：1

It needs 19683(3^9) sets of experiment for all the combination of factor in the table.

Taguchi method

- Use the **S/N ratio** and **Average** value of data to analyze the contribution and effect between the factor and result.
- Obtain the best and optimized experimental factor.
- Increase the phase purity and yield of the synthetic smectite.

Save more experimental resources and time!!

Orthogonal array

Noise experiments

3 noise factors **Outer array**

No	1	2	3	4
α	1	1	2	2
β	1	2	1	2
γ	1	2	2	1

Main experiments

Inner array **8 main factors**

No	A	B	C	D	E	F	G	H	DATA1	DATA2	DATA3	DATA4
1	1	1	1	1	1	1	1	1				
2	1	1	2	2	2	2	2	2				
3	1	1	3	3	3	3	3	3				
4	1	2	1	1	2	2	3	3				
5	1	2	2	2	3	3	1	1				
6	1	2	3	3	1	1	2	2				
7	1	3	1	2	1	3	2	3				
8	1	3	2	3	2	1	3	1				
9	1	3	3	1	3	2	1	2				
10	2	1	1	3	3	2	2	1				
11	2	1	2	1	1	3	3	2				
12	2	1	3	2	2	1	1	3				
13	2	2	1	2	3	1	3	2				
14	2	2	2	3	1	2	1	3				
15	2	2	3	1	2	3	2	1				
16	2	3	1	3	2	3	1	2				
17	2	3	2	1	3	1	2	3				
18	2	3	3	2	1	2	3	1				

It takes only 72 experiments

The main factors and levels of experiments.

No.	Factor	Level 1	Level 2	Level 3
A	Si source	Na_2SiO_3	TEOS	
B	Mineralizer amount (ml)	0.02	0.025	0.03
C	Reaction time (hours)	48	72	96
D	Initial pH	7	9	11
E	Mineralizer	HF	NaF	NH_4F
F	Na excess (%)	100	110	120
G	Al source	$Al(NO_3)_3$	$Al(OH)(CH_3COO)_2$	$Al(OH)_3$
H	Mg source	$Mg(NO_3)_2 \cdot 6H_2O$	$Mg(CH_3COO)_2 \cdot 4H_2O$	$Mg(OH)_2$

The noise factors and levels of experiments.

No.	Factor	Level 1	Level 2
α	Temperature	200℃	220℃
β	Alkali solution	NH_4OH	NaOH
γ	Mg : Al	9：1	8：2

The noise factor is the factor that we can't change much.

The calculation of phase purity & yield

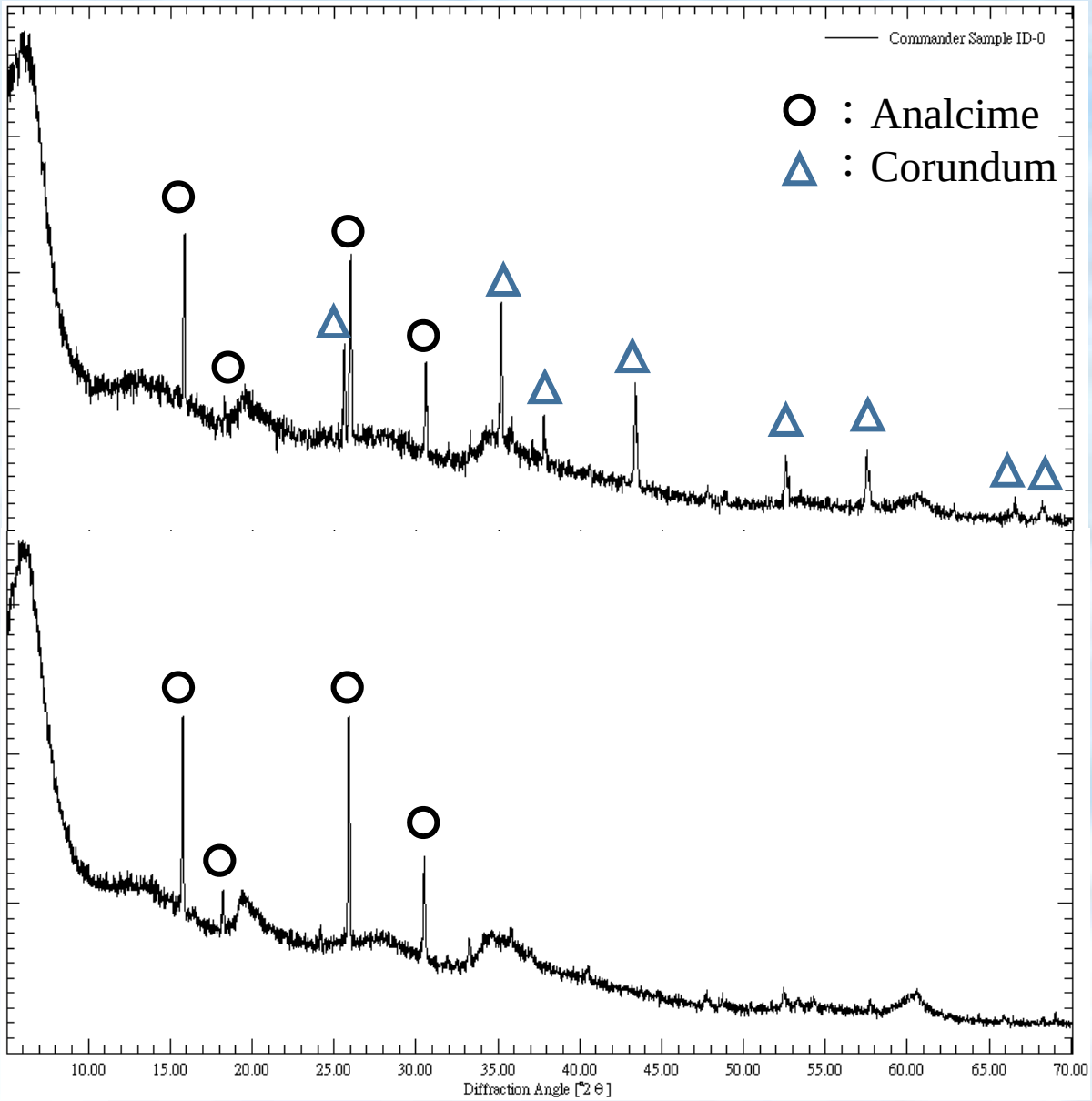
- The QPA of the synthetic clay sample is using Rietveld method software.

PROFEX (Doebelin and Kleeberg, 2015)

+

BGMN software (Bergmann et al., 1998)

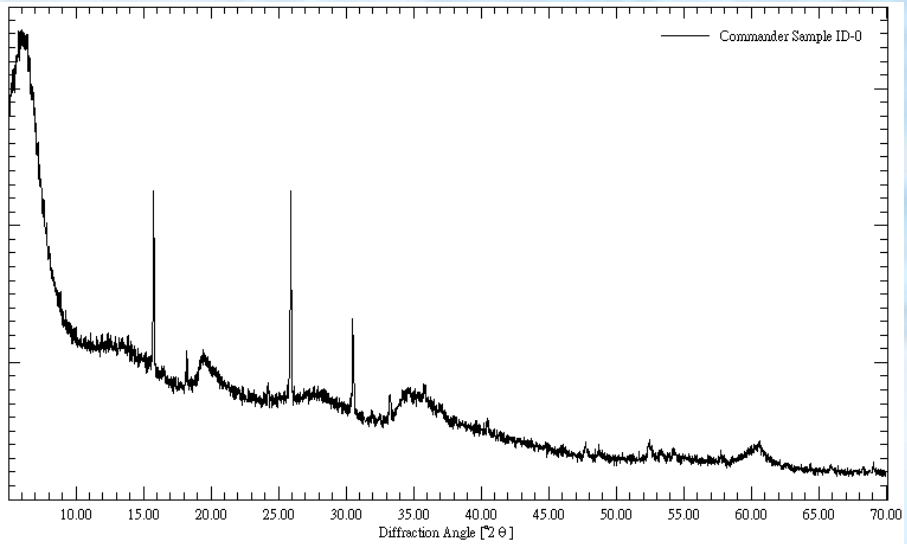
Smectite sample + Internal standard



Synthetic products
+
10% Corundum

Synthetic products

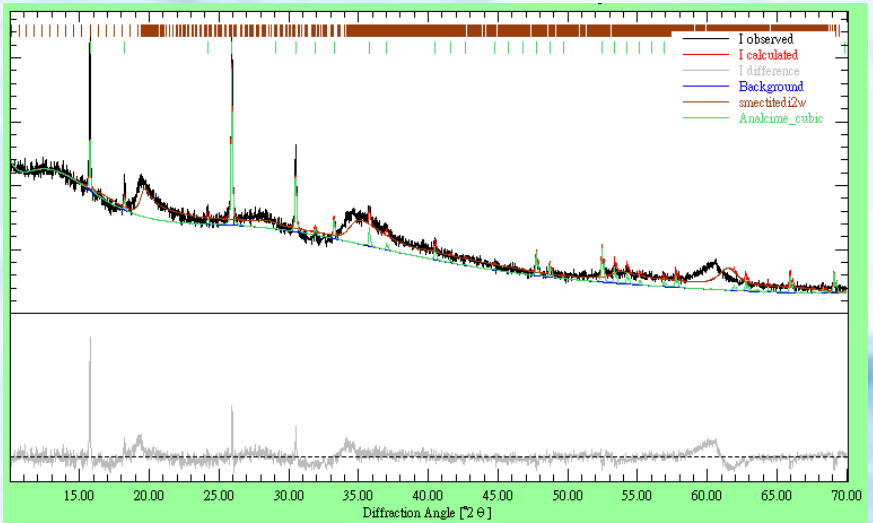
QPA without internal standard



Input XRD file

Select Mineral Phases

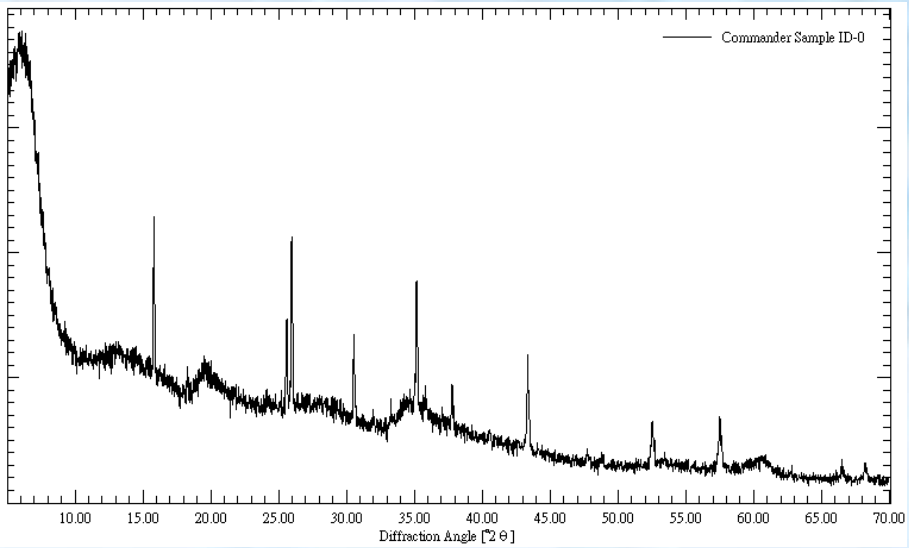
START!



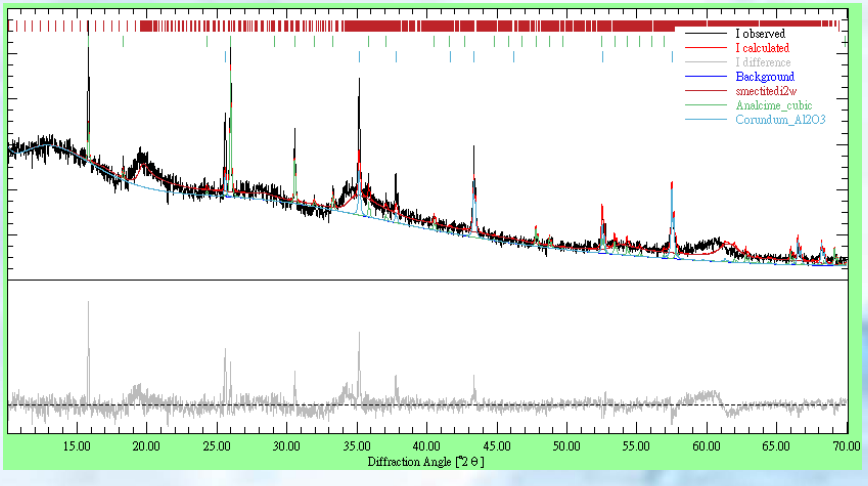
Smectite： 83.9%
Analcime： 16.1%

Amorphous?

QPA with internal standard



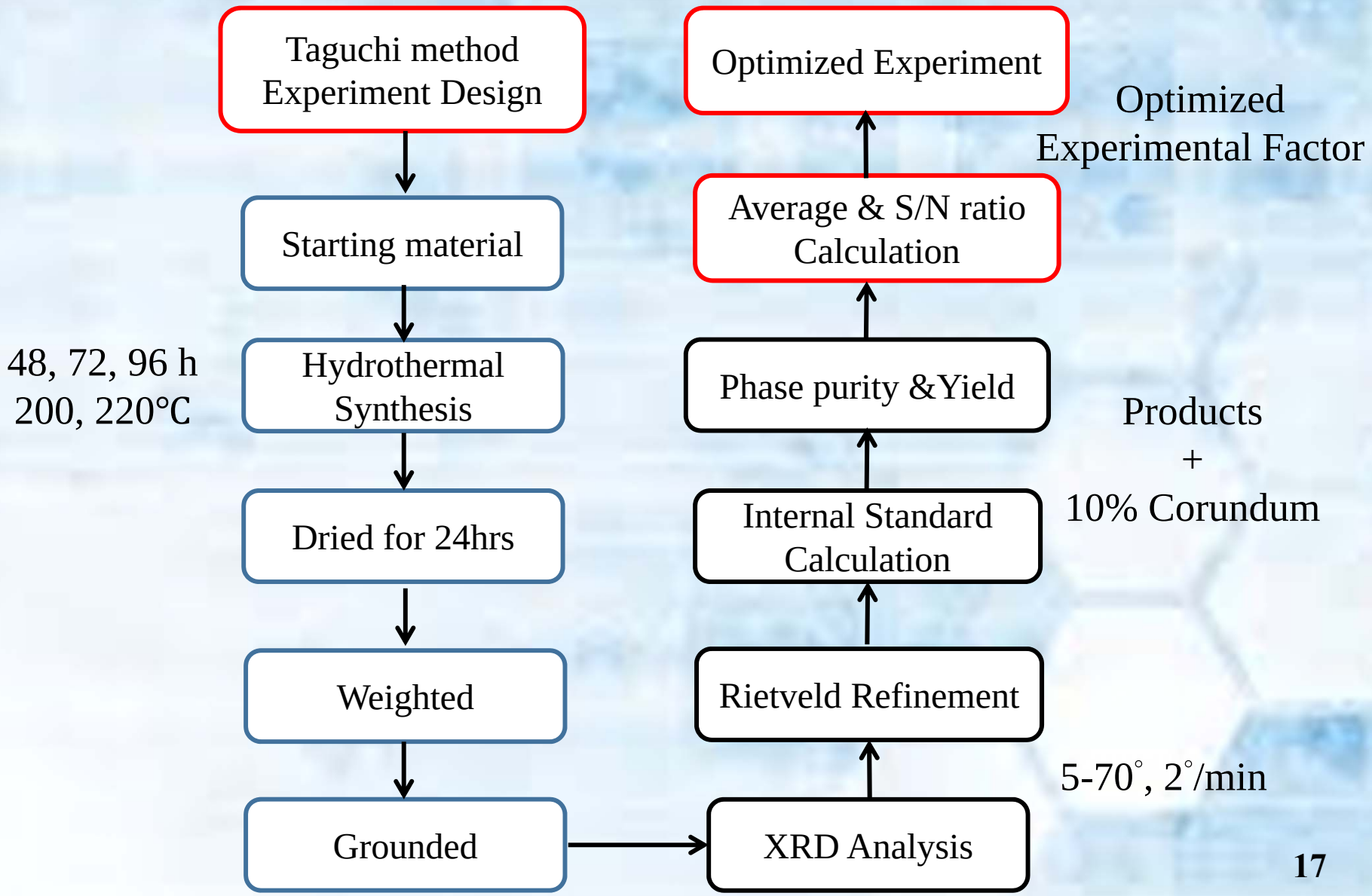
Synthetic product
+
10% Corundum
(Internal standard)



Smectite：75.9%
Analcime：13%
Corundum：
11.1%

Smectite：76.3%
Analcime：13.2%
Amorphous：10.5%

Experimental flow chart



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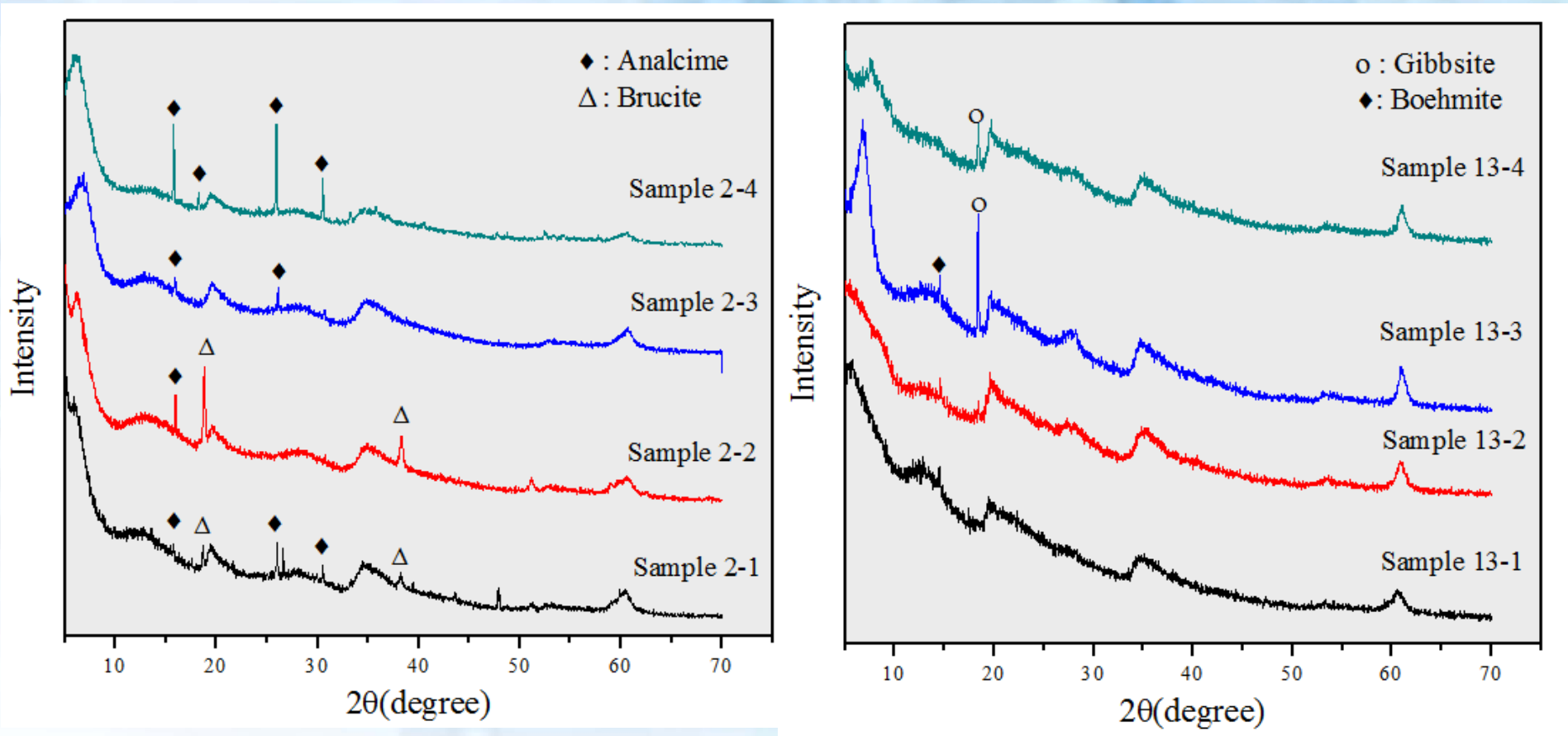
XRD analysis of smectite products

72 smectite samples were all analyzed by XRD.



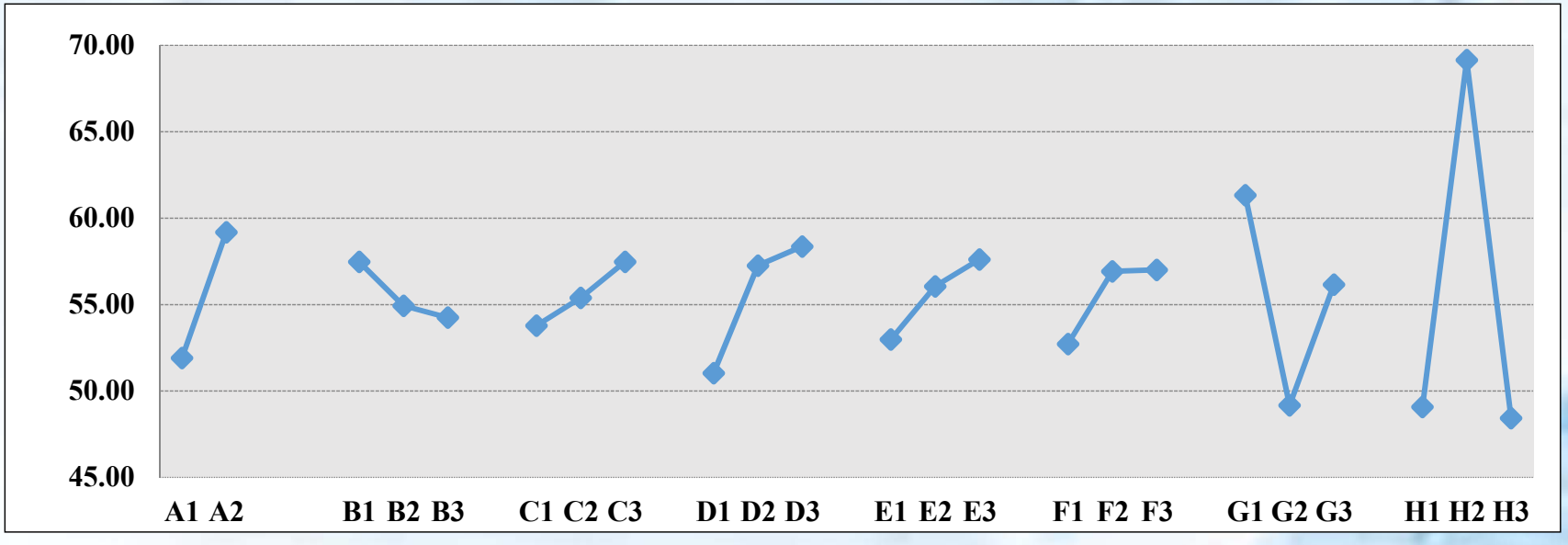
Other crystal phase and peaks are all smectite.

The phase purity of 72 products

Phase purity (%)	Data1	Data2	Data3	Data4	AVG	S/N
1	19.9	35.1	59.1	49.6	40.93	29.18
2	66.4	48	79.4	62	63.95	35.64
3	42	43.2	57.4	77.6	55.05	33.29
4	22.5	44.2	46.9	49.5	40.78	30.09
5	65	55.9	62.1	41.3	56.08	35.65
6	72.9	46.4	43.9	69.8	58.25	34.08
7	30.2	45.4	39.6	29	36.05	31.31
8	49.7	40.2	62.1	28.7	45.18	33.69
9	78.6	95.7	65.7	43.8	**70.95**	37.76
10	47.6	51.4	66.4	47.6	53.25	34.57
11	69.3	34.5	89	87.4	70.05	34.07
12	49.5	61.9	81.7	53.3	61.60	35.64
13	77.3	68.3	70.3	70.2	**71.53**	37.11
14	83.2	56.5	51.7	41.7	58.28	35.57
15	45.5	49.6	61.3	22.3	44.68	34.14
16	74.3	86.7	91.1	68.5	**80.15**	38.39
17	32.7	24.4	54.7	43.5	38.83	30.07
18	60.7	55.8	63.2	37.6	54.33	35.51
AVG					**55.6**	**34**

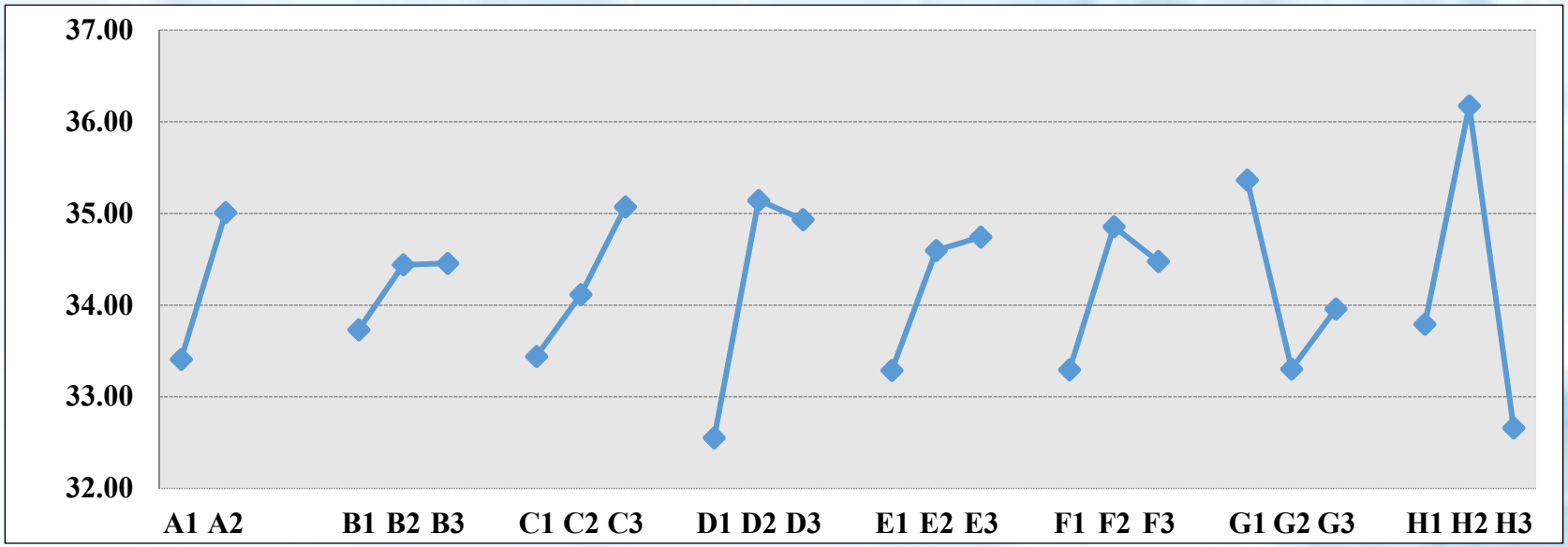
The AVG(%) response table of phase purity

Phase purity AVG(%)								
	A	B	C	D	E	F	G	H
Level 1	51.91	**57.47**	53.78	51.03	52.98	52.72	**61.33**	49.07
Level 2	**59.19**	54.93	55.39	57.25	56.05	56.92	49.17	**69.15**
Level 3		54.25	**57.48**	**58.36**	**57.61**	**57.01**	56.15	48.43



The S/N ratio(%) response table of phase purity

Phase purity S/N ratio(%)								
	A	B	C	D	E	F	G	H
Level 1	33.41	33.73	33.44	32.55	33.29	33.30	**35.37**	33.79
Level 2	**35.01**	34.44	34.12	**35.14**	34.60	**34.86**	33.30	**36.17**
Level 3		**34.46**	**35.07**	34.93	**34.74**	34.48	33.96	32.66

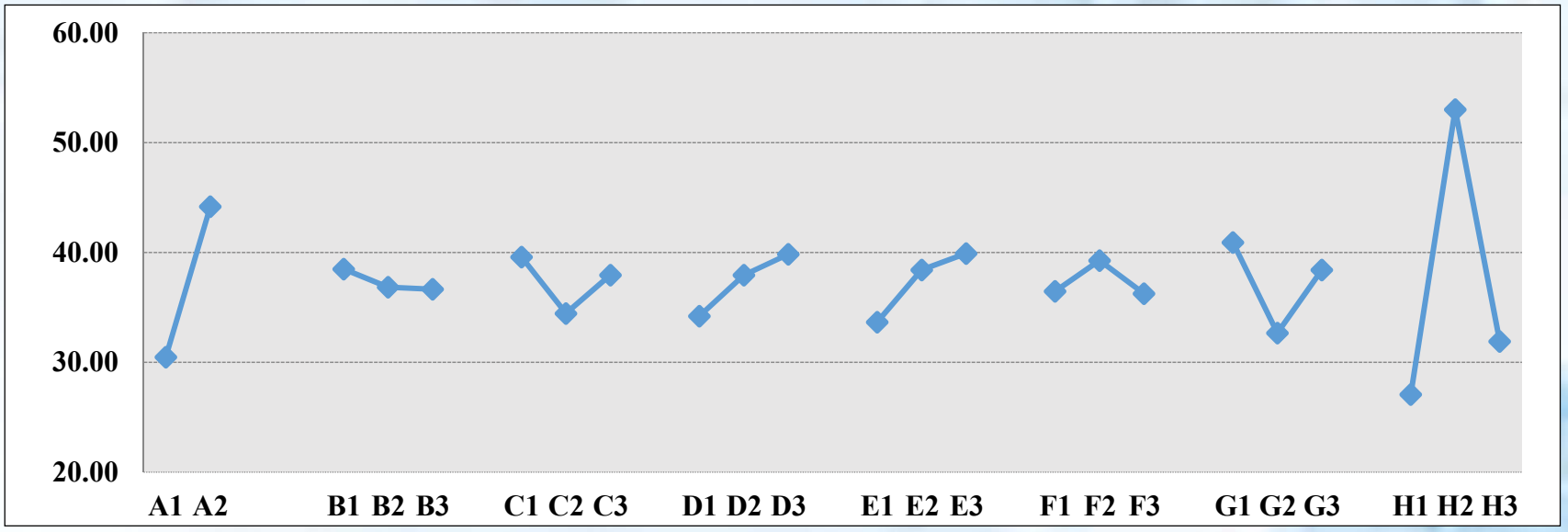


The yield of 72 products

Yield (%)	Data1	Data2	Data3	Data4	AVG	S/N
1	12.9	23.4	22.5	19.4	19.55	24.85
2	39.5	30.5	61.3	42.2	43.38	31.80
3	17.2	17.6	27.9	64.9	31.90	25.80
4	14.4	29.6	31.5	30.9	26.60	26.34
5	23.4	17.2	26	18.8	21.35	26.52
6	44.9	31	22.5	55.2	38.40	29.32
7	17.8	14.5	25.2	18.8	19.08	25.00
8	23	22.4	24.7	16.8	21.73	27.35
9	49.7	67	63.8	28.8	**52.33**	35.36
10	38.1	42.3	43.1	35.5	39.75	32.25
11	50.2	23.4	72.9	59.1	51.40	30.95
12	30.3	46.8	67.6	35.2	44.98	32.30
13	71.2	61.1	66.1	66.6	**66.25**	36.36
14	63.6	38.8	35.1	26.3	40.95	32.41
15	26.5	32.8	35.5	15.1	27.48	29.79
16	61.5	69.9	82.4	51.4	**66.30**	36.87
17	23.9	16	45	26.7	27.90	26.88
18	37.4	36.5	38.2	18.3	32.60	31.45
AVG					**37.3**	**30**

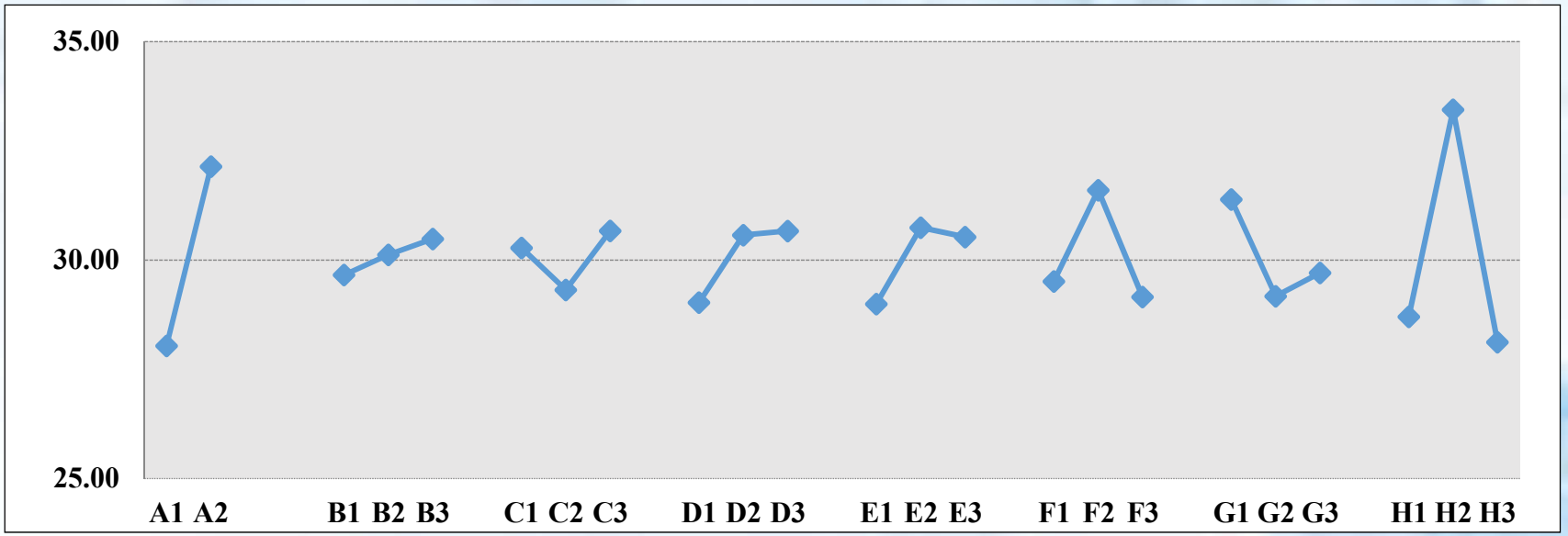
The AVG(%) response table of yield

Yield AVG(%)								
	A	B	C	D	E	F	G	H
Level 1	30.48	**38.49**	**39.59**	34.21	33.66	36.47	**40.91**	27.08
Level 2	**44.18**	36.84	34.45	37.94	38.41	**39.27**	32.66	**53.01**
Level 3		36.65	37.95	**39.84**	**39.91**	36.25	38.41	31.90



The S/N ratio(%) response table of yield

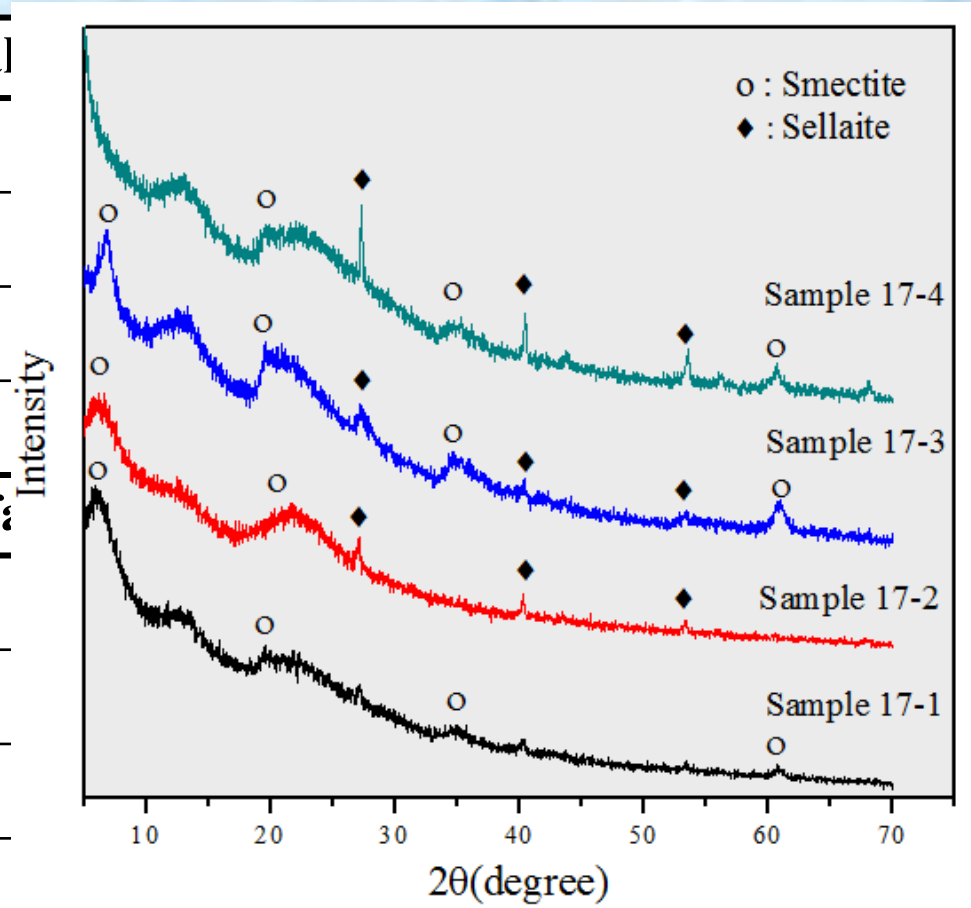
Yield S/N ratio(%)								
	A	B	C	D	E	F	G	H
Level 1	28.04	29.66	30.28	29.03	29.00	29.51	**31.39**	28.70
Level 2	**32.14**	30.12	29.32	30.57	**30.74**	**31.60**	29.17	**33.44**
Level 3		**30.48**	**30.67**	**30.67**	30.53	29.16	29.71	28.12



The optimal factor of AVG & S/N ratio

The optimal		
Factor	A	B
Phase purity	2	1
Yield	2	1
Optimal	**2**	**1**

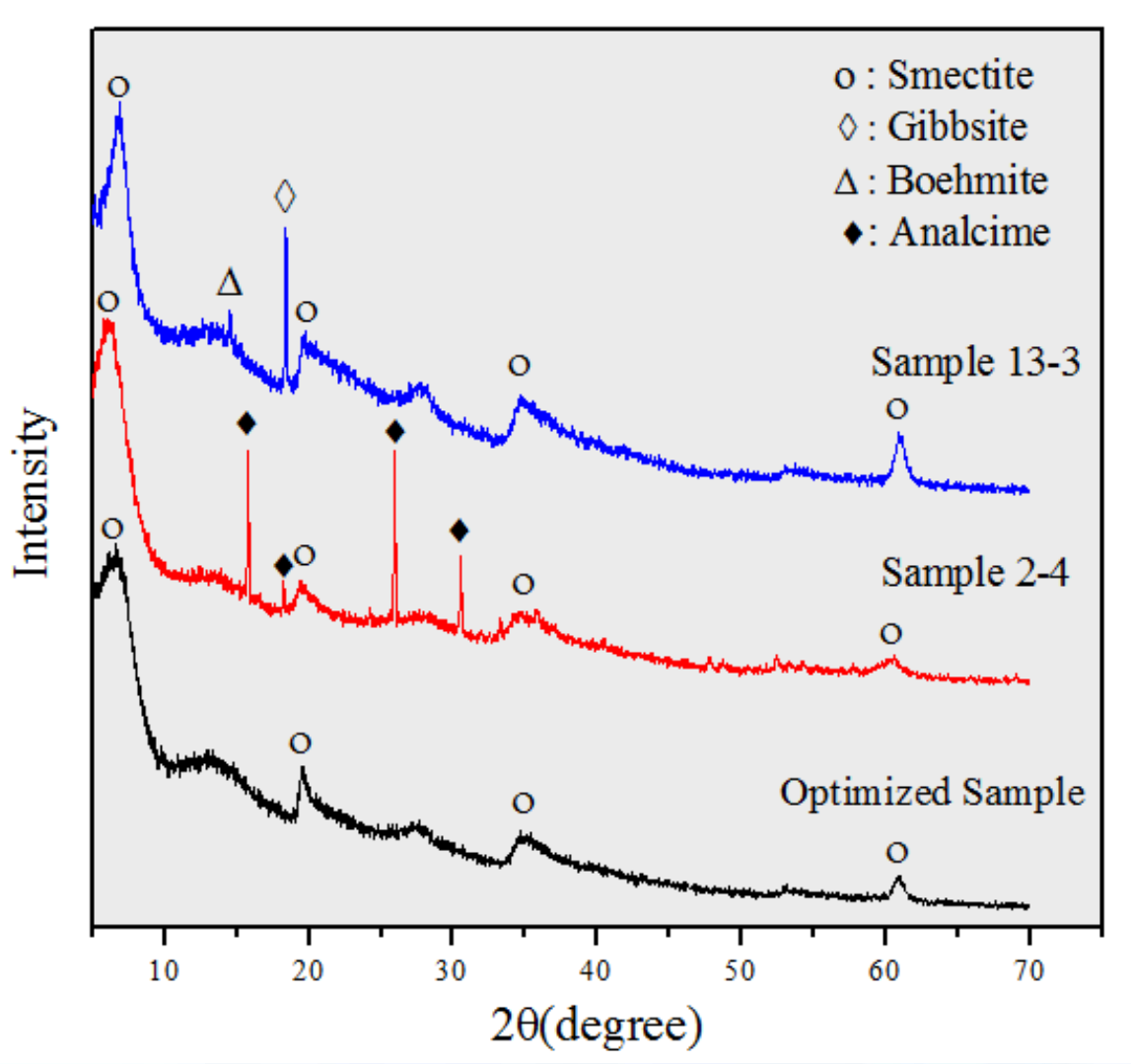
The optimal fa		
Factor	A	B
Phase purity	2	3
Yield	2	3
Optimal	**2**	**3**



The optimal factor level of AVG & S/N ratio								
Optimal	**2**	**1**	**3**	**3**	**3**	**2**	**1**	**2**

The optimized & previous sample

After the optimization of Taguchi method, the properties of synthetic product was improved.



96h, 220℃, pH=11, Mg : Al = 9 : 1

The phase purity & yield of optimized sample

Content	Sample Average (%)	Optimized Sample (%)
Smectite	**55.6**	**79.6**
Impurity	9.7	0
Amorphous	34.7	20.4
Yield	**37.3**	**81.6**

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- The synthesis process and products quality was improved by Taguchi method, and it can decide the most efficient way to synthesize the smectite product.

- Profex combine with software BGMN was chosen for QPA of the synthetic smectite sample. **The fast and easy analysis process that can provide a useful method to execute quantitative phase analysis not only for the natural clay mineral but also the synthetic clay sample**.

Conclusion

- Compare to the previous products. The yield and phase purity of the optimized synthetic products can reach up to **81.6** and **79.6 %** respectively. The phase purity and yield is much higher than the previous products.

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Thanks for your listening!!
Have a nice day!!