Table S1. Chemical data (in *weight % of oxides* and *atoms per formula unit = apfu*) of donwilhelmsite. The empirical formula calculated on the basis of 7 cations is $Ca_{1.02}Al_{3.92}Si_{2.06}O_{11}$, and the simplified formula is $CaAl_4 Si_2 O_{11}$.

	Al_2O_3 wt%	SiO_2 wt%	CaO wt%	Al 7 apfu	Si 7apfu	Ca 7 apfu
sp-01	52.3	32.1	15.7	3.9	2.0	1.1
sp-02	54.0	32.6	14.1	4.0	2.1	1.0
sp-03	46.8	35.2	17.7	3.5	2.3	1.2
sp-04	52.9	34.1	13.9	3.9	2.1	0.9
sp-05	54.0	32.2	14.3	4.0	2.0	1.0
sp-06	51.5	33.1	15.6	3.8	2.1	1.1
sp-07	52.8	32.7	14.9	3.9	2.1	1.0
sp-08	53.3	31.0	15.6	40	2.0	1.1
sp-09	53.2	31.6	15.3	4.0	2.0	1.0
sp-10	56.0	31.4	13.3	4.1	2.0	0.9
average	52.7	32.6	15.0	3.9	2.1	1.0
std. dev.	2.4	1.3	1.2	0.2	0.1	0.1
min.	46.8	31.0	13.3	3.5	2.0	0.9
max.	56.0	35.2	17.7	4.1	2.3	1.2

Table S2. Comparison of unit-cell parameters of the hexagonal crystal system. Diffraction data of the here described new mineral were obtained by precession electron diffraction tomography (PEDT). X-ray diffraction data in Gautorn et al. (1999) and Akaogi et al. (2009) were obtained on single crystals and powder samples, respectively. The synchrotron XRD patterns of micrometer-sized high aluminum silica (HAS) zagamiite grains are similar to those of powder-like samples (Ma et al. 2017). The cell parameters of these hexagonal crystals are given as a and c in [Å] and the cell volume $V = a^2c \sin(60°)$ in [Å^3]. The numbers in parentheses () represent the estimated standard deviations of the last digit. *Unit cell parameters from electron diffraction tomography (EDT) are known to have deviations and lower accuracies (Kolb et al., 2008). Z is the number of formula units per unit cell. The calculated density of donwilhelmsite is 3.903 [g/cm^{-3}].

Composition	Space group	a [Å]	c [Å]	V [Å^3]	Z	Reference
Donwilhelmsite **CaAl$_4$Si$_2$O$_{11}$**	$P6_3/mmc$	5.42 (1)	12.70 (3)*	323(4)	2	This work
synthetic **CaAl$_4$Si$_2$O$_{11}$**	$P6_3/mmc$	5.4223 (4)	12.7041 (6)	323.28 (5)	2	Gautron et al. 1999
synthetic **CaAl$_4$Si$_2$O$_{11}$**	$P6_3/mmc$	5.4239 (2)	12.6805 (5)	323.06 (3)	2	Akaogiet al. 2009
Zagamiite **CaAl$_2$Si$_{3.5}$O$_{11}$**	$P6_3/mmc$	5.403 (2)	12.77 (3)			Ma et al. 2017