Influences of As(V), Sb(III), and Hg(II) Ions on the Nucleation and Growth of Akaganeite

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SUPPORTING INFORMATION

XRD Analysis. X-ray diffraction (XRD) analysis was performed using a Panalytical Empyream Serie 2 X-ray diffractometer. The powder pattern was obtained using a Co-Ka radiation. The equipment operated with a scan range from 5° to 80°, a scan step size of 0.013° and a time per step of 59.670 s. The diffractograms were analyzed using a program based on the Rietveld methodology and Fourier transform called MAUD⁴² through which the values of scale factor, sample displacement, incident intensity, unit cell parameters, six-order polynomial background, and the average crystallite size were refined. The average crystallite size and texture were assumed to be anisotropic. The Rietveld analysis was performed using the CIF file of akaganeite with the monoclinic symmetry I2/m (COD code 9001319).⁴³

XRF Analysis. X-ray fluorescence (XRF) analysis was performed using a Thermo Scientific ARL OPTIM'X by wavelength dispersive x-ray fluorescence (WDXRF).

TEM and STEM. High-resolution transmission electron microscopy (TEM) and Scanning transmission electron microscopy analysis were performed with a FEI Tecnai G2 F20 Super-Twin transmission electron microscope operating at 200 kV. The preparation of the sample consisted of the dilution in ethanol of a small amount of the same, followed by the immersion of the mixture in an ultrasonic bath,

in order to disperse the sample in the solvent. Finally, the analysis was carried out by depositing a drop of the dispersion on a Cu grid coated with a carbon film.

Raman spectroscopy. Raman spectra were obtained with a Horiba Confocal Raman Spectrometer Yvon, High Resolution Labram HR Model, equipped with a He/Ne laser operating at 632.81 nm and a Nicon confocal microscope model BX-41 provided with lenses of 50X. The helium - neon laser was focused to a spot size of 1-2 μ m² at the samples. The scattered light was dispersed by a monochromator with 600 lines/mm grating and detected by a CCD system with resolution of 1024 x 256 pixels. The spectra range was from 100 to 4000 cm⁻¹, with a step size of 1.1 cm⁻¹. Moreover, the acquisition time was of 30 s with an accumulation of 2x5. The hold and slit were of 800 µm and 600 µm, respectively. Finally, with the objective of avoiding any transformation of the samples, the influence of the laser power was previously tested. In this way, it was concluded that the use of the D0.6 filter, which reduced the power to 0.85 mW, was the ideal for treating the samples. The Raman experiments were performed to the samples and its respective replicates, with the aim of confirming the reproducibility of all syntheses. In addition, to verify sample homogeneity the spectra collection was carried out repeatedly at different colored regions of each synthesis and each replicate, with the objective of confirming the existence or absence of other phases. Pure akaganeite appeared homogeneously red colored and the rest of the samples showed regions of red, white and dark colors. However, all repeated spectra showed to be identical and independent of color variation. All Raman spectral data were baseline-corrected and curve-fitted to obtain quantitative information of the band parameters like wavenumbers, widths, and relative intensities. All spectra were adjusted with Lorentzians lines as proposed in previous studies and obtained correlation coefficients r² higher than 0.98. It is important to note that Raman spectra were conveniently delimited in different regions in function of the excited vibrations in the structure in order to obtain the best fit in each case.

Infrared spectroscopy. A Perkin-Elmer model 1000 FTIR spectrometer was used for infrared (IR) spectra collection. The sample scans ranged from 400 to 4000 cm⁻¹ with 4 cm⁻¹ resolution. Some regions of the IR spectra were curve-fitted to obtain quantitative information on the wavenumbers, widths, and

relative intensities. For that purpose, we used Lorentzians lines and obtained correlation coefficients r² higher than 0.98.

Mössbauer spectroscopy. The transmission Mössbauer spectra were recorded at 300 K and 77 K using a 25 mCi ⁵⁷Co in rhodium source. Mössbauer spectra were collected in a time-mode spectrometer working in the transmission geometry using a constant acceleration drive with triangular reference signal. Calibration was achieved from standard α-iron foil at 300 K. The spectra were analyzed using a homemade program called MOSFIT which is based on non-linear least squares fitting procedures assuming Lorentzian Mössbauer lines while the values of isomer shift are quoted to that of α -Fe at 300 K.

Table SI_1. Amount of antimony, arsenic, iron and chloride present in the structure of samples coprecipitated with 60 ppm of the pollutants as determined by XRF. Only the amount of mercury was determined by STEM.

Element	Weight (%)	Element	Weight (%)	Element	Weight (%)
Hg	0.94 ± 0.01	Sb	1.41 ± 0.05	As	3.51 ± 0.08
Fe	65.53 ± 0.09	Fe	62.59 ± 0.11	Fe	63.50 ± 0.10
Cl	5.64 ± 0.12	Cl	7.25 ± 0.13	Cl	3.82 0.10

Table SI_2. Percentage distribution dissolved species of Hg^{2+} , Sb^{3+} and As^{5+} in the reaction medium at room temperature (25°C). In all experiments, the concentration of FeCl₃·6H₂O was fixed at 0.05M, while the concentrations of pollutants were fixed at 60ppm of Hg(II), Sb(III) and As(V). The pH was 1.19 for combined solution of FeCl₃ and Hg²⁺, 0.96 for combined solution of FeCl₃ and Sb³⁺ and 1.59 for combined solution of FeCl₃ and As⁵⁺. Only the most abundant species are listed.

Component	Species name	Total concentration (%)	Component	Species name	Total concentration (%)	Component	Species name	Total concentration (%)
Hg(OH) ₂	HgCl ₂ (aq)	63.608	Sb(OH) ₃	Sb(OH) ₂ +	79.37	AsO ₄ ³⁻	H ₃ AsO ₄	80.722
	HgCl ₃ ¹⁻	27.922		Sb(OH)₃	20.63		H ₂ AsO ₄ ⁻	19.278
Cl1-	Cl1-	82.583	Cl1-	Cl1-	83.867	Cl ¹⁻	Cl ¹⁻	84.432
	FeCl ₂ +	15.948		FeCl ₂ ⁺	16.133		FeCl ₂ +	15.568
								•
Fe ³⁺	Fe ³⁺	80.36	Fe ³⁺	Fe ³⁺	81.971	Fe ³⁺	Fe ³⁺	74.748
	FeCl ₂ +	15.948		FeCl ₂ +	16.134		FeCl ₂ ⁺	15.569
	Fe(OH) ₂ +	2.592		Fe(OH) ₂ ⁺	1.511		Fe(OH)2 ⁺	5.248

Table SI_3. Evolution of a, b, c, and β parameters, unit cell volume and mean crystallize size with the increase of Hg²⁺, Sb³⁺ and As⁵⁺ concentrations.

Sample	a [Å]	b [Å]	c [Å]	β	Cell volume [Å ³]	Mean crystallize size [nm]
	10,5083	3,0217	10,4972	89,975	301,616	134
рАк	$\pm 0,0012$	$\pm 0,0003$	±0,0012	±0,012	±0,0003	±19
		•			•	
A1-0.2511~	10,5001	3,0214	10,4991	90,061	288,207	93
SamplepAkpAkAk0.25HgAk5HgAk20HgAk60HgAk0.25SbAk10SbAk20SbAk60SbAk60SbAk60SbAk60SbAk60As	$\pm 0,0035$	±0,0002	$\pm 0,0035$	±0,015	±0,0007	±16
A1-511-	10,5066	3,0204	10,4910	90,006	296,681	76
Акэпд	$\pm 0,0034$	±0,001	$\pm 0,0039$	±0,033	±0,001	± 8
A1-2011a	10,5247	3,0233	10,4866	90,025	294,52	38
Акдонд	$\pm 0,0044$	±0,0013	$\pm 0,0044$	±0,019	±0,0012	± 3
A1:60Ug	10,5023	3,0150	10,4343	89,879	311,097	20
Акоонд	$\pm 0,0060$	±0,0018	±0,0061	±0,023	±0,0017	± 1
Ak0.25Sb	10,5129	3,0226	10,5021	89,990	299,805	94
	±0,0024	±0,0007	±0,0022	±0,014	±0,001	± 6
A1-10Sb	10,5224	3,0214	10,4982	90,019	295,497	86
AKIUSU	±0,0105	$\pm 0,0006$	±0,0023	±0,017	±0,001	±11
A1-20Sh	10,4927	3,0128	10,4563	90,063	285,560	33
AK2050	±0,0022	±0,0021	$\pm 0,0076$	$\pm 0,050$	$ \begin{array}{c c} \pm 0,001 \\ 295,497 \\ \pm 0,001 \\ \hline 285,560 \\ \pm 1,001 \\ \hline 2240,726 \\ \hline \end{array} $	± 3
A1-60Sh	10,4970	3,0086	10,3999	90,242	249,726	17
AKOUSU	$\pm 0,0078$	±0,0023	$\pm 0,0079$	±0,04	±0,002	±1
A1-0.25 A a	10,5035	3,0203	10,5032	89,999	298,105	82
AKU.23AS	±0,0046	±0,0007	±0,0031	±0,015	±0,001	±19
A 1-5 A a	10,4932	3,0225	10,5248	90,058	289,318	54
AKJAS	$\pm 0,0029$	$\pm 0,0029$ $\pm 0,0009$ $\pm 0,003$ $\pm 0,019$		±0,001	± 4	
A1-20 A a	10,4762	3,0218	10,5235	90,037	292,141	41
AKZUAS	$\pm 0,0043$	±0,0013	±0,0045	±0,031	±0,001	±6
A1-60 A a	10,4444	2,9958	10,3564	89,981	292,458	13
AKOUAS	±0,0102	±0,0029	±0,013	±0,072	±0,003	±2



Figure SI_1. X-ray diffraction patterns of the samples co-precipitated in the presence of varying concentrations of Hg²⁺.



Figure SI_2. X-ray diffraction patterns of the samples co-precipitated in the presence of varying concentrations of Sb³⁺.



Figure SI_3. X-ray diffraction patterns of the samples co-precipitated in the presence of varying concentrations of As⁵⁺.



Figure SI_4. Variations of the mean crystallite sizes (purple diamond) and unit cell volumes (pink star) with the pollutant concentration for akaganeite co-precipitated with Hg^{2+} (bottom), Sb^{3+} (middle) and As^{5+} .

Table SI_4. Comparison of the wavenumbers (in cm⁻¹) of the principal bands observed in the Raman spectrum of pure akaganeite in this study with some data reported in the literature. The data are divided into two wavenumber regions: low and high.

	Low wavenumber region (100 - 1800 cm ⁻¹)	High wavenumber region (3000 - 4000 cm ⁻¹)
This study	137, 310, 386, 491, 541, 600, 720, 886, 1149, broad band centered at about 1361.	3296, 3368, 3426, 3523
Oh and co-worker ⁵¹	314, 380, 549, 722	Non reported
Das and Hendry ⁵²	314, 400, 550, 720	Non reported
Richmond and co-workers ²⁵	137, 307, 387, 535, 597, 719, 906	Non reported
Réguer and co-workers ³	310, 390, 490, 540, 610, 720, 930, 1160, broad band from 1300 to 1500.	3242, 3360, 3422, 3530



Figure SI_5. Raman spectra of pAk, Ak0.25Hg, Ak5Hg, Ak20Hg and Ak60Hg in the wavenumber region from 100 to 1200 cm⁻¹.



Figure SI_6. Raman spectra of pAk, Ak0.25Hg, Ak5Hg, Ak20Hg and Ak60Hg in the wavenumber region from 2800 to 4000 cm⁻¹.



Figure SI_7. Raman spectra of pAk, Ak0.25Sb, Ak10Sb, Ak20Sb and Ak60Sb in the wavenumber region from 100 to 1200 cm⁻¹.



Figure SI_8. Raman spectra of pAk, Ak0.25Sb, Ak0.5Sb, Ak0.75Sb, Ak1Sb, Ak10Sb, Ak20Sb and Ak60Sb in the wavenumber region from 2800 to 4000 cm⁻¹.



Figure SI_9. Raman spectra of pAk, Ak0.25As, Ak5As, Ak20As and Ak60As in the wavenumber region from 100 to 1200 cm⁻¹.



Figure SI_10. Raman spectra of pAk, Ak0.25As, Ak5As, Ak20As and Ak60As in the wavenumber region from 2800 to 4000 cm⁻¹.



Figure SI_11. Decomposition of Raman spectra for pAk, Ak60Hg, Ak60Sb and Ak60As in the wavenumber region from 200 to 600 cm⁻¹.

Table SI_5. Wavenumbers and integrated areas of the Raman bands localized from 200 to 600 cm⁻¹ corresponding to the pure and co-precipitated akaganeites with 60 ppm of Hg^{2+} , Sb^{3+} and As^{5+} . The relative intensity ratio of the two Raman bands located at about 310 and 330 cm⁻¹ for each spectrum is also reported and the calculated chlorine content according to Figure 4 of the paper by Réguer and co-workers.³

			Relative	Cl content (%
Samnle	Wavenumbers	Integrated area	intensity ratio	mass) using
Sampic	[cm ⁻¹]	[%]	of selected	data from
			paired bands	Réguer <i>et al.</i> ³
	297 ± 3	11 ± 4		
	310 ± 2	24 ± 4		
	333 ± 1	12 ± 1	I (7	
pAk	363 ± 2	10 ± 1	Relative CI content (intensity ratio mass) using the selected of selected intensity ratio mass) using the selected intensity ratio [%] of selected intensity ratio data from Réguer et al. 11 ± 4 $\overline{24 \pm 4}$ $\overline{12 \pm 1}$ $\overline{1310}_{1333} = \frac{67}{33}$ $9 - 10$ 10 ± 1 $\overline{1310}_{1333} = \frac{67}{33}$ $9 - 10$ $9 - 10$ $\overline{27 \pm 1}$ $\overline{1333} = \frac{67}{33}$ $9 - 10$ $\overline{39 \pm 1}$ $\overline{14 \pm 1}$ $\overline{1333} = \frac{74}{26}$ Not graphe $\overline{30 \pm 1}$ $\overline{1339} = \frac{74}{26}$ Not graphe $\overline{37 \pm 1}$ $\overline{1339} = \frac{74}{26}$ Not graphe $\overline{26 \pm 2}$ $\overline{12 \pm 2}$ $\overline{11 \pm 5}$ $\overline{1339} = \frac{74}{26}$ Not graphe	9 – 10
	386 ± 1	27 ± 1		
	415 ± 1	20 ± 1		
	437 ± 2	9 ± 1		
	309 ± 1	39 ± 1		
A1:60Hz	341 ± 1	14 ± 1	I 74	
Ak60Hg	385 ± 1	32 ± 1	$\frac{I_{309}}{I_{341}} = \frac{74}{26}$	Not graphed
	419 ± 1	30 ± 1		
	443 ± 2	10 ± 1		
	308 ± 1	37 ± 1		
	339 ± 1	13 ± 1	I 74	
Ak60Sb	384 ± 1	28 ± 1	$\frac{I_{308}}{I_{339}} = \frac{74}{26}$	Not graphed
	417 ± 1	26 ± 2		
	437 ± 3	12 ± 2		
	301 ± 3	11 ± 5		
	313 ± 2	24 ± 4		

339 ± 1	13 ± 1	
366 ± 3	9 ± 1	
387 ± 1	23 ± 1	
411 ± 3	15 ± 3	
427 ± 2	19 ± 3	
447 ± 4	7 ± 2	



Figure SI_12. Raman spectra in the range from 775 and 1000 cm⁻¹ for pure and co-precipitated with 60 ppm As⁵⁺ akaganeites.



Figure SI_13. IR spectrum of pAk. The inset shows the region between 750 to 900 cm⁻¹.

Table SI_6. Wavenumbers of prominent bands observed in the infrared spectrum of pure akaganeite and
compared with other studies. The table also includes the band assignments given by different authors.

Wavenumbers [cm ⁻¹]	Wavenumbers [cm ⁻¹] published	Band assignments given by
of this study	in other studies	different authors
	420 and 490 [54]	Symmetrical stretching vibrations of
	Band from 439 to 491 [55]	Fe-O in the octahedral site. ⁵⁵
434 and 475	420 and 421 (dauktat) [56]	Translational modes of the FeO_6
	420 and 481 (doublet) 100	octahedral unit.56
	650 [54]	A symmetrical stratching wibrations of
	700 [55]	Asymmetrical stretching violations of
638+670 (doublet)		Fe-O-Fe ^[55]
	$644+697 (doublet)^{[56]}$	
822+831 (doublet)	820+850 (doublet) ^[54]	Librations of linear and bifurcated O-
	820+847 (doublet) [56]	HCl at the tunnels. ⁵⁶
1516 11624	1630 [54]	
1516 and 1634	Band from 1630 to 1648 [55]	OH and H_2O bending. ³⁴
	3390+3480 (doublet) ^[34]	OH structural and molecular $H_2O.^{54}$
3356+3468 (doublet)	3392-3458 (doublet) [56]	Vibration of linear and bifurcated O-
	Broad band from 3430 to 3458 [55]	HCl bonds. ⁵⁶



Figure SI_14. IR spectra of pAk, Ak60Hg, Ak60Sb and Ak60As in the wavenumber region from 3000 to 4000 cm⁻¹.



Figure SI_15. IR spectra of pAk, Ak60Hg, Ak60Sb and Ak60As in the wavenumber region from 400 to 750 cm⁻¹.



Figure SI_16. 300 K Mössbauer spectra of pAk, Ak60Hg, Ak60Sb and Ak60As.

Table SI_7. Refined values of hyperfine parameters resulting from the 300K and the 77K Mössbauer spectra of pure akaganeite: Isomer shift (δ), quadrupolar shift (2 ϵ), quadrupolar splitting (Δ), linewidth (Γ /2), magnetic hyperfine field (B_{hf}) and relative absorption area (A).

T [17]		δ	$2\varepsilon \text{ or } \Delta$		B _{hf} [T]	Area
T[K]	Components	[mm/s]	[mm/s] [mm/s]			[%]
300	D1	0.38±0.003	0.54±0.01	0.15±0.003	0	60
T [K] 300 77	D2	0.39±0.003	0.94±0.01	0.17±0.01	0	40
	S1	0.50±0.01	-0.09±0.01	0.18±0.01	48.0±0.5	28
T [K] 300 77	S2	0.50±0.01	-0.09±0.01	0.18±0.01	46.5±0.6	28
	\$3	0.47±0.01	-0.39±0.02	0.24±0.02	46.3±2.0	22
	S4	0.47±0.01	-0.39±0.02	0.24±0.02	44.0±1.2	22

Table SI_8. Refined values of hyperfine parameters resulting from the 300K Mössbauer spectra of akaganeites co-precipitated with Hg²⁺, Sb³⁺ and As⁵⁺. Isomer shift (δ), quadrupolar shift (2 ϵ), quadrupolar splitting (Δ), linewidth (Γ /2), magnetic hyperfine field (B_{hf}) and relative absorption area (A).

		Hg(II)		
Samula	Components	δ	2ε	Γ/2	Area
Sample	Components	[mm/s]	[mm/s]	[mm/s]	[%]
Ak20Hg	D1	0.38±0.002	0.54±0.01	0.17±0.004	60
	D2	0.38±0.003	0.93±0.02	0.19±0.01	40
Ak60Hg	D1	0.38±0.002	0.55±0.01	0.16±0.002	62
	D2	0.38±0.002	0.94±0.01	0.18±0.01	38
		Sb(III)		
Ak10Sb	D1	0.38±0.003	0.55±0.01	0.16±0.003	63
	D2	0.38±0.003	0.97±0.01	0.16±0.01	37
Ak20Sb	D1	0.38±0.003	0.56±0.01	0.16±0.002	67
	D2	0.38±0.003	0.98±0.01	0.18±0.01	33
Ak60Sb	D1	0.38±0.002	0.54±0.00	0.17±0.002	65
	D2	0.38±0.003	0.95±0.01	0.18±0.003	35
	1	As(V)	1	
Ak60As	D1	0.38±0.003	0.56±0.004	0.17±0.002	59
	D2	0.38±0.003	0.94±0.00	0.2±0.004	41

Table SI_9. Refined values of hyperfine parameters of the 77K Mössbauer spectra for akaganeites co-precipitated with Hg²⁺. Isomer shift (δ), quadrupolar shift (2 ϵ), hyperfine field (B_{hf}) and relative absorption area (A).

	S1				S2			83				<u>\$4</u>				
Sample	δ [mm/s]	2ε [mm/s]	$B_{hf}[T]$	Area [%] ±2	δ [mm/s]	2ε [mm/s]	B _{hf} (T)	Area [%] ±2	δ [mm/s]	2ε [mm/s]	B _{hf} [T]	Area [%] ±2	δ [mm/s]	2ε [mm/s]	B _{hf} [T]	Area [%] ±2
pAk	0.50 ±0.01	-0.09 ±0.01	48 ±0.5	28	0.50 ±0.01	-0.09 ±0.01	46.5 ±0.6	28	0.47 ±0.01	-0.39 ±0.02	46.3 ±2.0	22	0.47 ±0.01	-0.39 ±0.02	44.0 ±1.2	22
Ak5Hg	0.49 ±0.01	-0.09 ±0.01	47.9 ±0.5	27	0.49 ±0.01	-0.09 ±0.01	46.5 ±0.6	27	0.47 ±0.01	-0.38 ±0.02	46.2 ±2.1	23	0.47 ±0.01	-0.38 ±0.02	44.0 ±1.0	23
Ak20Hg	0.49 ±0.01	-0.10 ±0.01	47.9 ±0.5	28	0.49 ±0.01	-0.10 ±0.01	46.3 ±0.6	28	0.46 ±0.01	-0.39 ±0.02	45.9 ±1.9	22	0.46 ±0.01	-0.39 ±0.02	43.5 ±1.1	22
Ak60Hg	0.48 ±0.01	-0.13 ±0.01	47.2 ±0.5	30	0.48 ±0.01	-0.13 ±0.01	45.6 ±0.6	30	0.46 ±0.01	-0.40 ±0.02	45.1 ±1.7	20	0.46 ±0.01	-0.40 ±0.02	42.3 ±1.04	20

		S1				S2				S3				S4		
				Area				Area				Area				Area
	δ	2ε			δ	2ε	B_{hf}		δ	2ε	B_{hf}		δ	2ε	\mathbf{B}_{hf}	
Sample			$B_{hf}[T]$	[%]				[%]				[%]				[%]
_	[mm/s]	[mm/s]			[mm/s]	[mm/s]	(T)		[mm/s]	[mm/s]	[T]		[mm/s]	[mm/s]	[T]	
				±2				±2				±2				±2
	0.50	-0.09	48		0.50	-0.09	46.5		0.47	-0.39	46.3		0.47	-0.39	44.0	
pAk				28				28				22				22
	±0.01	±0.01	±0.5		±0.01	±0.01	±0.6		±0.01	±0.02	±2.0		±0.01	±0.02	±1.2	
	0.50	-0.13	47.8		0.50	-0.13	46.1		0.46	-0.33	45.6		0.46	-0.33	42.9	
Ak20Sb				35				35				15				15
	±0.004	±0.01	±0.4		±0.004	±0.01	±0.5		±0.01	±0.03	±1.4		±0.01	±0.03	±1.0	
	0.50	-0.14	47.2		0.50	-0.14	47.2		0.47	-0.34	45.1		0.47	-0.34	42.3	
Ak60Sb				30				30				20				20
	±0.004	±0.01	±0.5		±0.004	±0.01	±0.5		±0.01	±0.04	±1.6		±0.01	±0.04	±1.0	

Table SI_10. Refined values of hyperfine parameters of the 77K Mössbauer spectra for akaganeites coprecipitated with Sb³⁺. Isomer shift (δ), quadrupolar shift (2 ϵ), hyperfine field (B_{hf}) and relative absorption area (A).

Table SI_11. Refined values of hyperfine parameters of the 77K Mössbauer spectra for akaganeites coprecipitated with As⁵⁺. Isomer shift (δ), quadrupolar shift (2 ϵ), hyperfine field (B_{hf}) and relative absorption area (A).

		S1				S2				S3			84			
Sample	δ [mm/s]	2ε [mm/s]	B _{hf} [T]	Area [%] ±2	δ [mm/s]	2ε [mm/s]	B _{hf} (T)	Area [%] ±2	δ [mm/s]	2ε [mm/s]	B _{hf} [T]	Area [%] ±2	δ [mm/s]	2ε [mm/s]	B _{hf} [T]	Area [%] ±2
pAk	0.50 ±0.01	-0.09 ±0.01	48 ±0.5	28	0.50 ±0.01	-0.09 ±0.01	46.5 ±0.6	28	0.47 ±0.01	-0.39 ±0.02	46.3 ±2.0	22	0.47 ±0.01	-0.39 ±0.02	44.0 ±1.2	22
Ak5As	0.49 ±0.01	-0,09 ±0,01	48.0 ±0.5	27	0.49 ±0.01	-0,09 ±0,01	46.7 ±0.6	27	0.46 ±0.01	-0.39 ±0.02	46.3 ±1.9	23	0.46 ±0.01	-0.39 ±0.02	44.1 ±1.0	23
Ak20As	0.49 ±0.01	-0.11 ±0.01	47.9 ±0.5	30	0.49 ±0.01	-0.11 ±0.01	46.4 ±0.5	30	0.46 ±0.01	-0.41 ±0.02	46.0 ±1.6	20	0.46 ±0.01	-0.41 ±0.02	43.6 ±0.9	20
Ak60As	0.48 ±0.01	-0.14 ±0.01	46.7 ±0.7	27	0.48 ±0.01	-0.14 ±0.01	44.9 ±0.8	27	0.45 ±0.01	-0.28 ±0.02	44.2 ±2.5	23	0.45 ±0.01	-0.28 ±0.02	41.1 ±1.5	23

Table SI_12. Linewidths of the four sextet components of the 77 K Mössbauer spectra for the coprecipitated samples with Hg^{2+} , Sb^{3+} and As^{5+} .

	S1	82	S3	S4
Sample	Γ/2 [mm/s]	Γ/2 [mm/s]	Г/2 [mm/s]	Γ/2 [mm/s]
pAk	0.18±0.01	0.18±0.01	0.24±0.02	0.24±0.02
Ak5Hg	0.20±0.01	0.20±0.01	0.25±0.01	0.25±0.01
Ak20Hg	0.21±0.01	0.21±0.01	0.27±0.01	0.27±0.01
Ak60Hg	0.22±0.01	0.22±0.01	0.26±0.01	0.26±0.01
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Ak20Sb	0.22±0.01	0.22±0.01	0.22±0.01	0.22±0.01
Ak60Sb	0.21±0.01	0.21±0.01	0.25±0.02	0.25±0.02
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Ak5As	0.18±0.01	0.18±0.01	0.24±0.01	0.24±0.01
Ak20As	0.19±0.01	0.19±0.01	0.23±0.01	0.23±0.01
Ak60As	0.21±0.01	0.21±0.01	0.30±0.02	0.30±0.02