Supplementary Documentation

Highly Reactive Energetic Films by Pre-Stressing Nano-Aluminum Particles

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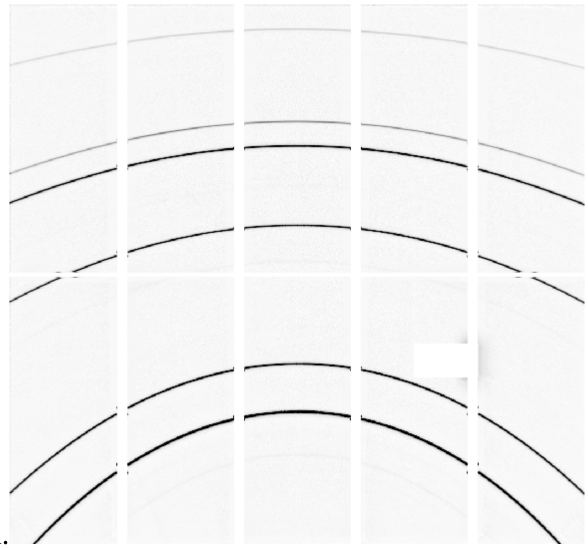
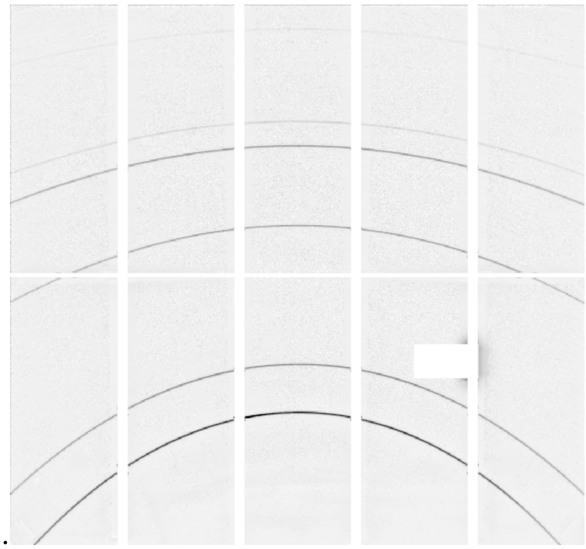
S1. Photograph showing films

A.


B.


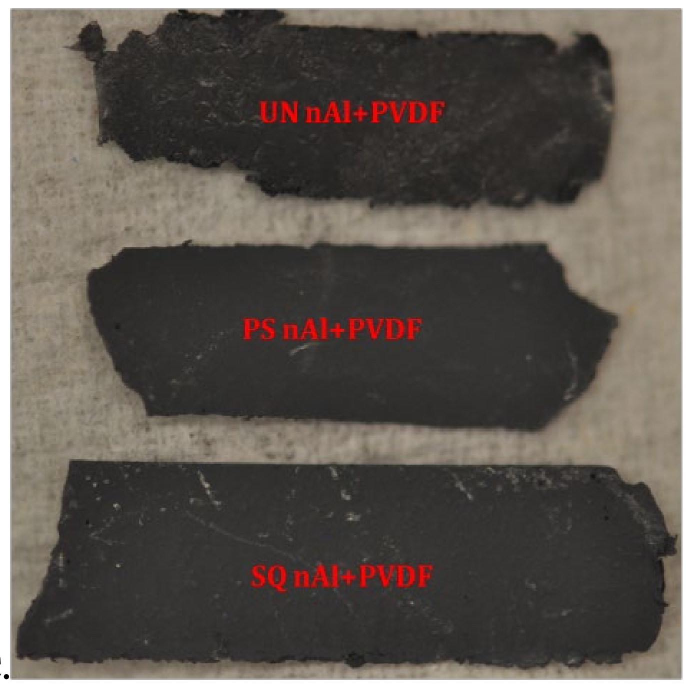
C.


Figure S1. A. Powder Synchrotron X-ray Diffraction Pattern of 80 nm Al powder pre-stressed (PS). B. Powder Synchrotron X-ray Diffraction Pattern of 80 nm Al powder super-quenched (SQ). C. Films photographed are labeled and approximately 2 cm in length when prepared. Films are cut to fit in the circular sample holder in the pressure cell at a constant mass for all samples. Further characterization includes PXRD analysis (Section S2).

S2. Powder X-ray Diffraction (PXRD) data.

In the pre-combustion films (Section S2.1), all samples contain trace amounts of Calcite ($Ca(CO_3)$) from the PXRD sample holder used in the analysis. Also, trace amounts of aluminum nitride (AlN) are detected in all films indicating some reaction between the Al particles and DMF from the solvent during film preparation.

The amorphous content in all films is attributed to poorly crystalline PVDF with peaks that match when comparing pure PVDF to the film sample in Section S2.2. Therefore, the final films contain 72-74 wt. % Al with 20-24 wt. % PVDF and the remaining composition includes trace amounts of AlN and $Ca(CO_3)$. This result implies that some PVDF dissolved in the acetone and DMF solution evaporates during film drying because the precursor included a 50:50 wt. % Al:PVDF concentration.

In the post-combustion residue (Section S2.3), some cross contamination from residue of previous experiments using this device (bismuth oxide) and also from the alloy used in making the pressure cell may contribute to detection of species that are not included in the reactant films: such as Na, K, and P. However, these contaminants effect all samples.

Whole Pattern Fitting and Rietveld Refinement

FILE: [UN nAl_PVDF.raw] UN nAl_PVDF - Pantoya
SCAN: 5.0/90.0/0.02/2.4(sec), Cu(30kV,15mA), I(p)=190131, 05/23/19 02:19a
PROC: [WPF Control File]

- ☑ K-alpha2 Peak Present
- ☑ Allow Negative Isotropic B
- ☑ Allow Negative Occupancy
- ☑ Apply Anomalous Scattering

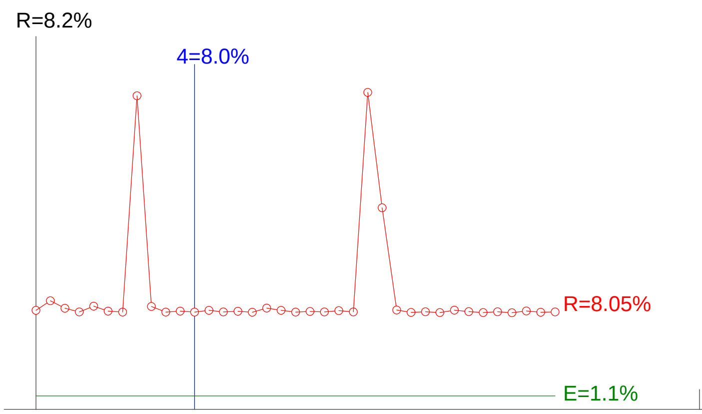
[Diffractometer LP] Two-Theta Range of Fit = 5.0 - 90.0(deg)
- ☑ Zero Offset of Goniometer - 2Theta = -0.467161(0.589593)
- ☑ Specimen Displacement - Cos(Theta) = 0.442423(0.552012)
- ☐ Monochromator Correction for LP Factor = 1.0
- ☑ K-alpha2/K-alpha1 Intensity Ratio = 0.487352(0.042854)

Profile Shape Function (PSF) for All Phases: pseudo-Voigt, Polynomial(4), Lambda=1.54059Å (Cu/K-alpha1)

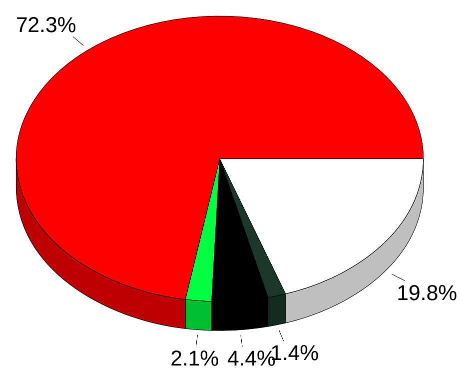
Phase ID (4)	Source	I/Ic	Wt%	#L	PC
Aluminum - Al	PDF#01-071-3760	4.10(5%)	72.3 (5.2)	5	(111)=1.000
Aluminum Nitride - AlN	PDF#04-014-9652	3.66(0%)	2.1 (0.2)	10	<None>
Calcite - $Ca(CO_3)$	PDF#01-083-0577	3.21(5%)	4.4 (0.3)	33	(104)=1.052
Aluminum Nitride - AlN	PDF#01-076-0702	1.48(5%)	1.4 (0.2)	12	<None>
Others + Amorphous			19.8 (0.9)		

XRF(Wt%): Ca=1.8%, Si=9.2%, Al=74.6%, O=12.7%, N=1.2%, C=0.5%

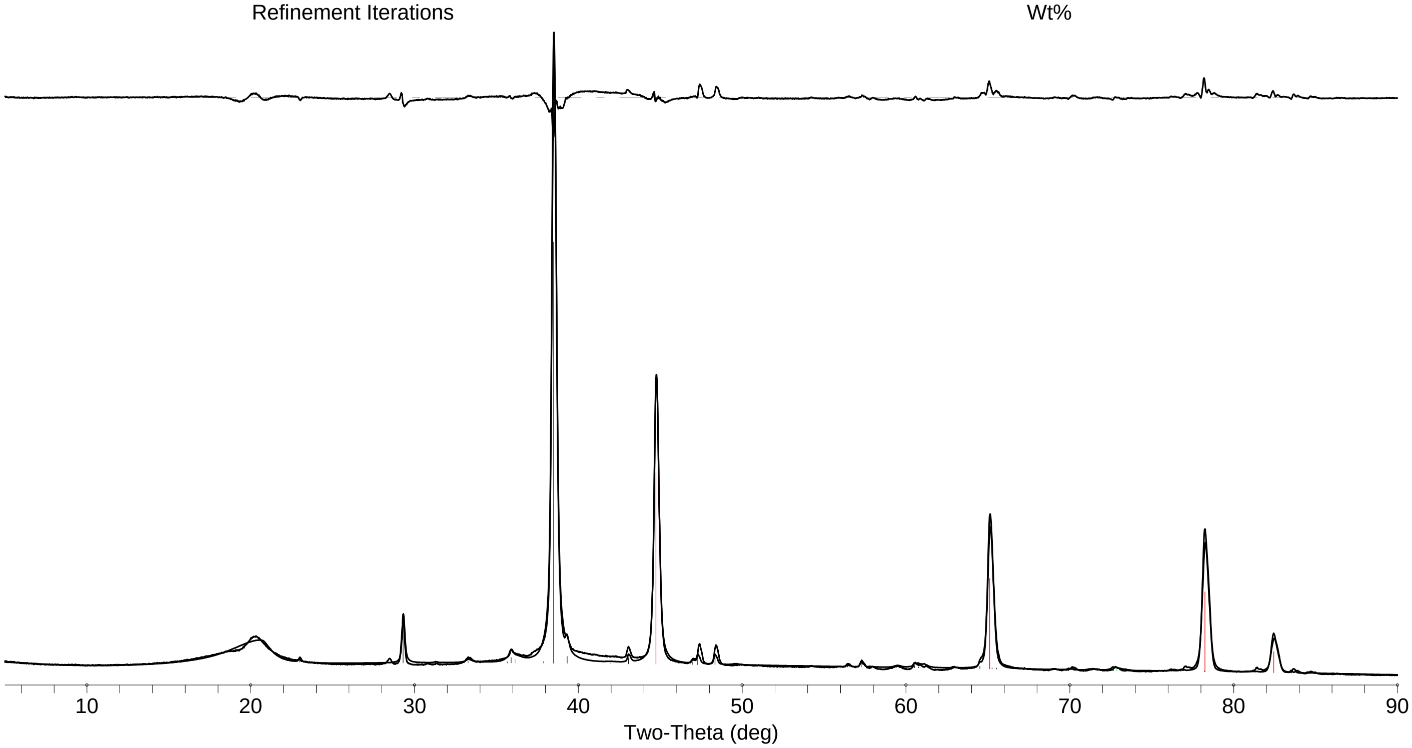
NOTE: Fitting Halted at Iteration 37(4): R=8.05% (E=1.1%, R/E=7.33, P=39, EPS=0.5)

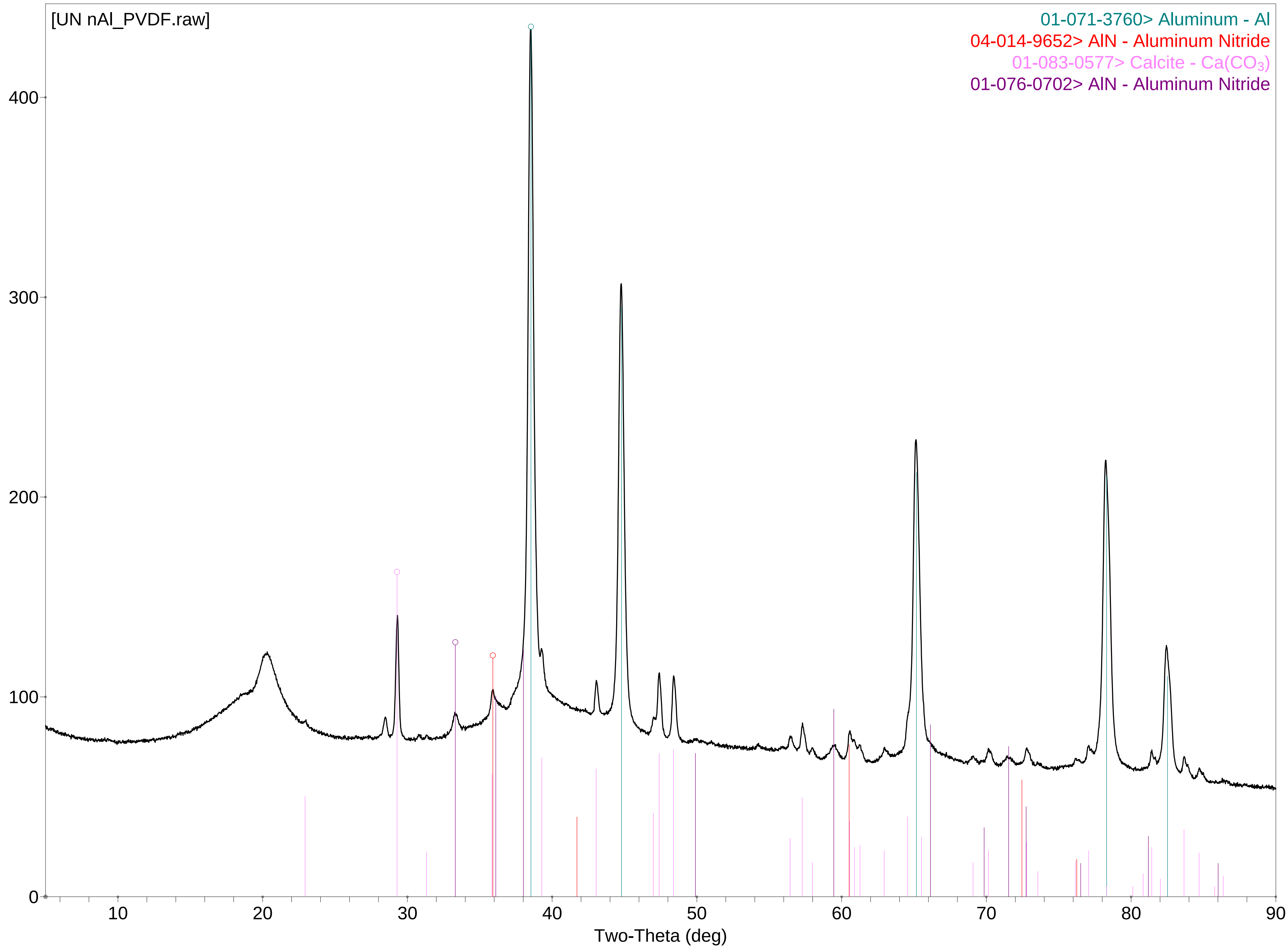


Refinement Iterations



Wt%




[UN nAl_PVDF.raw]
01-071-3760> Aluminum - Al
04-014-9652> AlN - Aluminum Nitride
01-083-0577> Calcite - Ca(CO3)
01-076-0702> AlN - Aluminum Nitride
400
300
200
100
0
10
20
30
40
50
60
70
80
90
Two-Theta (deg)

Whole Pattern Fitting and Rietveld Refinement

FILE: [PS nAl_PVDF.raw] PS nAl_PVDF - Pantoya
SCAN: 5.0/90.0/0.02/2.4(sec), Cu(30kV,15mA), I(p)=275558, 05/22/19 08:18p
PROC: [WPF Control File]

- ☑ K-alpha2 Peak Present
- ☑ Allow Negative Isotropic B
- ☑ Allow Negative Occupancy
- ☑ Apply Anomalous Scattering

[Diffractometer LP] Two-Theta Range of Fit = 5.0 - 90.0(deg)

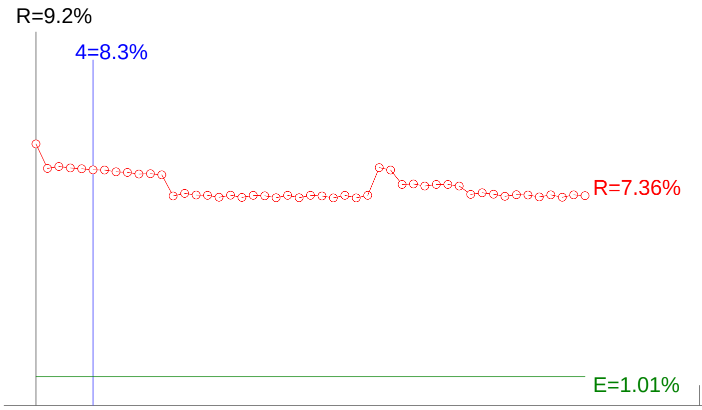
- ☑ Zero Offset of Goniometer - 2Theta = 0.923205
- ☑ Specimen Displacement - Cos(Theta) = -0.580608
- ☐ Monochromator Correction for LP Factor = 1.0
- ☑ K-alpha2/K-alpha1 Intensity Ratio = 0.528262(0.018076)

Profile Shape Function (PSF) for All Phases: pseudo-Voigt, Polynomial(5), Lambda=1.54059Å (Cu/K-alpha1)

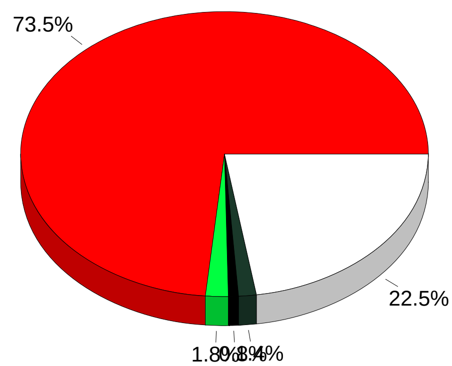
Phase ID (4)	Source	I/Ic	Wt%	#L	PC
Aluminum - Al	PDF#01-071-3760	4.10(5%)	73.5 (4.7)	5	SHF(6,2)
Aluminum Nitride - AlN	PDF#04-014-9652	3.57(0%)	1.8 (0.1)	10	<None>
Calcite - $Ca(CO_3)$	PDF#01-083-0577	3.21(5%)	0.8 (0.1)	33	(104)=3.308
Aluminum Nitride - AlN	PDF#01-076-0702	1.48(5%)	1.4 (0.2)	12	<None>
Others + Amorphous			22.5 (1.0)		

XRF(Wt%): Ca=0.3%, Si=10.5%, Al=75.6%, O=12.4%, N=1.1%, C=0.1%

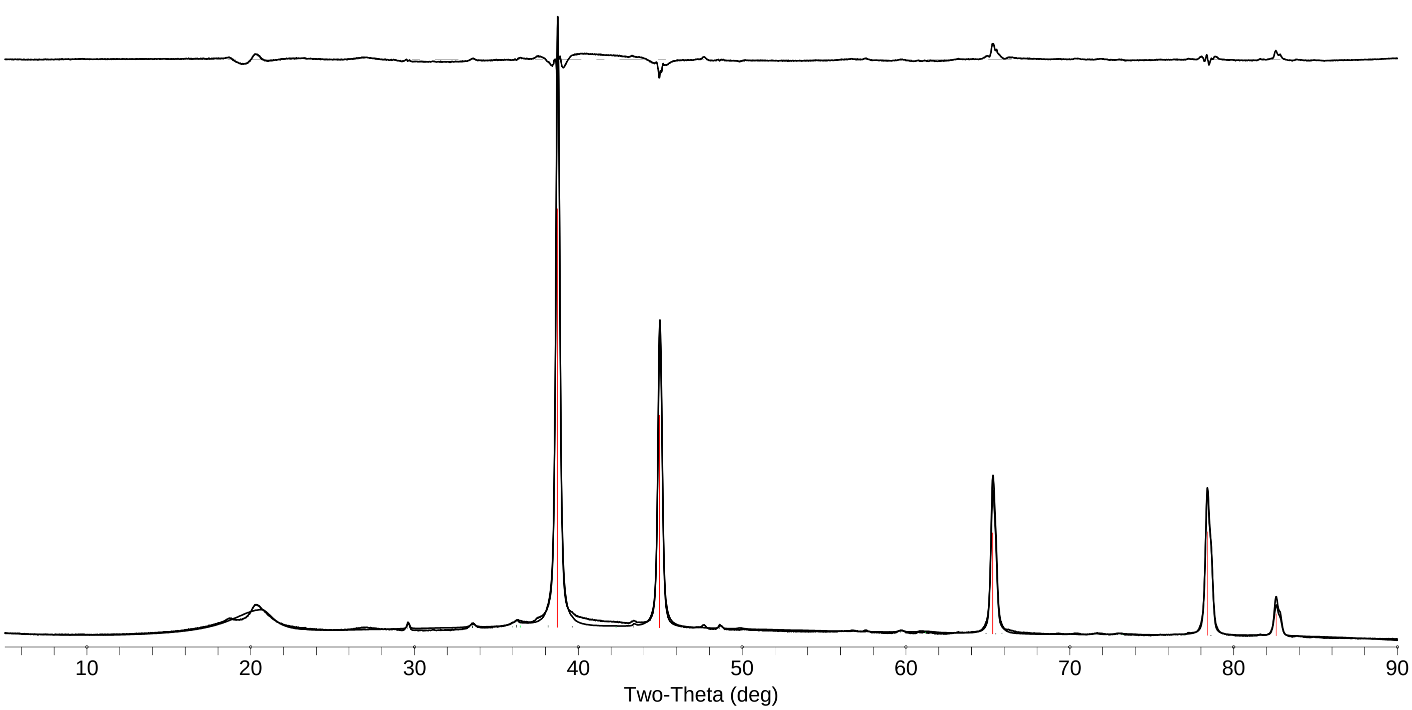
NOTE: Fitting Halted at Iteration 49(4): R=7.36% (E=1.01%, R/E=7.3, P=40, EPS=0.5)

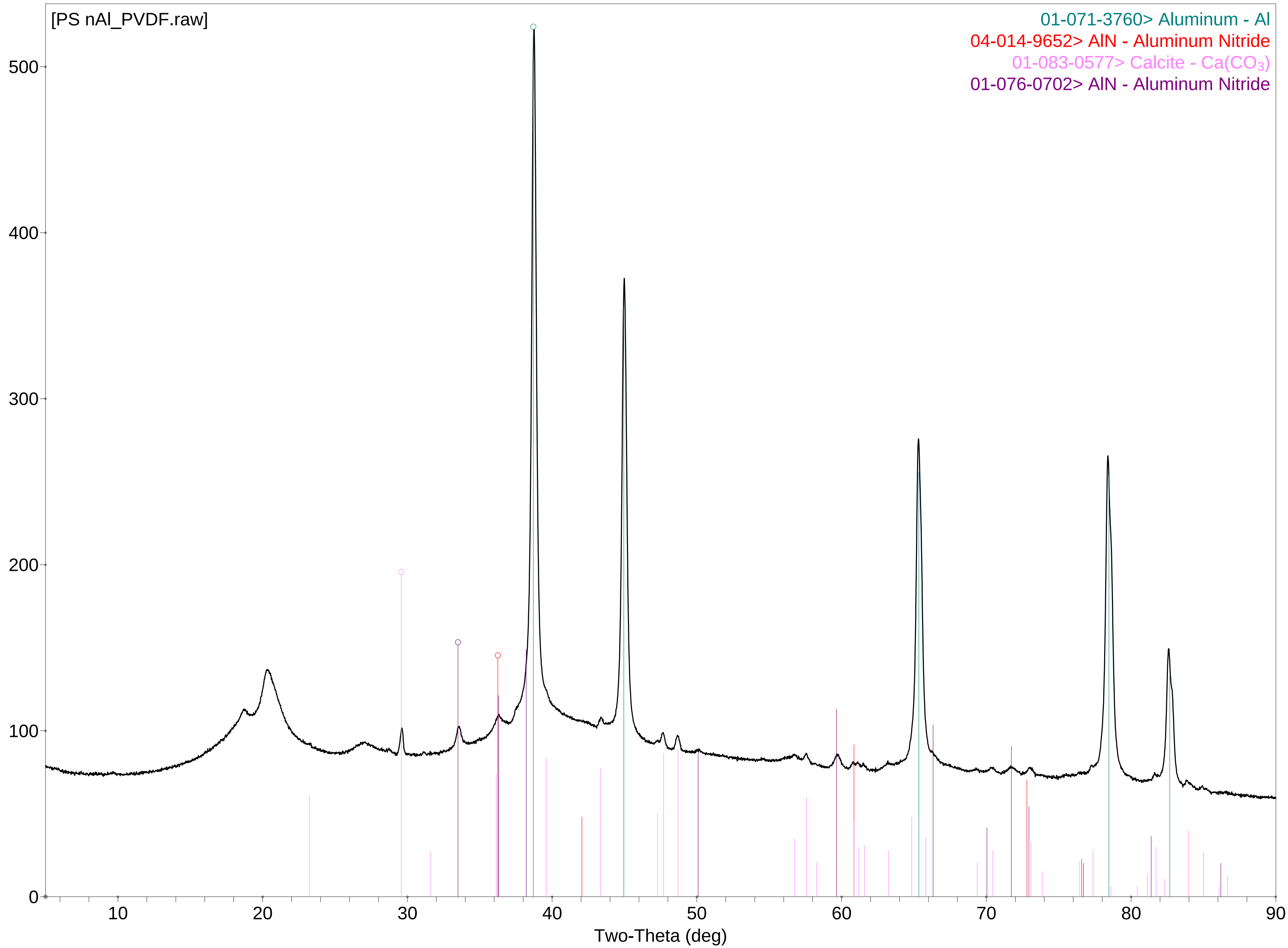


Refinement Iterations



Wt%





[PS nAl_PVDF.raw]
01-071-3760> Aluminum - Al
04-014-9652> AlN - Aluminum Nitride
01-083-0577> Calcite - Ca(CO3)
01-076-0702> AlN - Aluminum Nitride
0
100
200
300
400
500
10
20
30
40
50
60
70
80
90
Two-Theta (deg)

Whole Pattern Fitting and Rietveld Refinement

FILE: [SQ nAl_PVDF.raw] SQ nAl_PVDF - Pantoya
SCAN: 5.0/90.0/0.02/2.4(sec), Cu(30kV,15mA), I(p)=305819, 05/22/19 11:18p
PROC: [WPF Control File]

- ☑ K-alpha2 Peak Present
- ☑ Allow Negative Isotropic B
- ☑ Allow Negative Occupancy
- ☑ Apply Anomalous Scattering

[Diffractometer LP] Two-Theta Range of Fit = 5.0 - 90.0(deg)

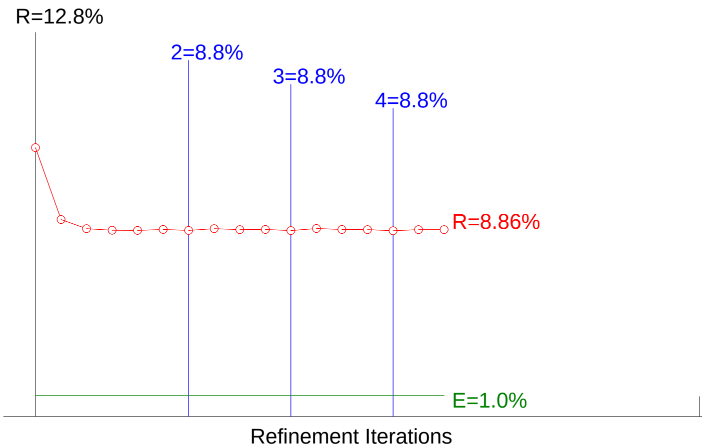
- ☑ Zero Offset of Goniometer - 2Theta = 0.955932
- ☑ Specimen Displacement - Cos(Theta) = -0.537551
- ☐ Monochromator Correction for LP Factor = 1.0
- ☑ K-alpha2/K-alpha1 Intensity Ratio = 0.510937(0.019619)

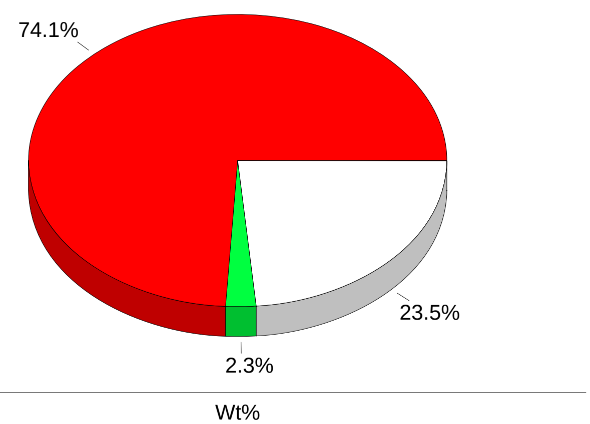
Profile Shape Function (PSF) for All Phases: pseudo-Voigt, Polynomial(5), Lambda=1.54059Å (Cu/K-alpha1)

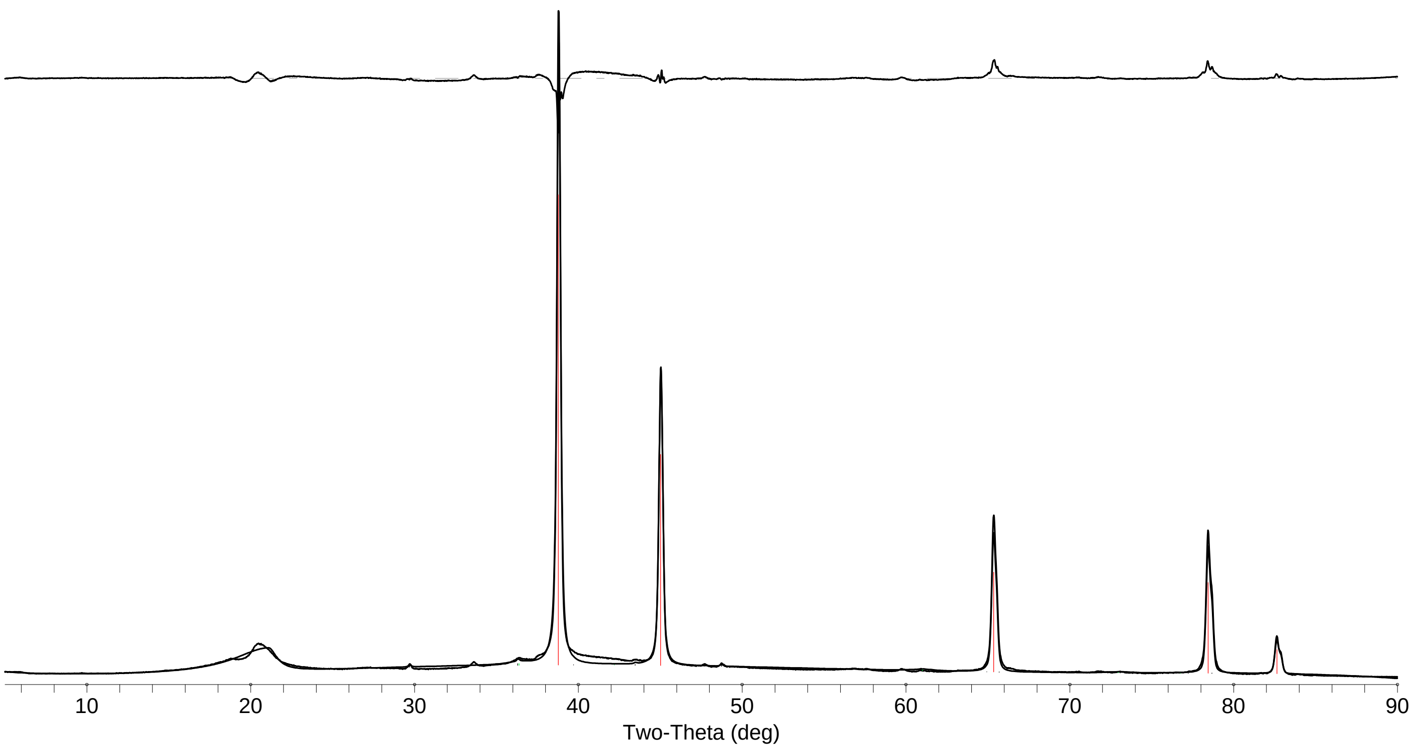
Phase ID (3)	Source	I/Ic	Wt%	#L	PC
Aluminum - Al	PDF#01-071-3760	4.10(5%)	74.1 (4.8)	5	(111)=1.000
Aluminum Nitride - AlN	PDF#04-014-9652	3.56(0%)	2.3 (0.2)	10	<None>
Calcite - $Ca(CO_3)$	PDF#01-083-0577	3.21(5%)	0.1 (0.0)	33	(104)=3.909
Others + Amorphous			23.5 (1.0)		

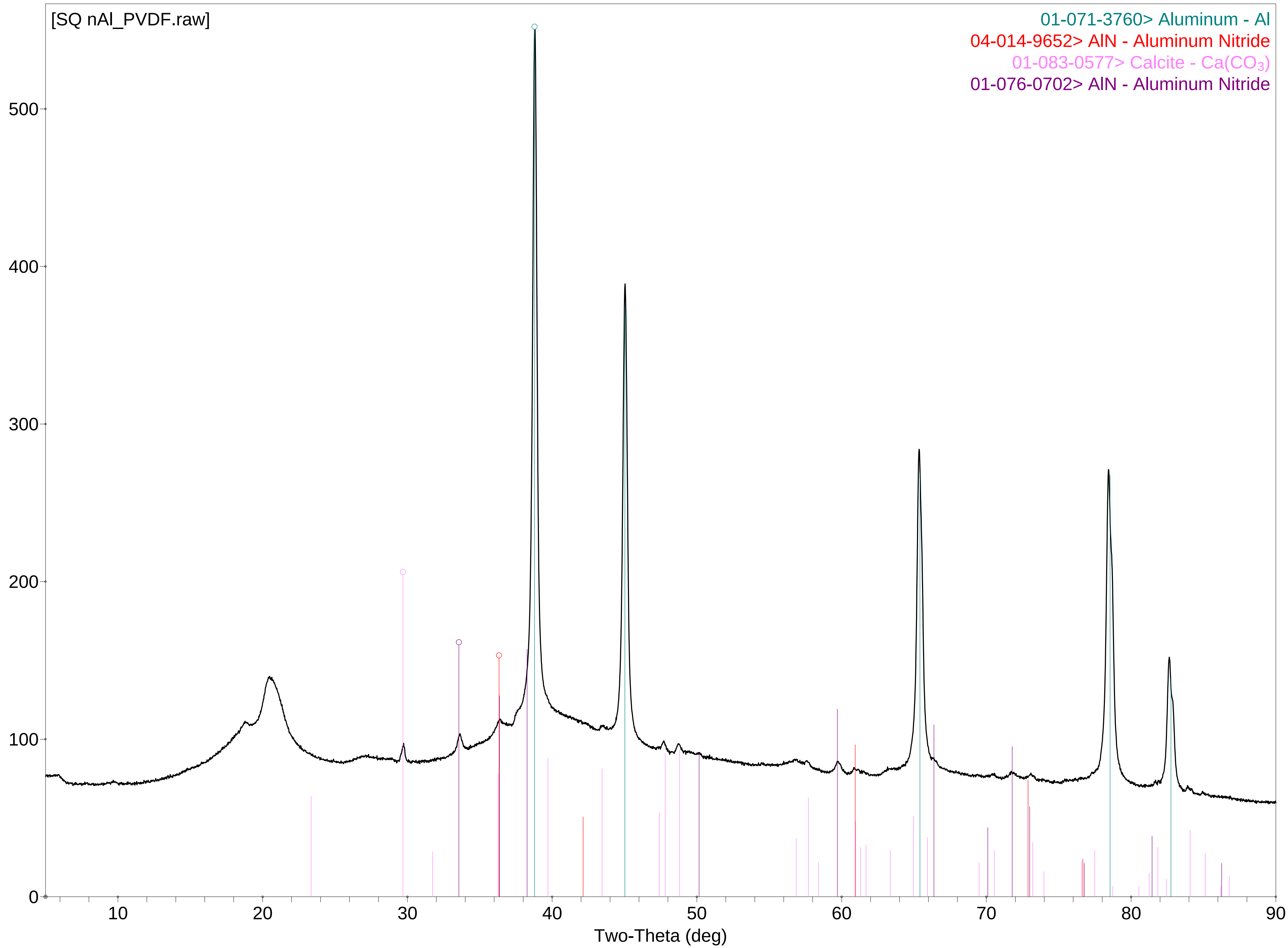
XRF(Wt%): Ca=0.0%, Si=11.0%, Al=75.6%, O=12.6%, N=0.8%, C=0.0%

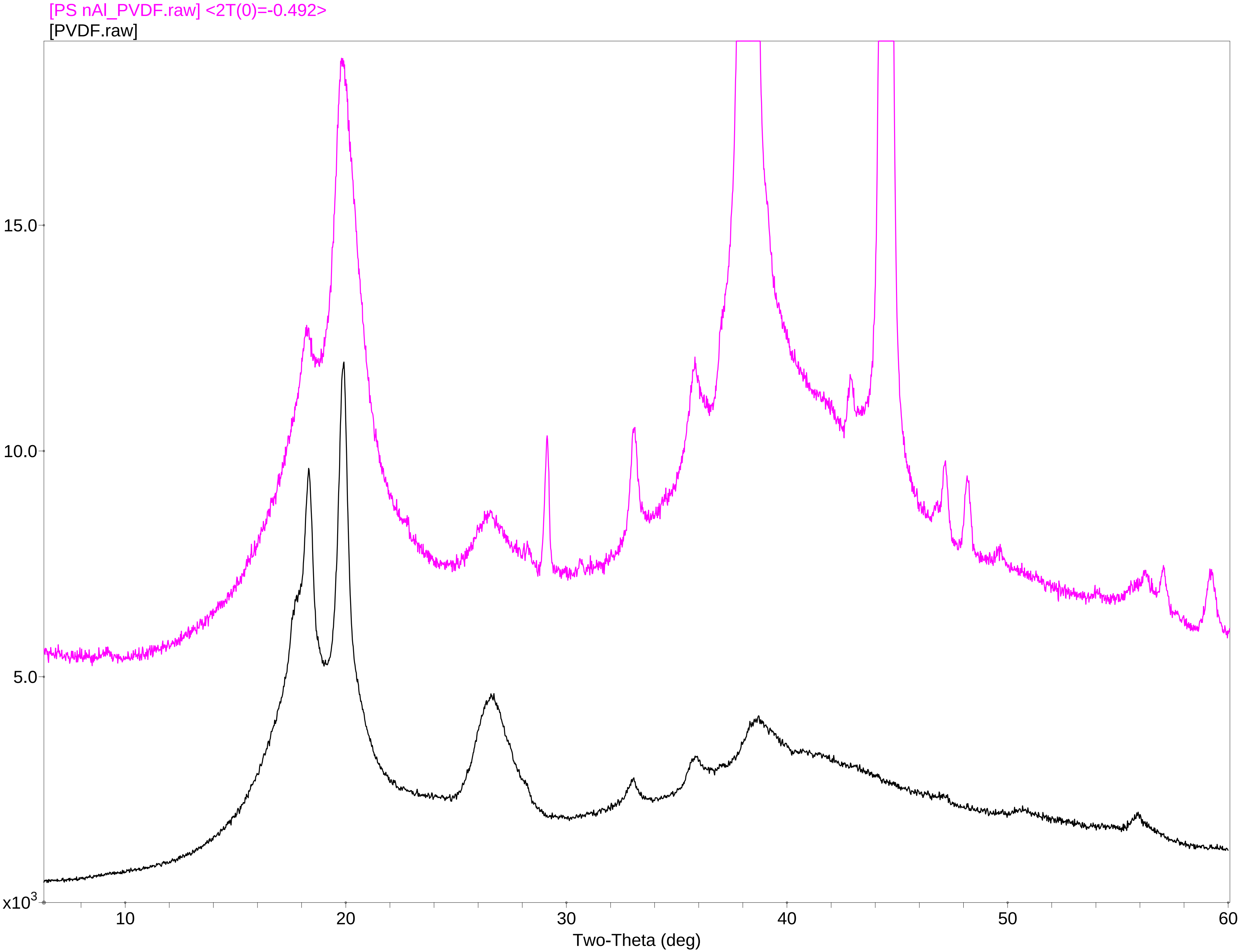
NOTE: Fitting Halted at Iteration 17(4): R=8.86% (E=1.0%, R/E=8.9, P=35, EPS=0.5)








[SQ nAl_PVDF.raw]
01-071-3760> Aluminum - Al
04-014-9652> AlN - Aluminum Nitride
01-083-0577> Calcite - Ca(CO3)
01-076-0702> AlN - Aluminum Nitride
0
100
200
300
400
500
10
20
30
40
50
60
70
80
90
Two-Theta (deg)



[PS nAl_PVDF.raw] <2T(0)=-0.492>
[PVDF.raw]
15.0
10.0
5.0
x10^3
10
20
30
40
50
60
Two-Theta (deg)

Whole Pattern Fitting and Rietveld Refinement

FILE: [50Al_50PVDF.raw] 50Al_50PVDF - Pantoya
SCAN: 3.0/90.0/0.02/5.217391(sec), Cu(30kV,15mA), I(p)=1529, 10/25/18 02:55p
PROC: [WPF Control File]

- ☑ K-alpha2 Peak Present
- ☑ Allow Negative Isotropic B
- ☑ Allow Negative Occupancy
- ☑ Apply Anomalous Scattering

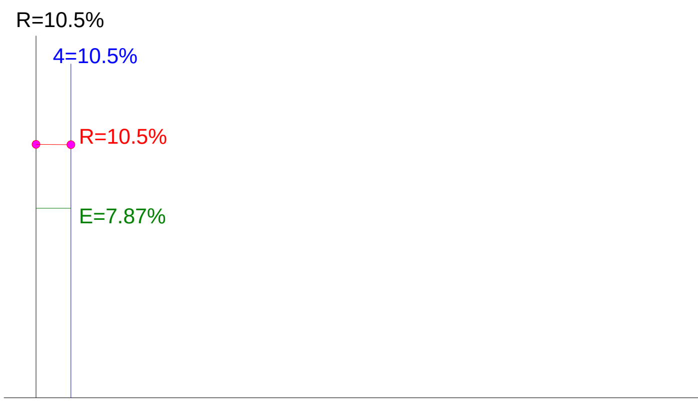
[Diffractometer LP] Two-Theta Range of Fit = 3.0 - 90.0(deg)
- ☑ Zero Offset of Goniometer - 2Theta = 0.017722(0.389361)
- ☑ Specimen Displacement - Cos(Theta) = 0.021622(0.369634)
- ☑ Monochromator Correction for LP Factor = 1.0(0.120435)
- ☑ K-alpha2/K-alpha1 Intensity Ratio = 0.500592(0.067132)

Profile Shape Function (PSF) for All Phases: pseudo-Voigt, Polynomial(5), Lambda=1.54059Å (Cu/K-alpha1)

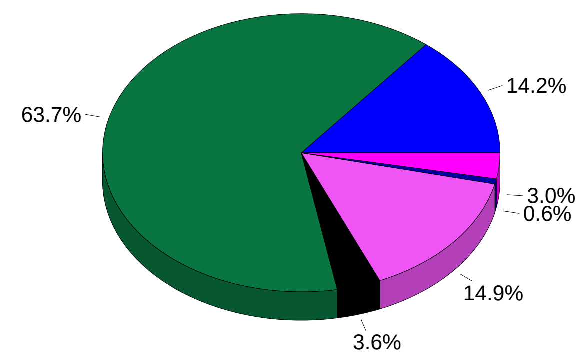
Phase ID (6)	Source	I/Ic	Wt%	#L	PC
Aluminum - Al	PDF#98-000-0062	4.52(0%)	14.2 (1.2)	5	(111)=0.900
Aluminum Fluoride - AlF_3	PDF#01-077-0252	3.22(5%)	63.7 (5.5)	29	<None>
Bismuth - $Bi_{0.916}Sb_{0.084}$	PDF#04-007-5318	23.28(0%)	3.6 (0.3)	31	<None>
Potassium Aluminum Fluoride - K_2AlF_5	PDF#00-052-0735	1.00(5%)	14.9 (2.8)	14	(100)=0.900
Aluminum Phosphate - $Al(PO_4)$	PDF#04-011-7504	17.27(0%)	0.6 (0.5)	4188	(110)=0.900
Sodium n - C_3H_7NaO	PDF#00-057-1658	5.30(5%)	3.0 (0.8)	27	(001)=0.900

XRF(Wt%): Bi=3.4%, Sb=0.2%, K=5.8%, P=0.2%, Al=36.8%, Na=0.8%, F=50.3%, O=0.9%, C=1.3%, H=0.3%

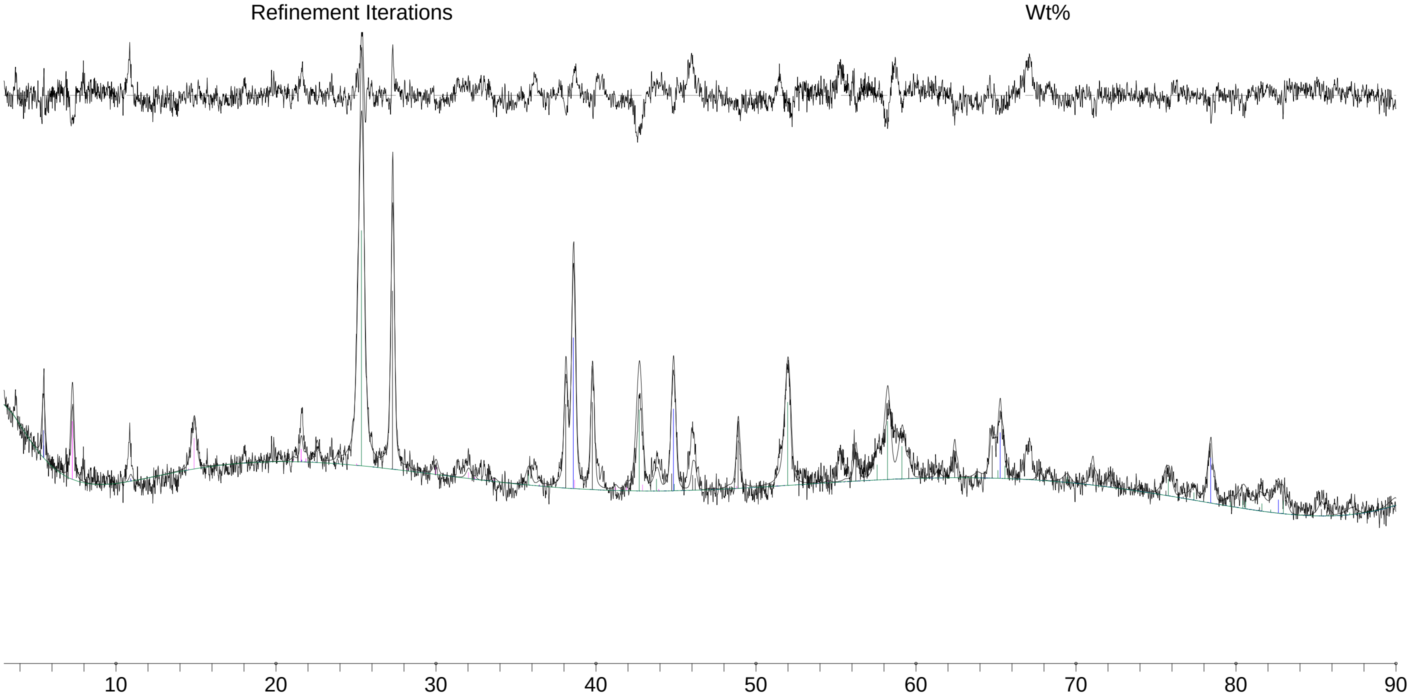
NOTE: Fitting Halted at Iteration 2(4): R=10.5% (E=7.87%, R/E=1.33, P=46, EPS=0.5)

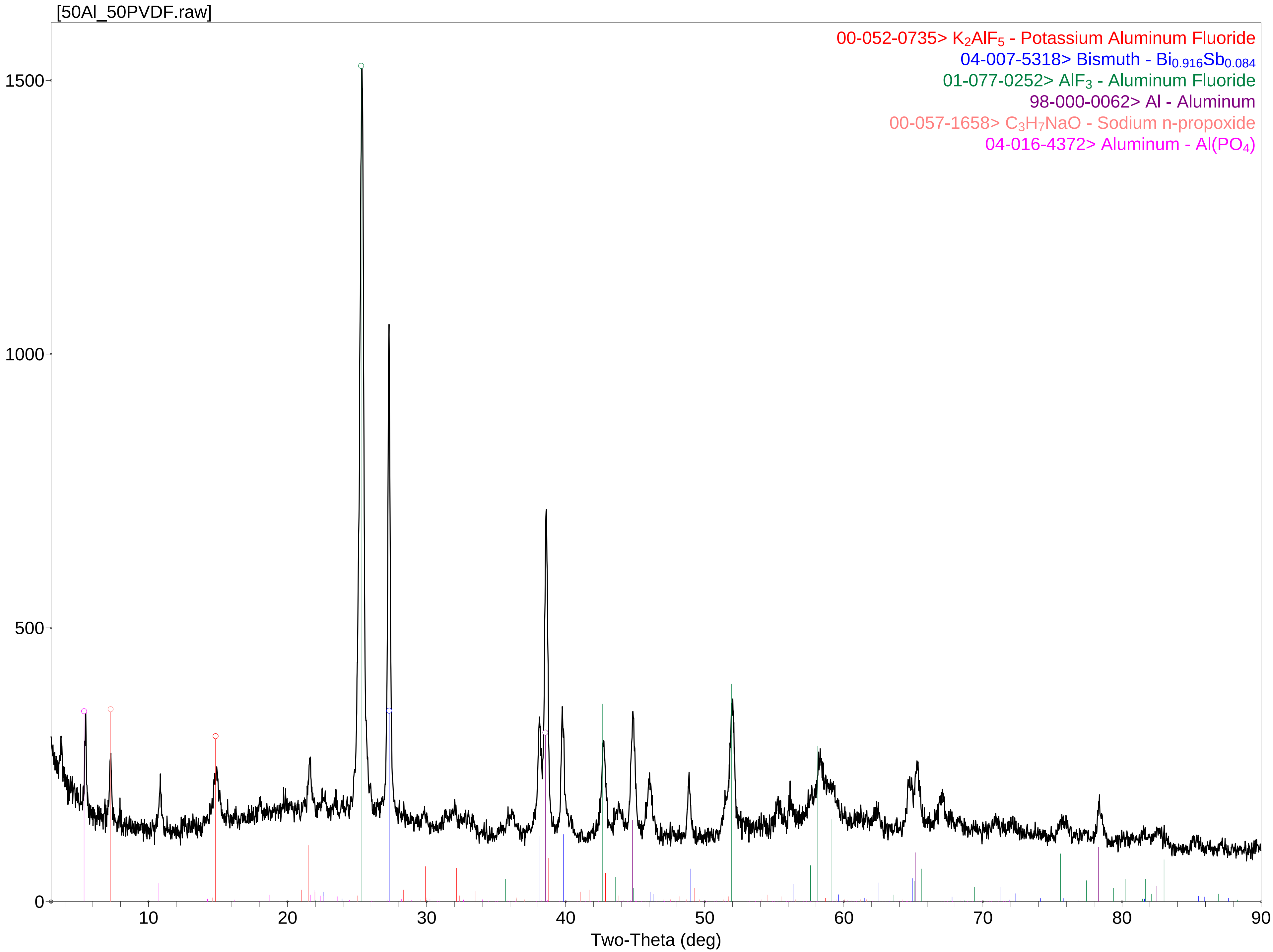


Refinement Iterations



Wt%




[50Al_50PVDF.raw]
00-052-0735> K2AlF5 - Potassium Aluminum Fluoride
04-007-5318> Bismuth - Bi0.916Sb0.084
01-077-0252> AlF3 - Aluminum Fluoride
98-000-0062> Al - Aluminum
00-057-1658> C3H7NaO - Sodium n-propoxide
04-016-4372> Aluminum - Al(PO4)
1500
1000
500
0
10
20
30
40
50
60
70
80
90
Two-Theta (deg)

Whole Pattern Fitting and Rietveld Refinement

FILE: [50PS_nAl_50PVDF.raw] 50PS_nAl_50PVDF - Pantoya
SCAN: 3.0/90.0/0.02/12(sec), Cu(30kV,15mA), I(p)=4334, 10/25/18 06:33a
PROC: [WPF Control File]

☑ K-alpha2 Peak Present
☑ Allow Negative Isotropic B
☑ Allow Negative Occupancy
☑ Apply Anomalous Scattering

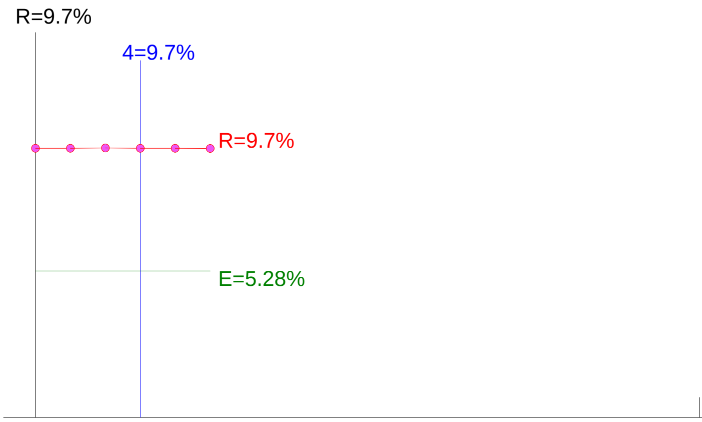
[Diffractometer LP] Two-Theta Range of Fit = 3.0 - 90.0(deg)
☑ Zero Offset of Goniometer - 2Theta = -0.983396(0.442371)
☑ Specimen Displacement - Cos(Theta) = 0.969797(0.42124)
☑ Monochromator Correction for LP Factor = 1.0(0.252622)
☑ K-alpha2/K-alpha1 Intensity Ratio = 0.502809(0.100378)

Profile Shape Function (PSF) for All Phases: pseudo-Voigt, Polynomial(5), Lambda=1.54059Å (Cu/K-alpha1)

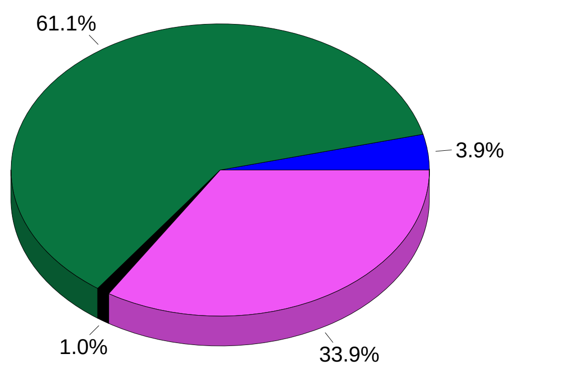
Phase ID (4)	Source	I/Ic	Wt%	#L	PC
Aluminum - Al	PDF#98-000-0062	4.54(0%)	3.9 (0.5)	5	(111)=0.900
Aluminum Fluoride - AlF_3	PDF#01-077-0252	3.22(5%)	61.1 (6.5)	29	(012)=0.894
Bismuth - $Bi_{0.916}Sb_{0.084}$	PDF#04-007-5318	24.07(0%)	1.0 (0.1)	32	(012)=0.900
Potassium Aluminum Fluoride - K_2AlF_5	PDF#00-052-0735	1.00(5%)	33.9 (4.0)	14	(100)=0.899

XRF(Wt%): Bi=1.0%, Sb=0.1%, K=13.3%, Al=28.1%, F=57.6%

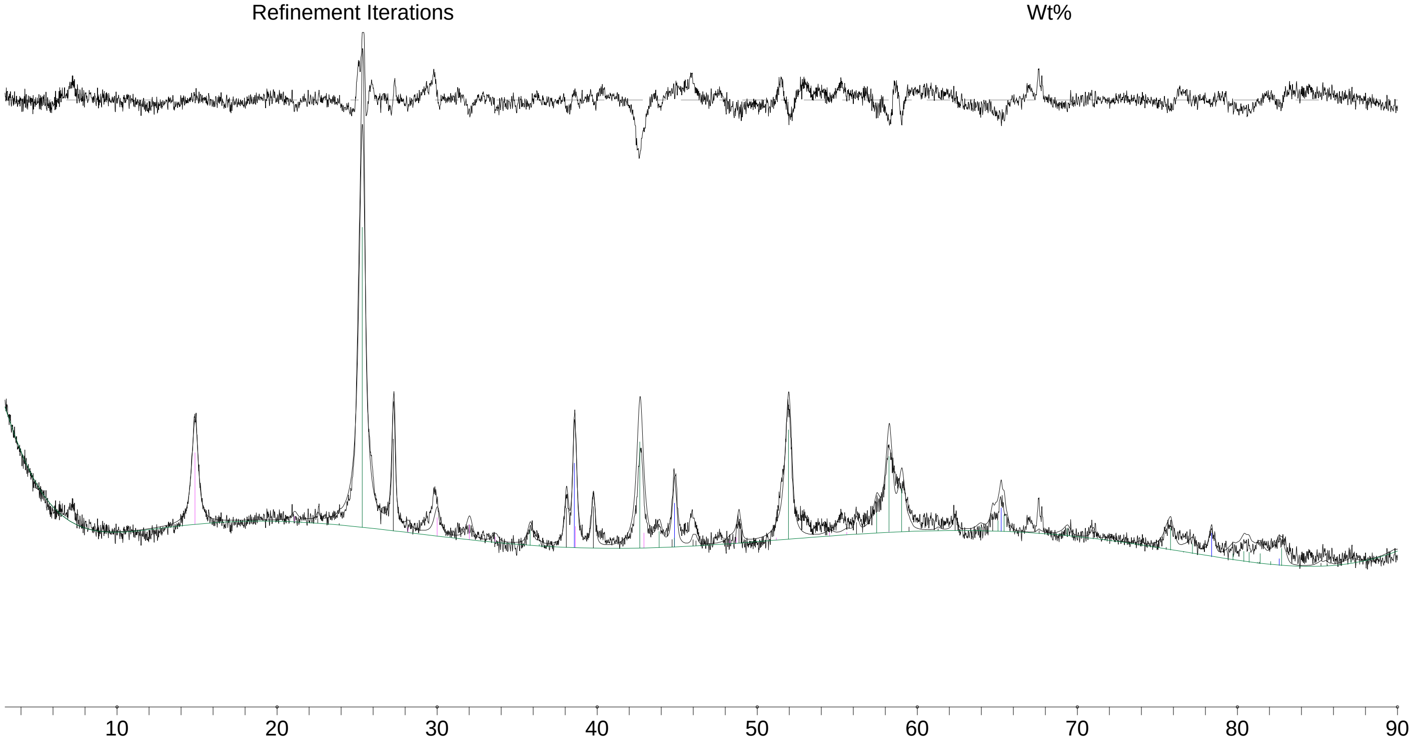
NOTE: Fitting Halted at Iteration 6(4): R=9.7% (E=5.28%, R/E=1.84, P=36, EPS=0.5)

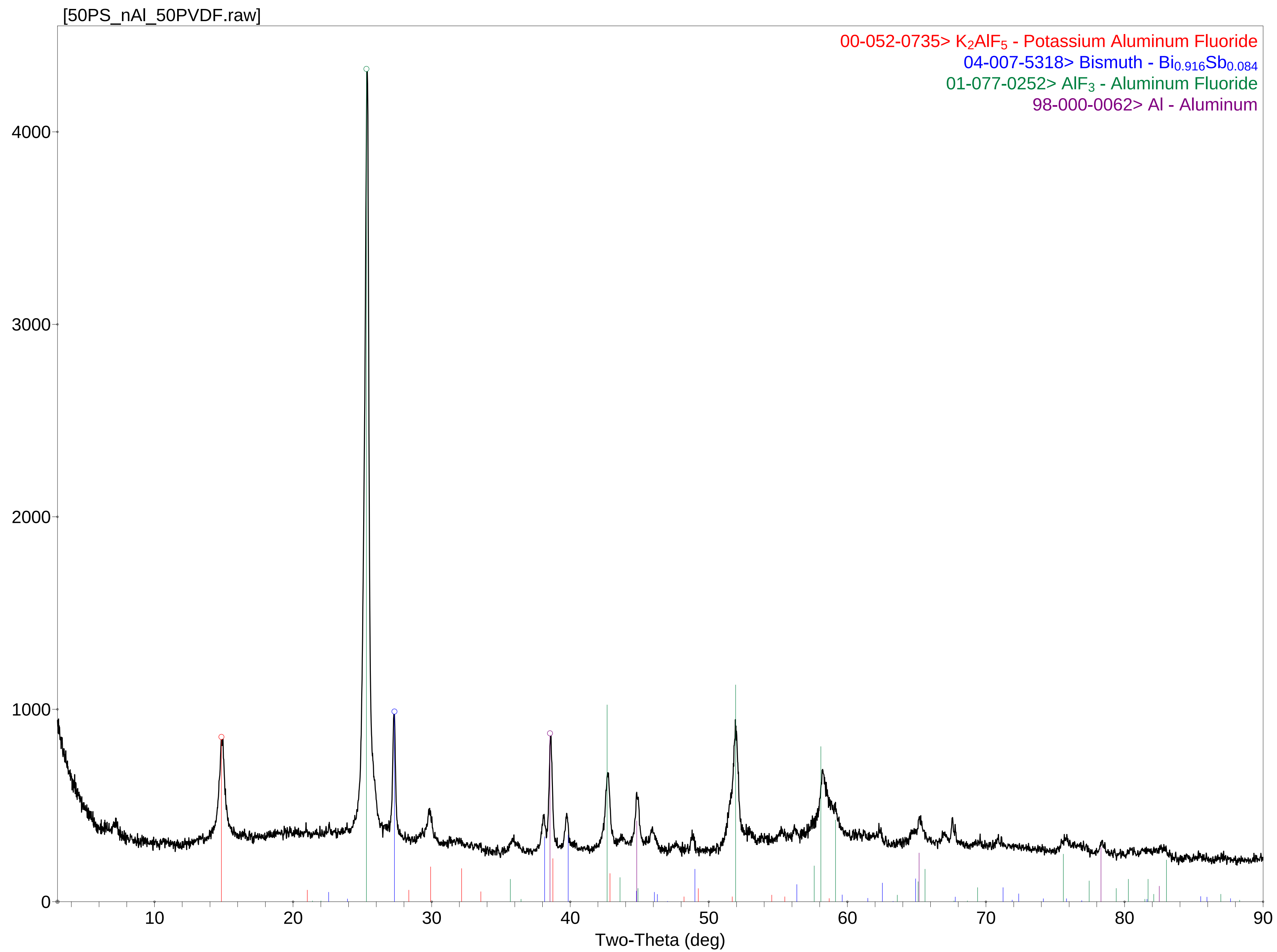


Refinement Iterations



Wt%




[50PS_nAl_50PVDF.raw]
00-052-0735> K_2AlF_5 - Potassium Aluminum Fluoride
04-007-5318> Bismuth - $Bi_{0.916}Sb_{0.084}$
01-077-0252> AlF_3 - Aluminum Fluoride
98-000-0062> Al - Aluminum
4000
3000
2000
1000
0
10
20
30
40
50
60
70
80
90
Two-Theta (deg)

Whole Pattern Fitting and Rietveld Refinement

FILE: [50SQ_nAl_50PVDF.raw] 50SQ_nAl_50PVDF - Pantoya
SCAN: 3.0/90.0/0.02/12(sec), Cu(30kV,15mA), I(p)=14082, 10/26/18 08:08a
PROC: [WPF Control File]

- ☑ K-alpha2 Peak Present
- ☑ Allow Negative Isotropic B
- ☑ Allow Negative Occupancy
- ☑ Apply Anomalous Scattering

[Diffractometer LP] Two-Theta Range of Fit = 3.0 - 90.0(deg)

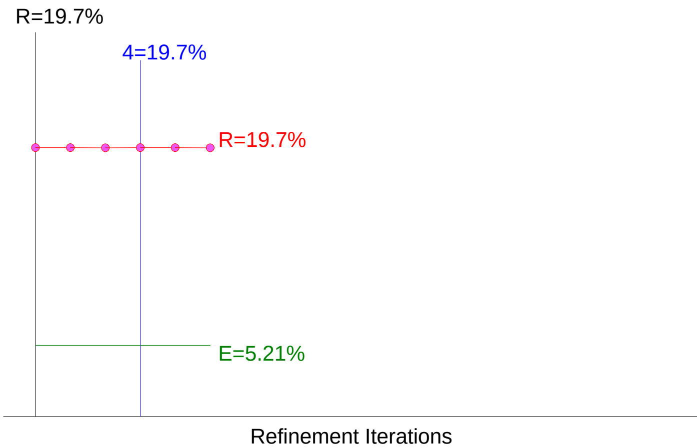
- ☑ Zero Offset of Goniometer - 2Theta = -0.896797(0.707023)
- ☑ Specimen Displacement - Cos(Theta) = 0.879222(0.672211)
- ☑ Monochromator Correction for LP Factor = 1.0(0.379075)
- ☑ K-alpha2/K-alpha1 Intensity Ratio = 0.502811(0.086697)

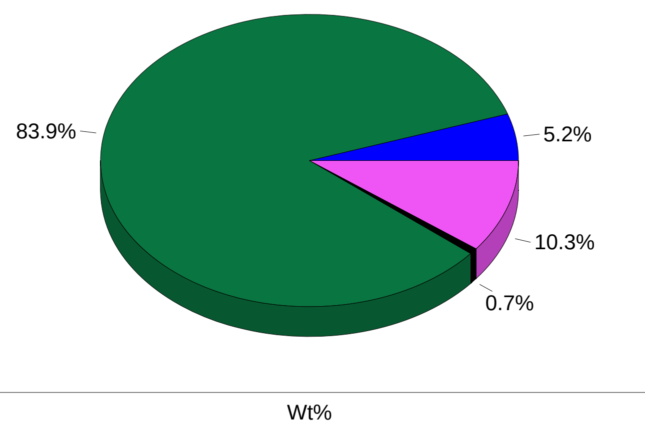
Profile Shape Function (PSF) for All Phases: pseudo-Voigt, Polynomial(5), Lambda=1.54059Å (Cu/K-alpha1)

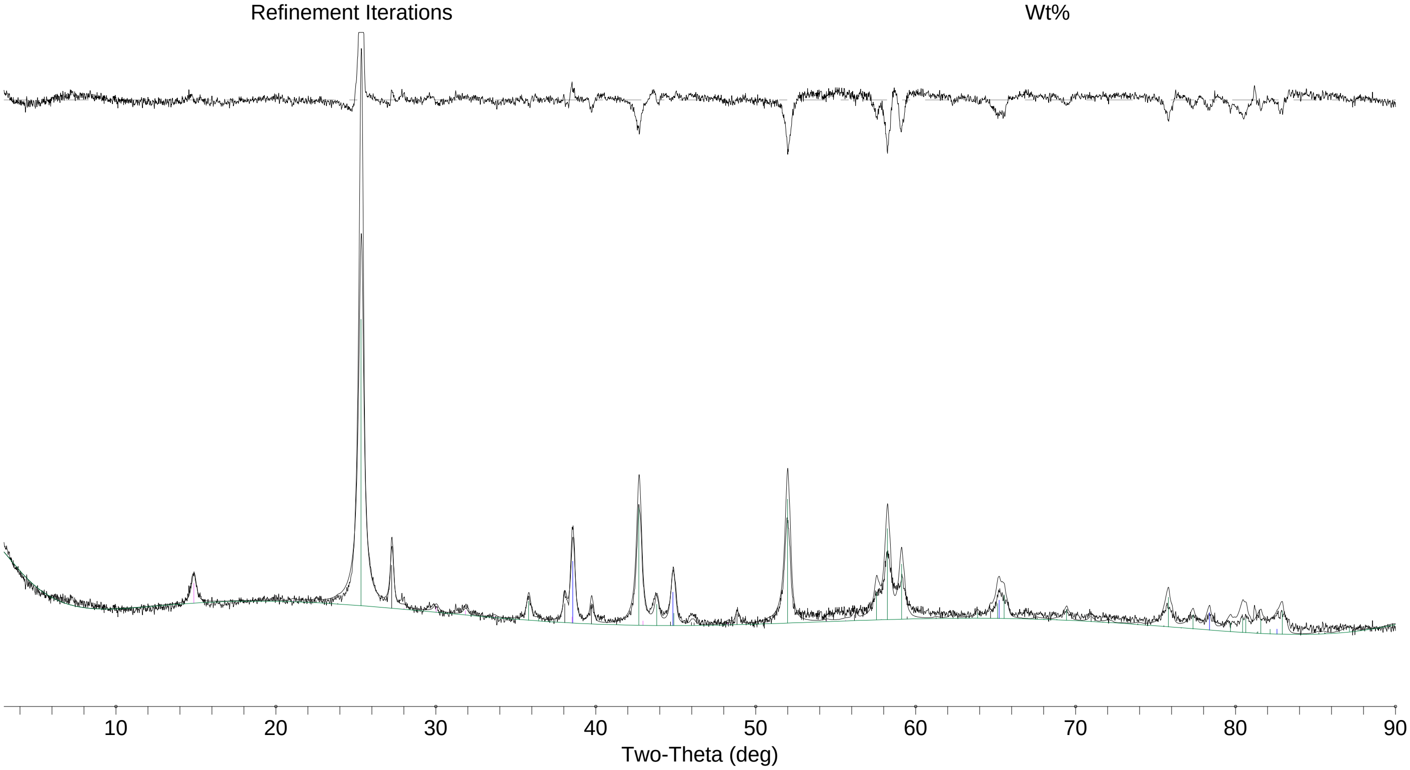
Phase ID (4)	Source	I/Ic	Wt%	#L	PC
Aluminum - Al	PDF#98-000-0062	4.54(0%)	5.2 (0.8)	5	(111)=0.900
Aluminum Fluoride - AlF_3	PDF#01-077-0252	3.22(5%)	83.9 (9.0)	29	(012)=0.893
Bismuth - $Bi_{0.916}Sb_{0.084}$	PDF#04-007-5318	24.07(0%)	0.7 (0.1)	32	(012)=0.900
Potassium Aluminum Fluoride - K_2AlF_5	PDF#00-052-0735	1.00(5%)	10.3 (3.0)	14	(100)=0.899

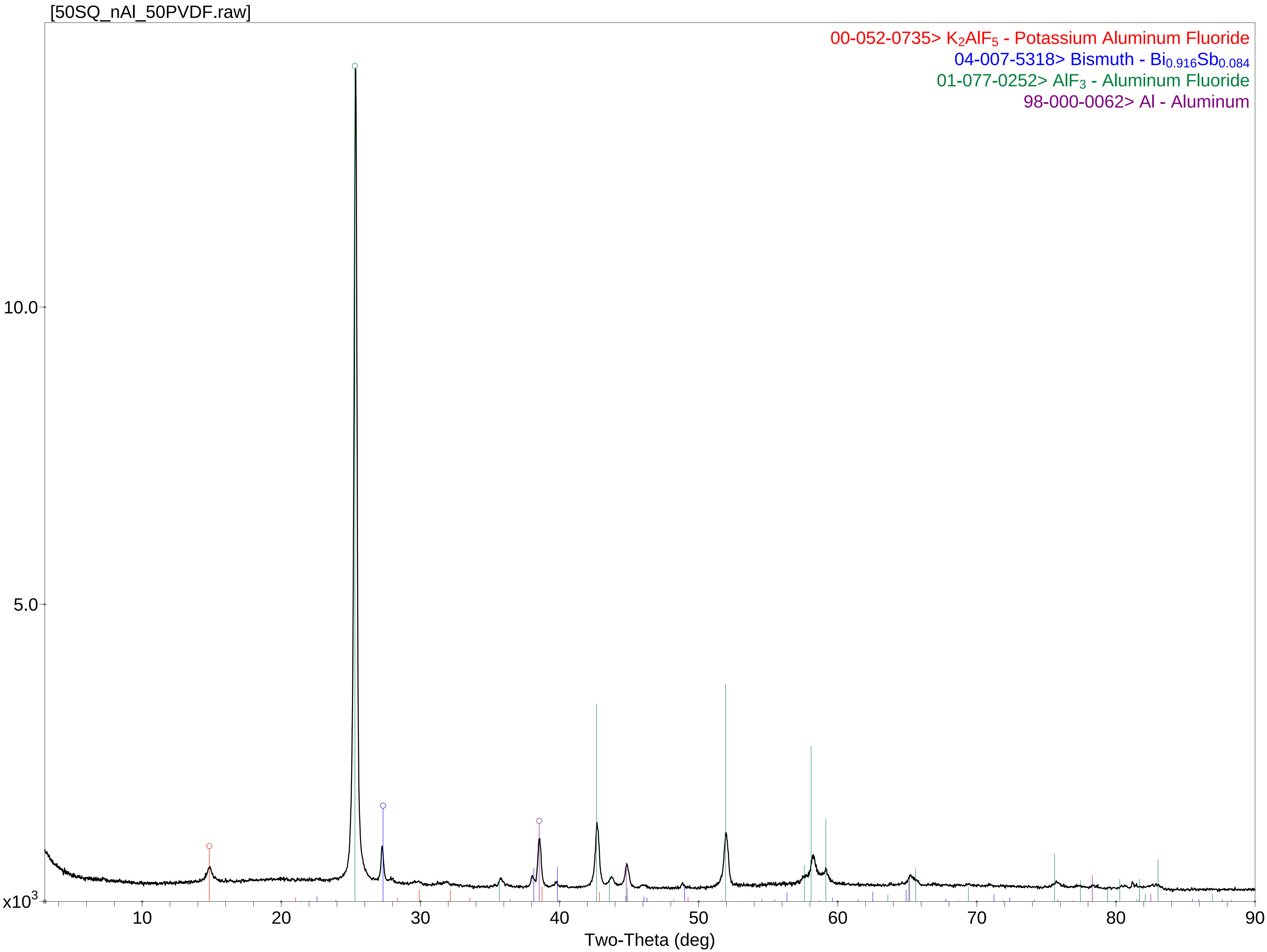
XRF(Wt%): Bi=0.6%, Sb=0.0%, K=4.0%, Al=33.5%, F=61.8%

NOTE: Fitting Halted at Iteration 6(4): R=19.7% (E=5.21%, R/E=3.78, P=36, EPS=0.5)









[50SQ_nAl_50PVDF.raw]
00-052-0735> K2AlF5 - Potassium Aluminum Fluoride
04-007-5318> Bismuth - Bi0.916Sb0.084
01-077-0252> AlF3 - Aluminum Fluoride
98-000-0062> Al - Aluminum
10.0
5.0
x10^3
10
20
30
40
50
60
70
80
90
Two-Theta (deg)

S3. Derivation of the equation for stresses in a spherical particle covered by a shell

S3.1. Stresses and strains in the hollow sphere (alumina shell) with constant creep strain

We will generalize approach for spherically symmetric problem presented in [1] for homogeneously distributed creep strain. Using spherical coordinate system with radial coordinate *r*, the radial ε_r and hoop ε_h strains are expressed through displacements *u*,

$$\varepsilon_r = \frac{du}{dr};\ \varepsilon_h = \frac{u}{r}. \tag{S.1}$$

These strainssatisfy the compatibility condition

$$\varepsilon_r = \frac{d(r\varepsilon_h)}{dr} = r\frac{d\varepsilon_h}{dr} + \varepsilon_h. \tag{S.2}$$

The equilibrium equation is

$$\frac{d\sigma_r}{dr} + 2\frac{\sigma_r - \sigma_h}{r} = 0, \tag{S.3}$$

where subscripts *r* and *h* are for radial and hoop stresses. The Hooke's law combined with additivity of the elastic (superscript *e*) and inelastic (superscript *in*) strains is presented as

$$\varepsilon_r = \frac{\sigma_r - 2\nu\sigma_h}{E} + \varepsilon_r^{in};\ \varepsilon_h = \frac{(1-\nu)\sigma_h - \nu\sigma_r}{E} + \varepsilon_h^{in}, \tag{S.4}$$

where E is the Young modulus and ν is the Poison's ratio. Substituting strains (S.4) into compatibility conditions (S.2) results in

$$r\frac{d}{dr}((1-\nu)\sigma_h - \nu\sigma_r) + (1+\nu)(\sigma_h - \sigma_r) = E(\varepsilon_r^{in} - \varepsilon_h^{in}). \tag{S.5}$$

Expressing $\sigma_h - \sigma_r$ via equilibrium equation (S.3) and substituting into Eq. (S.5) leads to

$$\frac{d}{dr}(\sigma_r + 2\sigma_h) = \frac{2E(\varepsilon_r^{in} - \varepsilon_h^{in})}{r(1-\nu)}, \tag{S.6}$$

or after integration

$$(\sigma_r + 2\sigma_h) = \frac{2E(\varepsilon_r^{in} - \varepsilon_h^{in})}{(1-\nu)}\ln r + A, \tag{S.7}$$

where *A* is the integration constant. Let us evaluate

$$\frac{2}{3}\frac{d(\sigma_h-\sigma_r)}{dr}=-\frac{d\sigma_r}{dr}+\frac{2E(\varepsilon_r^{in}-\varepsilon_h^{in})}{3r(1-\nu)}=-\frac{2(\sigma_h-\sigma_r)}{r}+\frac{2E(\varepsilon_r^{in}-\varepsilon_h^{in})}{3r(1-\nu)}, \quad (S.8)$$

where first Eq. (S.6) and then Eq. (S.3) were utilized. After integration we obtain

$$\sigma_h-\sigma_r=\frac{3C}{r^3}+\frac{E(\varepsilon_r^{in}-\varepsilon_h^{in})}{3(1-\nu)}, \quad (S.9)$$

where C is the integration constant. Solving Eqs. (S.7) and (S.9) for stresses,

we obtain for the hollow sphere

$$\sigma_r=\frac{A}{3}-\frac{2C}{r^3}+\frac{2E(\varepsilon_r^{in}-\varepsilon_h^{in})}{3(1-\nu)}(\ln r-{}^1\!/_3); \qquad \sigma_h=\frac{A}{3}+\frac{C}{r^3}+\frac{2E(\varepsilon_r^{in}-\varepsilon_h^{in})}{3(1-\nu)}\ln r. \quad (S.10)$$

Substituting these expressions in Eq. (S.4) for ε_h and Eq. (S.1) for u, one obtains

$$u=\varepsilon_h r=r(\frac{A(1-2\nu)r^3+3C(1+\nu)}{3Er^3}+\frac{(\varepsilon_r^{in}-\varepsilon_h^{in})((1+\nu)+6(1-2\nu)\ln r)}{9(1-\nu)}+\varepsilon_h^{in}). \quad (S.11)$$

Inelastic strains consists of isotropic thermal strain ε^T and volume preserving creep strains, i.e.

$$\varepsilon_h^{in}=\varepsilon^T+\varepsilon_h^c;\ \varepsilon_r^{in}=\varepsilon^T+\varepsilon_r^c. \quad (S.12)$$

Since two hoop strains are equal, the condition of absence of volumetric creep strain results in

$$\varepsilon_r^c=-2\varepsilon_h^c;\ \varepsilon_r^{in}=\varepsilon^T-\varepsilon_h^c. \quad (S.13)$$

Below it will be more convenient to present results in terms of bulk K and shear G moduli using

$$E=\frac{9KG}{(3K+G)}; \qquad \nu=\frac{(3K-2G)}{(3K+G)}. \quad (S.14)$$

S3.2. Stresses and strains in the internal sphere (aluminum core)

Aluminum core deforms thermoelastically and is under hydrostatic stress state

$$u_1=Br; \quad \varepsilon_{r1}=\varepsilon_{h1}=B;\ \sigma_{r1}=\sigma_{h1}=3K_1(B-\varepsilon_1^T), \quad (S.15)$$

where ε_1^T is the thermal strain in a core. We used subscript 1 for the core and will use below the subscript 2 for the shell.

S2.3. Stresses in a core-shell system

To find stresses in a core-shell system, we connect solution for the internal and external sphere with the displacement and radial stress continuity conditions at the core-shell boundary $r = r_1$ and boundary condition for the radial stress at the external shell boundary $r = r_2$:

$$u_1(r_1) = u_2(r_1);\ \sigma_{r1}(r_1) = \sigma_{r2}(r_1) - \frac{2\Gamma_1}{r_1};\ \sigma_{r2}(r_2) = -\frac{2\Gamma_2}{r_2}, \quad \text{(S.16)}$$

where Γ_1 and Γ_2 are the surface energies at the core-shell and shell-gas boundaries. After placing expressions for displacements and radial stresses (S.10), (S.11), and (S.15) in Eq. (S.16), one obtains system of 3 linear equations for 3 parameters A, B, and *C*. Substituting these parameters in expressions for stresses and strains, we obtain explicit expressions for them. In particular, we obtain for themean stress (negative pressure) in the Al core, $\sigma_0 = \sigma_{r1}$,and hoop stress in the shell at the boundary with the core, $\sigma_h = \sigma_{h2}(r_1)$:

$$\begin{aligned}\sigma_h = &-\frac{6(m^3+2)(\varepsilon_2^T-\varepsilon_1^T)G_2K_1K_2}{H} - \\ &\frac{18\varepsilon_c^h G_2K_2\left(4G_2K_2\left(m^3-1-3m^3\ln m\right)+K_1(4G_2+3K_2m^3+2(2G_2-3K_2)m^3\ln m)\right)}{(4G_2+3K_2)H} \\ &-\frac{2\Gamma_2 m^2(-2G_2K_1+3(2G_2+K_1)K_2)}{RH} - \frac{4(m^3+2)G_2K_2\Gamma_1}{RH};\end{aligned} \quad \text{(S.17)}$$

$$\begin{aligned}\sigma_0 = &\frac{12G_2K_1K_2((m^3-1)(\varepsilon_2^T-\varepsilon_1^T)+3\varepsilon_c^h m^3\ln m)}{H} - \frac{2\Gamma_2 m^2K_1(4G_2+3K_2)}{RH} \\ &-\frac{2K_1(4G_2+3m^3K_2)\Gamma_1}{RH};\end{aligned} \quad \text{(S.18)}$$

Here $H = 3m^3K_1K_2 + 4G_2(K_1 + (m^3-1)K_2)$, subscripts 1 and 2 designate Al and alumina, respectively, $R=r_1$ is the Al core radius, $m = 1 + 1/M$,and $M=R/\delta$.

References

[1] Lubliner J. L. Plasticity Theory (New York: Macmillan) 1990.