Crystal growth, structure and magnetic properties of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$: A compound in the $\text{Sm}_{2n+m}\text{Ni}_{4n+m}\text{Al}_{15n+4m}$ homologous series

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Abstract. $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ was obtained as large rod shaped single crystals from reactive aluminium flux. Single crystal X-ray diffraction suggests that $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ crystallizes in the $\text{Gd}_3\text{Ni}_5\text{Al}_{19}$ structure type, orthorhombic space group $Cmcm$ and lattice constants $a = 4.0974(1) \text{ \AA}$, $b = 16.0172(6) \text{ \AA}$ and $c = 27.0774(10) \text{ \AA}$. $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ is a member of the $\text{Sm}_{2n+m}\text{Ni}_{4n+m}\text{Al}_{15n+4m}$ series with $n = 1$ and $m = 1$. The crystal structure of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ consists of SmNiAl_4 and $\text{Sm}_2\text{Al}_4\text{Al}_{15}$ slabs intergrown along the b -axis. Magnetic susceptibility data on $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ confirms the paramagnetic nature with a complicated magnetic ordering below 18 K. The inverse susceptibility data follows modified Curie-Weiss law above 150 K with effective magnetic moment $1.3 \mu_B/\text{Sm}$ atoms suggests trivalent Sm atoms.

Keywords. Intermetallics; crystal growth; crystal structure; magnetism.

1. Introduction

The rare earth based intermetallic compounds have considerable interest in the past few decades because of their interesting properties such as Kondo behaviour,¹ heavy fermions,² superconductivity³ and magneto resistance.^{4–6} Considering that a large amount of reports are available in the literature, only a few samarium based intermetallics were studied for detailed structure and property studies. The classical examples are: magneto-resistance in SmRuSn_3 ,⁷ ferromagnetic ordering in $\text{Sm}_8\text{Ru}_{12}\text{Al}_{53}\text{Si}_{17}$,⁸ structural distortion and magnetic properties influenced by different Cu to In ratio in SmCu_6In_6 ⁹ and topological surface state, which can lead into topological insulator, observed in the Kondo insulator SmB_6 .^{10,11}

During our systematic work on RE-Ni-Al phases, $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ was obtained as large rod shaped single crystals. A few compounds were already reported in the Sm-Ni-Al family: Wang *et al.* reported that the hydrogenation and field cooling lead to an abnormal diamagnetism in SmNiAl and its hydride.¹² Takeshita *et al.* reported the hydrogenation of RENi_4Al ($\text{RE} = \text{La}, \text{Ce}, \text{Pr}, \text{Nd}, \text{Sm}, \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}$ and Tm) at moderate pressure and ambient temperature.¹³ Other compounds reported in the family are $\text{Sm}_3\text{Ni}_6\text{Al}_2$,¹⁴ $\text{Sm}_2\text{Ni}_2\text{Al}$,¹⁵ SmNiAl ,^{16–19} SmNiAl_2 ,²⁰ SmNiAl_4 ²¹ and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$.²²

only for their brief X-ray diffraction (XRD) studies. Our detailed research on different databases including Pearson Crystal Data (PCD)²³ and Inorganic crystal Structure Database (ICSD)²⁴ suggest that $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ was not reported. However, Delsante *et al.* proposed the existence of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ by preliminary powder X-ray diffraction (XRD) and Electron Probe Micro Analysis (EPMA).²⁵ Apart from that, no report on this compound is available in the literature. The compounds in the family $\text{RE}_3\text{Ni}_5\text{Al}_{19}$ with other rare earths (Y, Gd-Lu) were reported in the past.^{25–31} All these compounds crystallize in the orthorhombic $Cmcm$ space group with RE atoms in the trivalent state except Yb, which exists in the mixed valent state.

In our work, the single crystals of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ were obtained from the excess aluminium metal. In recent years we have used metal flux as successful growth technique for new of new intermetallic compounds.^{9,32–52} The crystal structure of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ was refined using single crystal XRD data. $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ is classified as a member in the $\text{RE}_{2n+m}\text{Ni}_{4n+m}\text{Al}_{15n+4m}$ homologous series. The structure relation between $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and other members in the homologous series SmNiAl_4 and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ is explained in detail. After establishing the crystal structure, $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ was successfully synthesized using arc melting technique as well. The phase purity of these compounds was checked by powder XRD and microstructure analysis. The magnetic

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susceptibility measurements suggest trivalent Sm with a weak antiferromagnetic ordering at 18 K.

2. Experimental

2.1 Reagents

The following reagents were used as purchased from Alfa Aesar without further purification: Sm (Ingot, 99.99%), Ni (Powder, 99.99%) and Al (Chunk, 99.99%).

2.2 Synthesis

2.2a Metal flux technique: For the synthesis of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$, samarium metal (0.2 g), nickel (0.1 g) and aluminium (0.6 g) were loaded in an alumina crucible. The crucible was placed in a 13 mm diameter fused silica tube, under an inert (argon) atmosphere inside a glove box, which was flame sealed under a vacuum of 10^{-5} torr to prevent oxidation during heating. The reactants were then heated to 1000°C over 10 h, maintained at that temperature for 5 h to allow proper homogenization, then cooled to 800°C in 2 h and kept at this temperature for 48 h. Finally, the sample was allowed to cool down to 30°C over 48 h. The reaction product was isolated from the excess aluminium flux by heating at 700°C and subsequently centrifuging through a coarse frit. Any unreacted flux was removed by immersion and sonication in hydrochloric acid for 12 h. The final crystalline product was rinsed with water and dried with acetone. This method produced the target compound $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ with ca. 99.99% purity and relatively in a high yield (ca. 70%) on the basis of the initial amount of Sm metal used in the reaction. The rod shaped single crystals were metallic in colour and up to $1\text{ mm} \times 0.1\text{ mm} \times 0.1\text{ mm}$ in size. They were stable in air and no decomposition was observed even after several months. Single crystals of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ were carefully selected for the XRD data collection.

2.2b Arc melting: After establishing the crystal structure, $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ in bulk quantity for the magnetic measurements was prepared by arc melting in Ar atmosphere. The samples were melted repeatedly (four times) in the same atmosphere to ensure homogeneity. The weight losses of the final material were found to be less than 1%. The samples obtained via arc melting method were used for the magnetic studies.

2.3 Elemental analysis

Semi-quantitative microanalyses were performed on the single crystals obtained from the flux techniques

Figure 1. Typical SEM image of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ single crystal grown from the active aluminum flux.

using a Leica 220i scanning electron microscope (SEM) equipped with Bruker 129 eV energy dispersive X-ray analyzer (EDS). Data were acquired with an accelerating voltage of 20 kV and 90 s accumulation time. SEM image of a typical single crystal of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ is shown in figure 1. The EDS analyses were performed on visibly clean surfaces of the single crystals obtained from the flux method indicated that the atomic composition was close to 3:5:19, which is in good agreement with the composition obtained from the single crystal X-ray data refinement.

2.4 Powder XRD

Phase identity and purity of the $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ sample was determined by powder XRD experiments that were carried out with a Bruker D8 Discover diffractometer using $\text{Cu-K}\alpha$ radiation ($\lambda = 1.5406\text{ \AA}$). The experimental powder pattern of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ was found to be in good agreement with the pattern calculated from the single-crystal X-ray structure refinement with a small amount of Ni_2Al_3 phase (figure 2).

2.5 Single crystal XRD

XRD data was collected on a selected single crystal of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ at room temperature using a Bruker Smart

Figure 2. The comparison between the PXRD patterns for the compound $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ obtained from the arc melting synthesis technique and simulated from the single crystal XRD data refinement (star marks corresponds to Ni_2Al_3 phase).

CCD diffractometer equipped with a normal focus, 2.4 kW sealed tube X-ray source with graphite monochromatic Mo-K α radiation ($\lambda = 0.71073$ Å) operating at 50 kV and 30 mA, with ω scan mode. A crystal of suitable size (0.05 x 0.05 x 0.1 mm³) was cut from a rod shaped crystal and mounted on a thin glass (~0.1 mm) fibre with commercially available super glue. A full sphere of 90 frames was acquired up to 66.47° in 2θ . The individual frames were measured with steps of 0.50° and an exposure time of 30 s per frame. The programme SAINT⁵³ was used for integration of diffraction profiles and absorption correction was made with SADABS programme.⁵⁴ The systematic absences led to the centrosymmetric space group *Cmcm*.

2.6 Structure refinement

The structure of Sm₃Ni₅Al₁₉ was solved by SHELXS 97⁵⁵ and refined by a full matrix least-squares method using SHELXL⁵⁶ with anisotropic atomic displacement parameters for all atoms. As a check for the correct composition, the occupancy parameters were refined in a separate series of least-squares cycles. Single crystals of Sm₃Ni₅Al₁₉ from different syntheses batches were used for data collection. The lattice parameters of the

Gd₃Ni₅Al₁₉ structure were taken at the initial step and the refinement resulted in fifteen crystallographic positions in the Sm₃Ni₅Al₁₉ structure; which are two samarium atoms, three nickel atoms and ten aluminium atoms. Among them, samarium atoms occupy 8*f* and 4*c* sites of point symmetry *m.* and *m2m*, respectively, two nickel atoms occupy 8*f* sites (point symmetry *m.*), other nickel atom occupies 4*c* site (point symmetry *m2m*), nine aluminium atoms occupy the 8*f* sites (point symmetry *m.*) and one aluminium atom occupies 4*c* site (point symmetry *m2m*).

As a check for the correct composition, the occupancy parameters were refined in a separate series of least-squares cycles. All bond lengths are within the acceptable range compared to the theoretical values. Packing diagrams were generated with Diamond.⁵⁷ The data collection and structure refinement for Sm₃Ni₅Al₁₉ are listed in table 1. The standard atomic positions and isotropic atomic displacement parameters of this compound are listed in table 2. The anisotropic displacement parameters and important bond lengths are listed in tables 3 and 4, respectively. Further information on the structure refinements is available from Cambridge Crystallographic Data Centre by quoting the Registry No. CCDC 993911.

Table 1. Crystal data and structure refinement for Sm₃Ni₅Al₁₉.

Empirical formula	Sm ₃ Ni ₅ Al ₁₉
Formula weight	1257.22
Temperature	293(2) K
Wavelength	0.71073 Å
Crystal system	Orthorhombic
Space group	<i>Cmcm</i>
Unit cell dimensions	$a = 4.0974(1)$ Å $b = 16.0172(6)$ Å $c = 27.0774(10)$ Å
Volume	1777.06(10) Å ³
Z	4
Density (calculated)	4.7 g/cm ³
Absorption coefficient	15.85 mm ⁻¹
F(000)	2291
Crystal size	0.05 x 0.05 x 0.10 mm ³
θ range for data collection	2.54 to 33.23°
Index ranges	$-6 \leq h \leq 6$, $-24 \leq k \leq 23$, $-32 \leq l \leq 41$
Reflections collected	16247
Independent reflections	1942 [$R_{\text{int}} = 0.0368$]
Completeness to $\theta = 32.17^\circ$	100%
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	1942 / 0 / 86
Goodness-of-fit	1.319
Final R indices [$>2\sigma(I)$]	$R_{\text{obs}} = 0.018$, $wR_{\text{obs}} = 0.034$
R indices [all data]	$R_{\text{all}} = 0.019$, $wR_{\text{all}} = 0.049$
Extinction coefficient	0.004
Largest diff. peak and hole	1.358 and $-2.129 \text{ e} \cdot \text{Å}^{-3}$

$^a R = \sum ||F_o| - |F_c|| / \sum |F_o|$, $wR = \{ \sum [w(|F_o|^2 - |F_c|^2)^2] / \sum [w(|F_o|^4)] \}^{1/2}$ and calc. $w = 1/[\sigma^2(F_o^2) + (0.0613P)^2 + 42.3592P]$ where $P = (F_o^2 + 2F_c^2)/3$

Table 2. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ at 296(2) K with estimated standard deviations in parentheses.

Label	Wyckoff site	x	y	z	Occupancy	U_{eq}^*
Sm1	8f	0	1659(1)	1352(1)	1	5(1)
Sm2	4c	0	3863(2)	2500	1	6(1)
Ni1	8f	0	532(4)	5835(2)	1	7(1)
Ni2	8f	0	3405(3)	5418(2)	1	5(1)
Ni3	4c	0	451(5)	2500	1	6(1)
Al1	8f	0	355(9)	6737(5)	1	9(1)
Al2	8f	0	678(9)	304(5)	1	8(1)
Al3	8f	0	2084(9)	5883(5)	1	7(1)
Al4	8f	0	2242(9)	6932(5)	1	9(1)
Al5	8f	0	2344(9)	284(5)	1	7(1)
Al6	8f	0	3701(9)	1348(5)	1	9(1)
Al7	8f	0	4051(9)	370(5)	1	9(1)
Al8	8f	0	5362(9)	1001(5)	1	8(1)
Al9	8f	0	5779(9)	2018(5)	1	8(1)
Al10	4f	0	1930(12)	2500	1	9(1)

* U_{eq} is defined as one third of the trace of the orthogonalized U_{ij} tensor.

2.7 Magnetic measurements

Magnetic measurements were carried out using a Quantum Design MPMS-SQUID magnetometer. Measurements were performed on polycrystalline sample, which were ground and screened by powder XRD to verify phase identity and purity. Temperature dependent data were collected in field cooled mode (FC) between 2 and 300 K in an applied field (H) of 1000 Oe. Magnetization data were also collected for $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ at 2 K and 300 K with field sweeping from -60000 Oe to 60000 Oe.

3. Results and Discussion

3.1 Crystal chemistry

The reports on the compounds $\text{RE}_3\text{Ni}_5\text{Al}_{19}$ with rare earths $\text{RE} = \text{Y}, \text{Sm}, \text{Gd-Lu}$ suggest that this composition is favoured by small rare earths.^{25–31} The absence of Eu in this series is probably due to the large size of divalent Eu atom, which suggests that in addition to size, the valency of rare earth also plays a crucial role in the formation of these compounds. The lattice parameters of all reported compounds in the $\text{RE}_3\text{Ni}_5\text{Al}_{19}$ are listed

Table 3. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ at 293(2) K with estimated standard deviations in parentheses.

Label	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Sm1	6(1)	5(1)	5(1)	0	0	0
Sm2	7(1)	6(1)	5(1)	0	0	0
Ni1	8(2)	6(1)	8(1)	0	0	0
Ni2	6(2)	5(2)	5(2)	0	0	0
Ni3	7(3)	6(3)	5(3)	0	0	0
Al1	10(5)	9(1)	7(6)	0	0	1(1)
Al2	9(5)	6(1)	10(1)	0	0	0
Al3	9(5)	6(1)	7(6)	0	0	1(1)
Al4	9(5)	11(6)	8(6)	0	0	2(1)
Al5	7(5)	8(6)	7(6)	0	0	2(1)
Al6	10(6)	8(6)	8(6)	0	0	0
Al7	11(5)	10(6)	6(1)	0	0	$-2(1)$
Al8	8(5)	7(6)	9(6)	0	0	0
Al9	7(5)	13(6)	5(3)	0	0	1(1)
Al10	12(8)	7(1)	7(1)	0	0	0

The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11} + \dots + 2hka^*b^*U_{12}]$.

Table 4. Selected bond lengths [Å] for Sm₃Ni₅Al₁₉ at 293(2) K with estimated standard deviations in parentheses.

Label	Distances	Label	Distances
Sm1—Al1	3.3909(15)	Ni2—Al7	2.3751(15)
Sm1—Al2	3.2438(14)	Ni2—Al8	2.5277(15)
Sm1—Al3	3.1403(11)	Ni3—Al1	2.4360(15)
Sm1—Al4	3.1239(11)	Ni3—Al9	2.4847(8)
Sm1—Al5	3.0934(14)	Ni3—Al10	2.370(2)
Sm1—Al8	3.068(1)	Al1—Al6	2.7551(13)
Sm1—Al9	3.0723(11)	Al1—Al9	2.8421(14)
Sm1—Al10	3.1379(4)	Al2—Al2	2.727(3)
Sm1—Ni2	3.2555(2)	Al2—Al5	2.6697(19)
Sm2—Al1	3.1671(11)	Al2—Al7	2.7794(14)
Sm2—Al4	3.1139(11)	Al2—Al8	2.8320(14)
Sm2—Al6	3.1298(14)	Al3—Al4	2.853(2)
Sm2—Al10	3.096(2)	Al3—Al5	2.7683(13)
Sm2—Ni3	3.2655(7)	Al3—Al6	2.7141(13)
Ni1—Al1	2.4568(15)	Al4—Al6	2.9979(15)
Ni1—Al2	2.4143(15)	Al4—Al10	2.8833(14)
Ni1—Al3	2.4893(15)	Al5—Al5	2.6102(17)
Ni1—Al6	2.7619(10)	Al5—Al7	2.745(2)
Ni1—Al7	2.4956(9)	Al6—Al7	2.705(2)
Ni1—Al8	2.5402(9)	Al6—Al8	2.821(2)
Ni2—Al2	2.5394(9)	Al7—Al8	2.707(2)
Ni2—Al3	2.4613(15)	Al8—Al9	2.832(2)
Ni2—Al5	2.5511(15)	Al9—Al9	2.608(3)
Ni2—Al5	2.4025(8)		

in table 5 and cell volume against the rare earths is plotted in figure 3. The decrease in cell volume from Sm to Lu corroborate with the lanthanide contraction and the deviation from the curve with a huge hump for Yb sample indicate the probable mixed valent behaviour of Yb, which is in fact reported by Bauer *et al.*³¹

Sm₃Ni₅Al₁₉ is the compound with largest volume in this family, which crystallizes in the orthorhombic Gd₃Ni₅Al₁₉ type structure (space group *Cmcm*) and can be considered as a member of the homologous series Sm_{2n+m}Ni_{4n+m}Al_{15n+4m} (*n* = 1 and *m* = 1).²⁷ The crystal structure of Sm₃Ni₅Al₁₉ along the *a*-axis is

Table 5. The lattice parameters (*a*, *b* and *c*) of all compounds reported in the RE₃Ni₅Al₁₉ (RE = Sm, Y, Gd-Lu) family.

Compound name	<i>a</i> (Å)	<i>b</i> (Å)	<i>c</i> (Å)	reference
Y ₃ Ni ₅ Al ₁₉	4.08	16.04	27.29	²⁶
Sm ₃ Ni ₅ Al ₁₉	4.09	16.01	27.07	²⁵
Sm ₃ Ni ₅ Al ₁₉	4.0974(1)	16.0172(6)	27.0774(10)	[This work]
Gd ₃ Ni ₅ Al ₁₉	4.08	15.99	27.09	²⁷
Tb ₃ Ni ₅ Al ₁₉	4.035	15.91	27.08	²⁸
Dy ₃ Ni ₅ Al ₁₉	4.021	15.86	27.01	²⁸
Ho ₃ Ni ₅ Al ₁₉	3.991	15.75	26.92	²⁹
Er ₃ Ni ₅ Al ₁₉	3.960	15.63	26.81	²⁹
Tm ₃ Ni ₅ Al ₁₉	3.92	15.46	26.59	³⁰
Yb ₃ Ni ₅ Al ₁₉	4.06	15.90	26.98	³¹
Lu ₃ Ni ₅ Al ₁₉	3.92	15.40	26.54	³⁰

Figure 3. The cell volume ($\text{\AA}^3$) of the compounds reported in the $\text{RE}_3\text{Ni}_5\text{Al}_{19}$ ($\text{RE} = \text{Sm, Gd-Lu}$) plotted against the rare earth atoms.

shown in figure 4. The structure can be explained as $n\text{Sm}_2\text{Ni}_4\text{Al}_{15}$ slabs intergrown with $m\text{SmNiAl}_4$ slabs along the b -axis. SmNiAl_4 slab in $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ is a Sm-centered pentagonal, while $\text{Sm}_2\text{Ni}_4\text{Al}_{15}$ slab contains two Ni-centred trigonal prisms, one Al-centred tetragonal prism and two Sm centred pentagonal prisms. The crystal structure of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ can be compared with the other two members in the homologous series SmNiAl_4 ($n = 0$ and $m = 1$) and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ ($n = 1$ and $m = 2$) as shown in figure 5. SmNiAl_4 ²¹ is an orthorhombic system crystallizes in the YNiAl_4 ⁵⁸ type structure with $Cmcm$ space group, while, $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ ²² is a monoclinic system that crystallizes in the $\text{Y}_4\text{Ni}_6\text{Al}_{23}$ ^{59,60} structure type with space group $C2/m$. The compounds where m is odd ($m = 1, 3$, etc.) adopt orthorhombic structure and where m is even ($m = 0, 2$, etc.) adopt the monoclinic structure. SmNiAl_4 contains two SmNiAl_4 slabs rotated by 180° with respect to each other. On the other hand $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ contains two SmNiAl_4 and one $\text{Sm}_2\text{Ni}_4\text{Al}_{15}$ slabs intergrown similar in $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$. The bond angles in all three compounds are very close. As an example, the Al-Ni-Al bond

Figure 4. The crystal structure of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ is shown along the a -axis. The two slabs SmNiAl_4 and $\text{Sm}_2\text{Ni}_4\text{Al}_{15}$ are shaded with yellow and green colors, respectively, intergrown along the b -axis.

angle in SmNiAl_4 slabs of SmNiAl_4 , $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ are 113.28° , 110.62° and 114.87° , respectively, and Al-Sm-Al bond angles in trigonal prisms are 46.66° , 49.76° and 49.46° for SmNiAl_4 , $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$, respectively.

The local coordination environments of all Sm and Ni atoms in the compounds SmNiAl_4 , $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ are presented in figure 6. Sm and Ni coordination sphere of all three compounds are very similar. Noticeably the coordination sphere of Ni2 and Ni3 are interchanged in $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ if standard atomic coordinates are taken from the literature. The coordination environments of all Sm atoms are surrounded with 13 aluminium and 2 nickel atoms. The coordination environment of Ni, Ni3 and Ni2 in SmNiAl_4 , $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$, respectively, are similar to seven Al and two Sm neighbouring atoms. The coordination environment of Ni1, Ni2 in $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and Ni1, Ni3 in $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ are similar surrounded with ten Al atoms and Ni2 in $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and Ni3 in $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ are similarly surrounded with eight Al and two Sm atoms. Such similarity in the coordination environment is also observed for the Al atoms in all three structures.

The shortest and largest bond distances of Sm-Al in $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ 3.068(1) $\text{\AA}$ and 3.391(2) $\text{\AA}$, respectively, are comparable with the distances in SmNiAl_4 (3.022 $\text{\AA}$ and 3.358 $\text{\AA}$) and in $\text{Sm}_4\text{Ni}_6\text{Al}_{24}$ (3.06 $\text{\AA}$ and 3.381 $\text{\AA}$).^{21,22} Similarly the shortest Sm-Ni bond distance of 3.2555 $\text{\AA}$ in $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ is also similar in the compounds SmNiAl_4 and $\text{Sm}_4\text{Ni}_6\text{Al}_{24}$ with the values 3.209 $\text{\AA}$ and 3.268 $\text{\AA}$, respectively.^{21,22}

3.2 Magnetic properties

The temperature dependent magnetic susceptibility and modified inverse molar magnetic susceptibility of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ at an applied field of 1000 Oe is shown in figure 7. The temperature dependent molar susceptibility (χ_m) of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ obeys modified Curie-Weiss law, $\chi = C/(T-\theta) + \chi_0$ above 150 K with a complicated magnetic ordering below 18 K. A Curie-Weiss fit in the temperature range of 150–300 K gave the magnetic moment $2.25\mu_B$ per formula.⁶¹

The field dependence of the magnetization $M(H)$ for $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ ground samples were measured at 2 K and 300 K can be seen in figure 8. The data measured at 300 K exhibit linear behaviour and no signs of saturation up to our highest attainable field of 60000 Oe. The magnetization curve taken at 2 K (within the magnetic ordering) shows a slight field dependent response up to ~ 30000 Oe, continues to rise slowly up to the highest obtainable field (60000 Oe).

Figure 5. The crystal structures of (a) SmNiAl_4 , (b) $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and (c) $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$. The pentagonal, tetragonal and trigonal prisms are shaded with orange, green and brown colors. The unit cell is outlined in solid blue line.

Figure 6. The coordination sphere of Sm and Ni atoms in (a) SmNiAl_4 , (b) $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and (c) $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$.

Figure 7. Temperature dependence of the magnetic susceptibility (χ_m) and modified inverse magnetic susceptibility ($1/\chi_m$) of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$. The inset shows antiferromagnetic like ordering at 18 K.

Figure 8. Magnetization as a function of applied magnetic field at 2 K and 300 K for a polycrystalline sample of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$.

4. Conclusions

Well-shaped single crystals of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ were obtained from the metal flux technique and the crystal structure of $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ was studied using single crystal X-ray diffraction. The flux method proves to be an excellent tool for growing intermetallic compounds with complex crystal structures. The compound $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ is a member in the $\text{Sm}_{2n+m}\text{Ni}_{4n+m}\text{Al}_{15n+4m}$ series with $n = 1$ and $m = 1$. Although a few compounds other than SmNiAl_4 , $\text{Sm}_3\text{Ni}_5\text{Al}_{19}$ and $\text{Sm}_4\text{Ni}_6\text{Al}_{23}$ were hypothetically predicted in this series, none of them are experimentally realized. Our work will be directed in the synthesis of those compounds using metal flux technique.

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