Electronic Supporting Information

Design, Synthesis and Isolation of a New 1,2,5-Selenadiazolidyl and Structural and Magnetic Characterization of its Alkali-Metal Salts

Nikolay A. Semenov,*[a] Ekaterina A. Radiush,[a] Elena A. Chulanova,[a,b] Alexandra M. Z. Slawin,[c] J. Derek Woollins,[d] Evgeny M. Kadilenko,[b,e] Irina Yu. Bagryanskaya,[a] Irina G. Irtegova,[a] Artem S. Bogomyakov,[b,f] Leonid A. Shundrin,[a,b] Nina P. Gritsan,[e,g] and Andrey V. Zibarev [a,g]

[a] *Institute of Organic Chemistry, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia. E-mail: klaus@nioch.nsc.ru*

[b] *Department of Natural Sciences, Novosibirsk State University, 630090 Novosibirsk, Russia*

[c] *School of Chemistry, University of St. Andrews, St Andrews, Fife KY16 9ST, United Kingdom*

[d] *Department of Chemistry, Khalifa University, Abu Dhabi, United Arab Emirates*

[e] *Institute of Chemical Kinetics and Combustion, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia*

[f] *International Tomography Center, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia*

[g] *Department of Physics, Novosibirsk State University, 630090 Novosibirsk, Russia*

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1. Single-crystal X-ray diffraction

Table S1. Crystallographic data for compounds

Compound	**1**	**2**	**2**·thf	**3**·thf
Empirical formula	C_6N_6Se	$C_{18}H_{24}KN_6O_6Se$	$C_{22}H_{32}KN_6O_7Se$	$C_{22}H_{32}NaN_6O_7Se$
Formula mass	235.07	538.49	610.59	594.48
Temperature [K]	93	296	296	200
Wavelenght [Å]	0.71075	0.71073	0.71073	0.71073
Crystal system	Orthorhombic	Monoclinic	Monoclinic	Monoclinic
Space group	Pbca	P 2_1/c	P 2_1/n	P 2_1/n
a [Å]	18.847(5)	12.0108(5)	12.3642(7)	8.7796(7)
b [Å]	10.756(3)	19.0336(8)	9.2915(5)	14.7106(10)
c [Å]	7.0728(18)	10.8353(4)	24.4420(15)	22.4551(19)
α [˚]	90.0000	90.000	90.000	90.000
β [˚]	90.0000	105.018(2)	92.661(3)	90.426(3)
γ [˚]	90.0000	90.000	90.000	90.000
Cell Volume (Å^3)	1433.8(7)	2392.44(17)	2804.9(3)	2900.1(4)
Z	8	4	4	4
Denisity (calcd). [Mgm^{-3}]	2.178	1.495	1.446	1.362
Absorption coefficient [mm^{-1}]	5.188	1.788	1.537	1.358
F (000)	896	1100	1260	1228
Crystal size [mm]	0.10 × 0.10 × 0.02	0.90 × 0.50 × 0.40	0.38 × 0.12 × 0.03	0.50 × 0.12 × 0.10
Θ range [˚]	2.16-25.35	2.22-30.05	2.35-27.56	2.28-27.54
	–22 ≤ h ≤ 22	–16 ≤ h ≤ 16	–14 ≤ h ≤ 15	–11 ≤ h ≤ 11
Index limit	–12 ≤ k ≤ 12	–26 ≤ k ≤ 25	–12 ≤ k ≤ 12	–17 ≤ k ≤ 19
	–8 ≤ l ≤ 8	–15 ≤ l ≤ 12	–31 ≤ l ≤ 31	–29 ≤ l ≤ 29
Reflections collected	8956	46821	32646	57566
Independent reflections	1314	6549	6388	6694
	[R_{int} = 0.0443]	[R_{int} = 0.0447]	[R_{int} = 0.0518]	[R_{int} = 0.0523]
Completeness to Θ [%]	99.5	99.9 (2Θ≤50°)	98.8	100.0
Data/restraints/parameters	1314 / 0 / 118	6549 / 0 / 289	6391/0/334	6694/6/344
Goodness of fit on F^2	0.958	1.026	1.029	1.043
R indices [I>2σ(I)]	R_1 = 0.0210	R_1 = 0.0466	R_1 = 0.0486	R_1 = 0.0366
	wR_2 = 0.0481	wR_2 = 0.1053	wR_2 = 0.1068	wR_2 = 0.0801
R indices (all data)	R_1 = 0.0265	R_1 = 0.0917	R_1 = 0.1223	R_1 = 0.0681
	wR_2 = 0.0489	wR_2 = 0.1262	wR_2 = 0.1297	wR_2 = 0.0925
CCDC	**1917743**	**1917744**	**1917745**	**1917746**

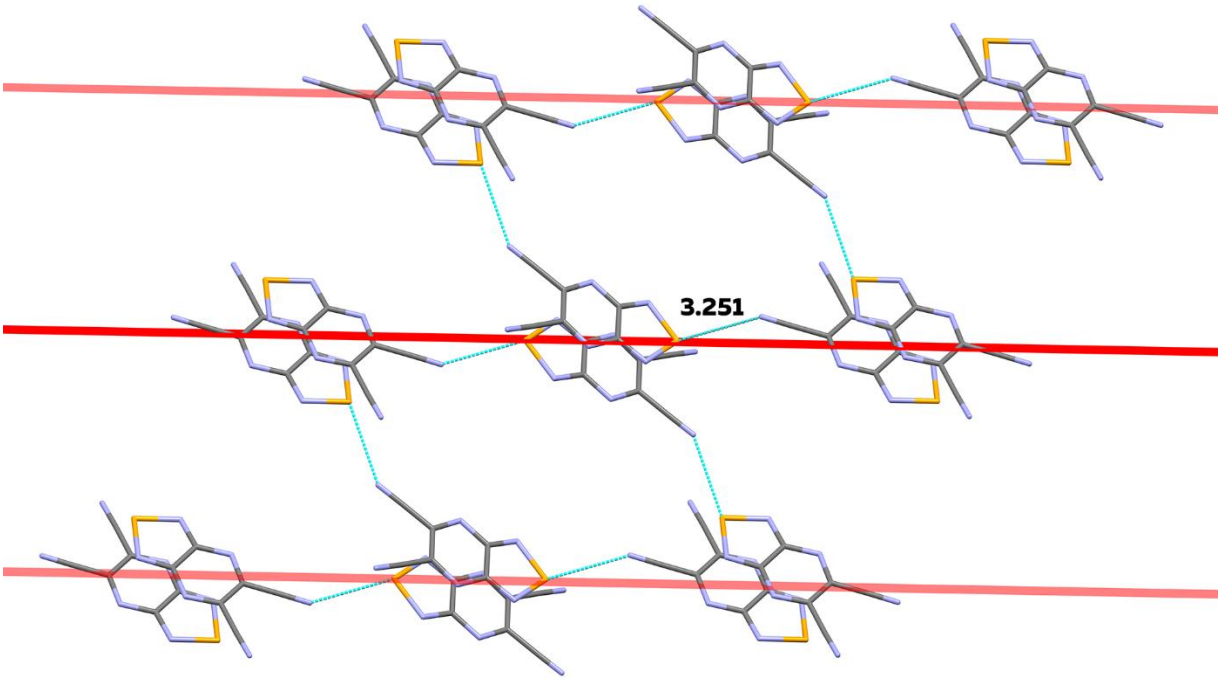

Figure S1. Fragment of crystal packing of **2** viewed along the crystallographic axis *a* and showing layers of RA dimers connected by shortened contacts Se…N (light blue lines). [K(18-crown-6)$^{+}$] cations are omitted for clarity.

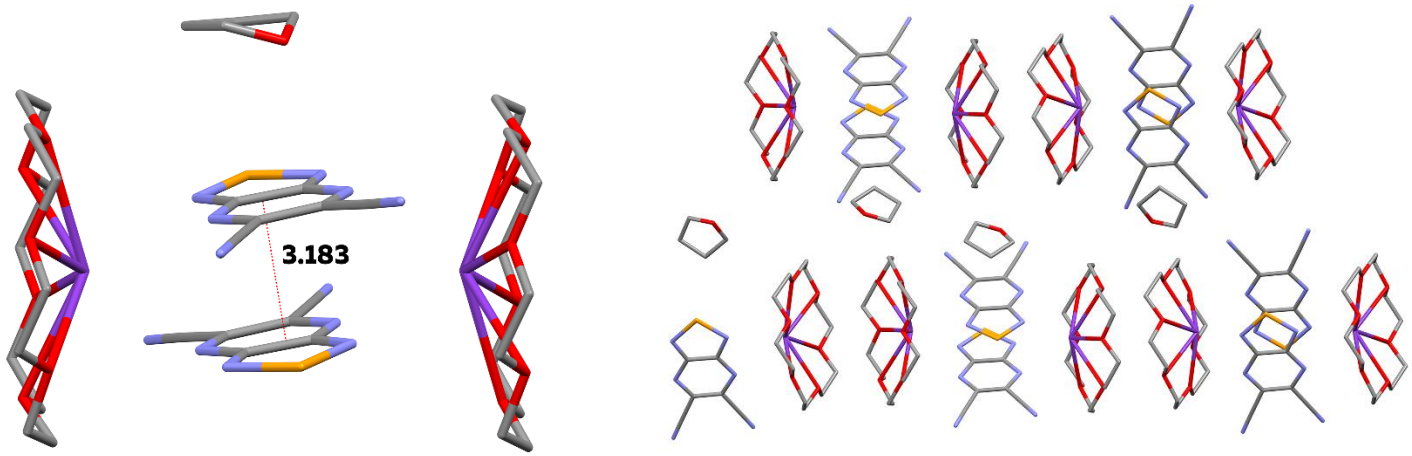

Figure S2. Left: XRD molecular structure of **2**·thf, H atoms are omitted for clarity. Right: Fragment of crystal packing viewed along the crystallographic axis *b* and showing layers of RA dimers. [K(18-crown-6)$^{+}$]$_2$[**1**]$^{\bullet -}{}_2$ are separated by layers of thf molecules.

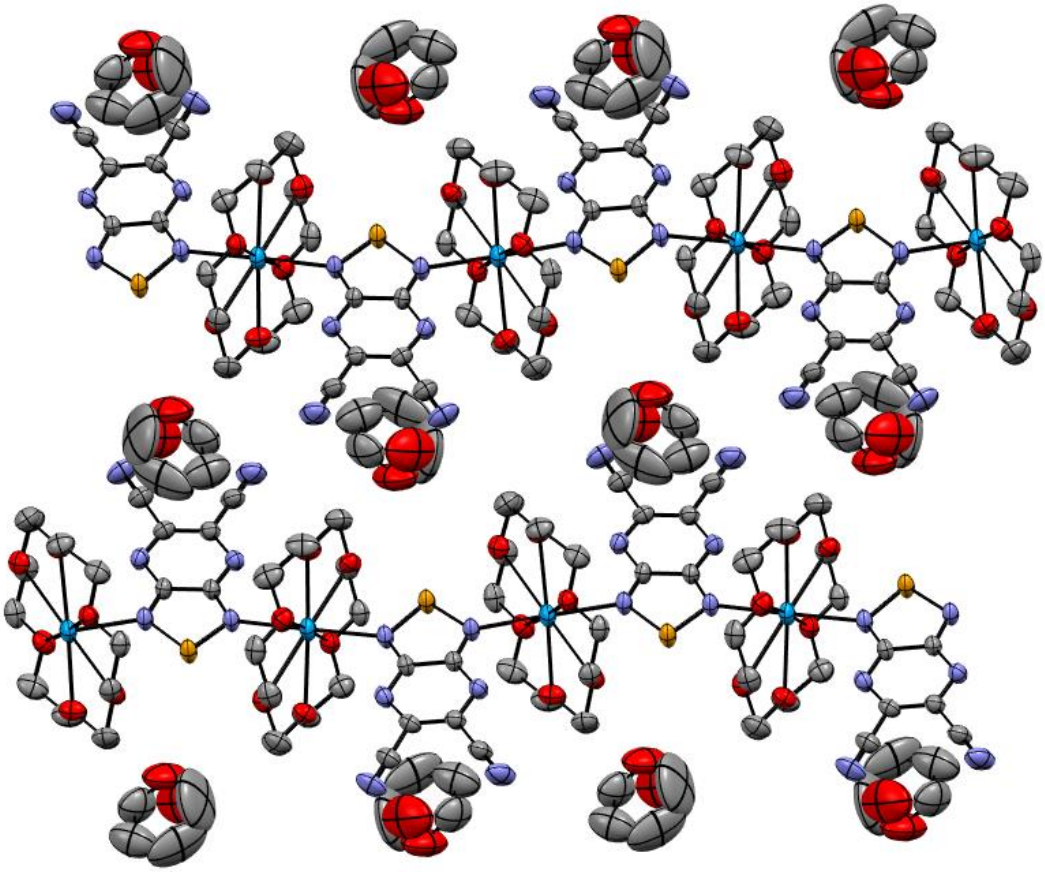

Figure S3. Fragment of crystal packing of **3**·thf featuring two parallel infinite chains of alternating cations and RAs with thf molecules filling the space between the chains (H atoms are omitted for clarity).

2. Powder X-ray diffraction

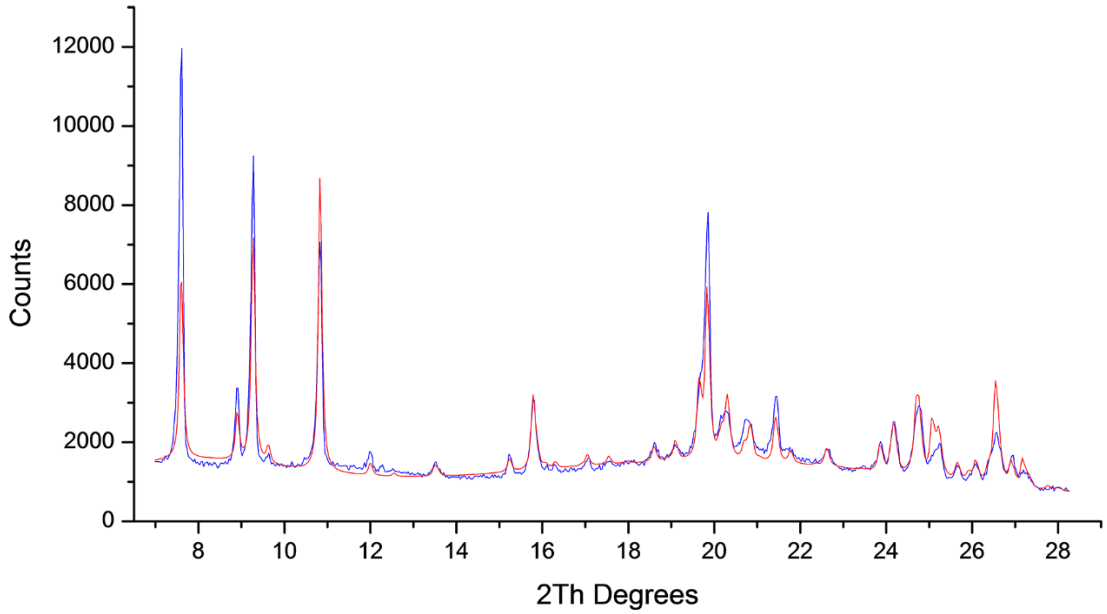

Figure S4. Powder XRD pattern for the non-crystalline sample of salt **2** (blue) and calculated pattern for the single-crystal XRD data for **2** (red).

3. Cyclic voltammetry

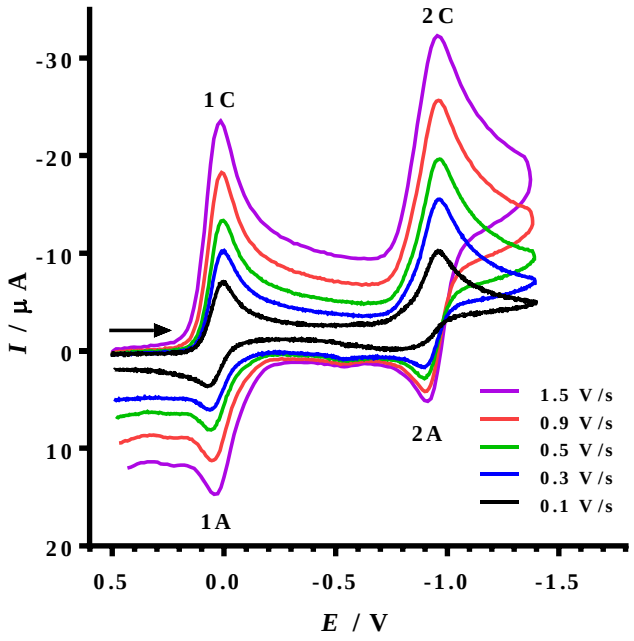

Figure S5. Cyclic voltammograms of reduction of compound **1** in MeCN solution with the potential sweep rates varied from 0.1 to 1.5 $V \cdot s^{-1}$ (depicted with colours). Black arrow indicates the direction of potential sweep.

4. EPR spectroscopy

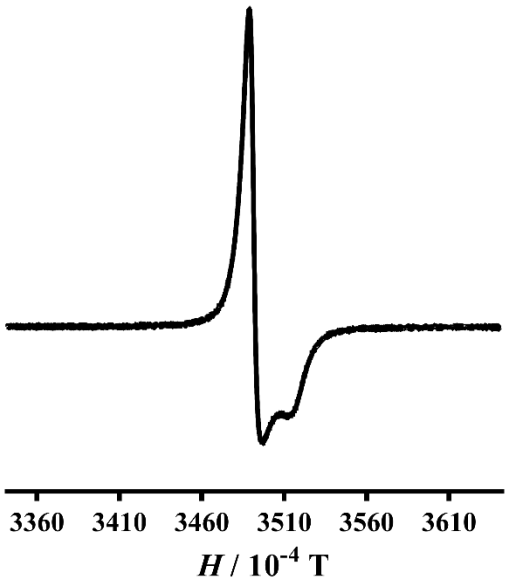

Figure S6. Weak signal in the solid-state EPR spectrum of salt **2**.

5. SQUID magnetometry

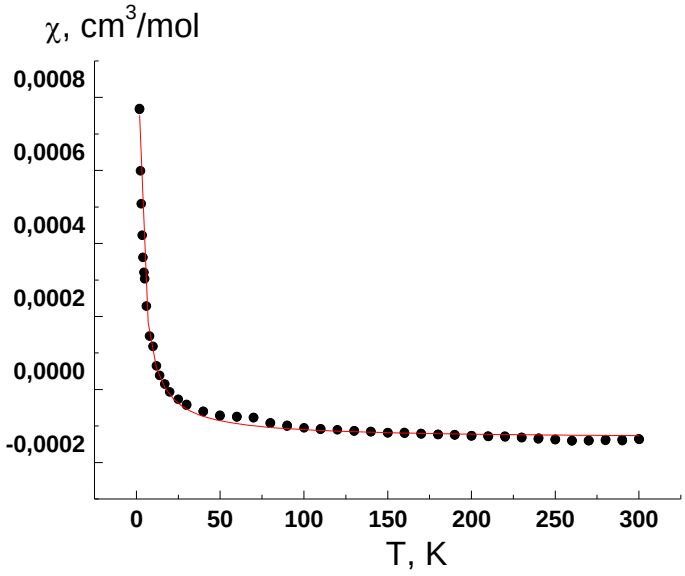

Figure S7. Temperature dependence of molar magnetic susceptibility χ(T) of salt **2** in the temperature range 2-300 K. Solid line corresponds to the best fit, using an equation $\chi = \chi_D + C/(T - \Theta)$. The best fit parameter values of χ_D, C and Θ are $-130 \cdot 10^{-6}$ cm^3 mol^{-1}, 0.0025 K·cm3/mol and –0.82 K, respectively. So, diamagnetic susceptibility $\chi_D = -130 \cdot 10^{-6}$ cm^3 mol^{-1}, and Curie constant value allows to estimate admixture of paramagnetic centers with spin S = 1/2 as 0.0025/0.375 = 0.0067 = 0.67%.

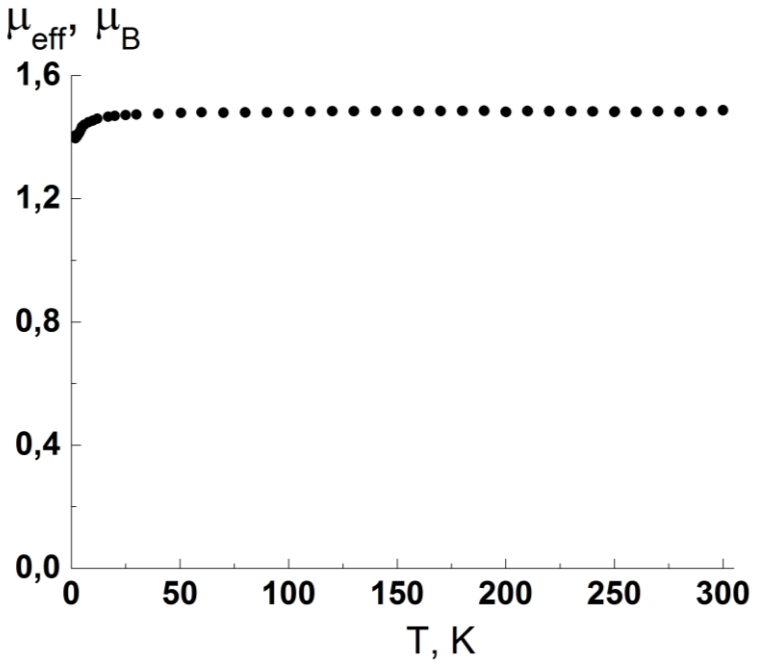

Figure S8. Temperature dependence of the effective magnetic moment μ_{eff}(T) of salt **3** in the temperature range 2-300 K.

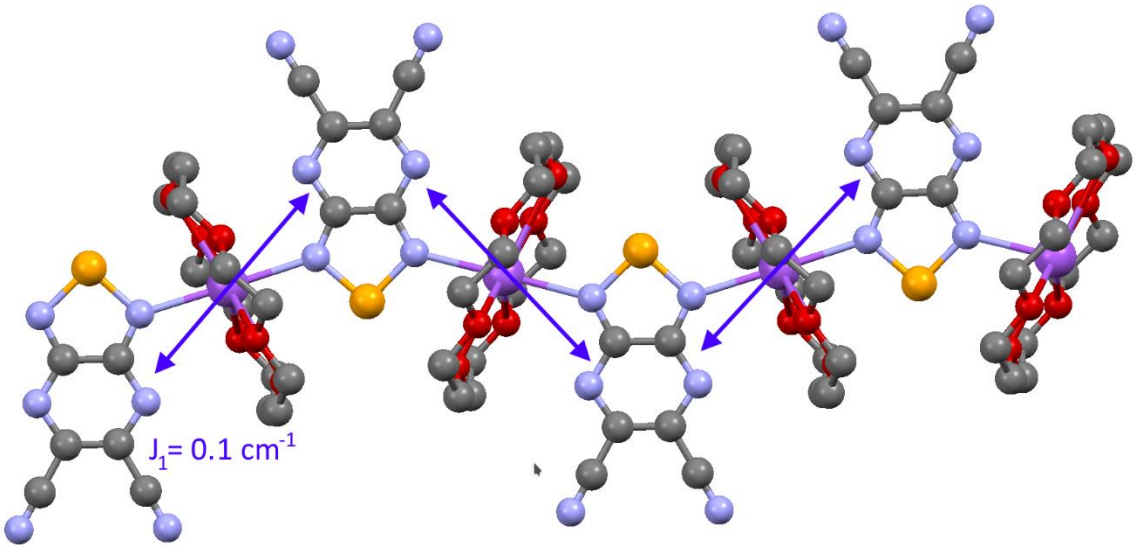

Figure S9. Fragment of the XRD structure of salt **3**·thf (alternating chain of RAs and cations) and intrachain pair exchange interactions indicated by arrows.