Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Supplementary information

Table S1. pH values in the course of U(VI) hydrazinate synthesis (0.1 M uranyl nitrate aqueous solution and 0.1M hydrazine aqueous solution in 1:3 volume ratio).

Volume of N_2H_4 added, %	0	25	50	75	100	125	150	175	200	225	250	275	300
pH	2.5	3.2	3.5	3.8	4.0	4.4	4.8	5.0	5.4	6.3	7.3	7.6	

UV-vis spectroscopy: The initial solution of 0.1M hydrazine hydrate, this solution diluted with water (3:1 molar ratio) and mother liquor after reaction with uranyl nitrate have been analyzed, the results are shown at Fig. S1. It has been found, that the intensity of characteristic hydrazine peaks at wavelength ~1050 nm is significantly lower for mother liquor compared to diluted hydrazine solution, moreover, the shape of the peaks is also different.

Besides that, the obtained precipitate (20 mg) has been dissolved in the 17M hydrochloric acid (2 ml) and mixed (1:200) with *p*-dimethylaminobenzaldehyde (76 mg) solution (4 ml EtOH plus 1 ml 17M HCl). *p*-dimethylaminobenzaldehyde reacts with hydrazine to form *p*-Dimethylaminobenzalazine that has a distinct yellow color with corresponding absorbance peak at 457 nm. The UV-vis spectrum from this sample is shown at Fig. S2.

Therefore, UV-vis spectroscopy data unequivocally confirms a conclusion, that U(VI) binds hydrazine.

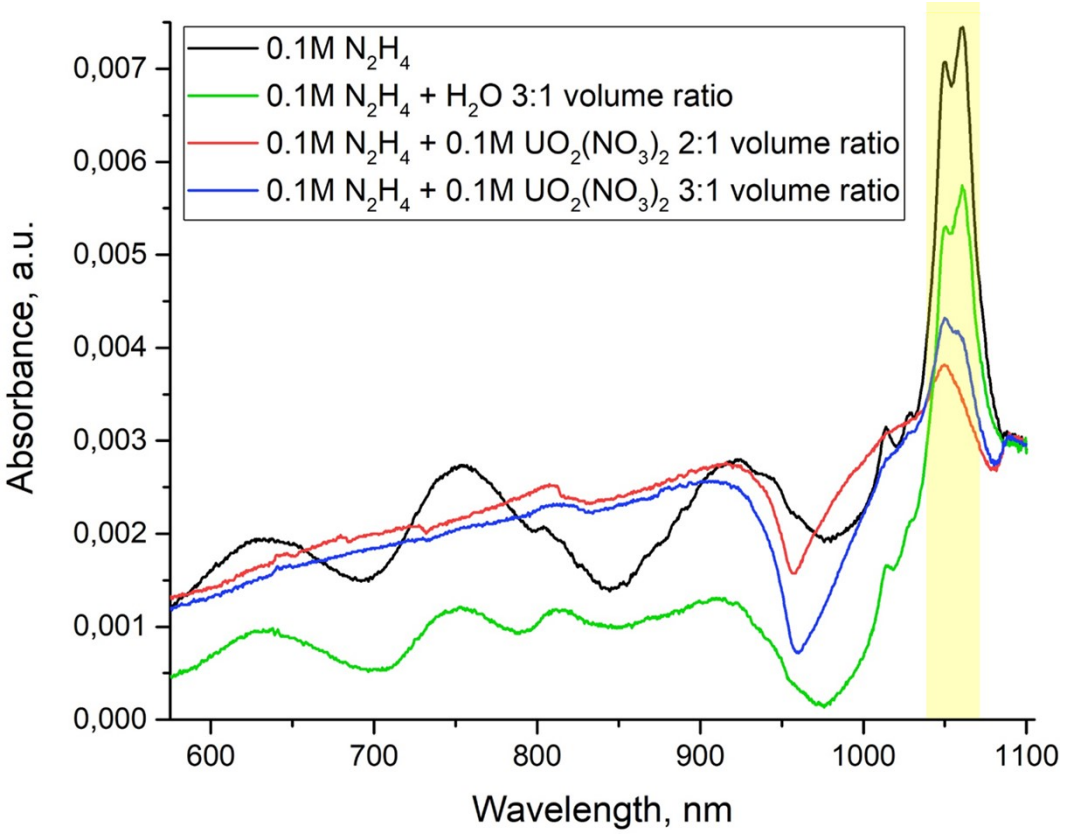

Fig. S1. UV-vis spectra of hydrazine hydrate solutions: initial solution (black), diluted solution and solutions after reaction with uranyl nitrate 2:1 volume ratio (red) and 3:1 volume ratio (blue). Wavelengths of the characteristic hydrazine peaks are marked with a yellow box.

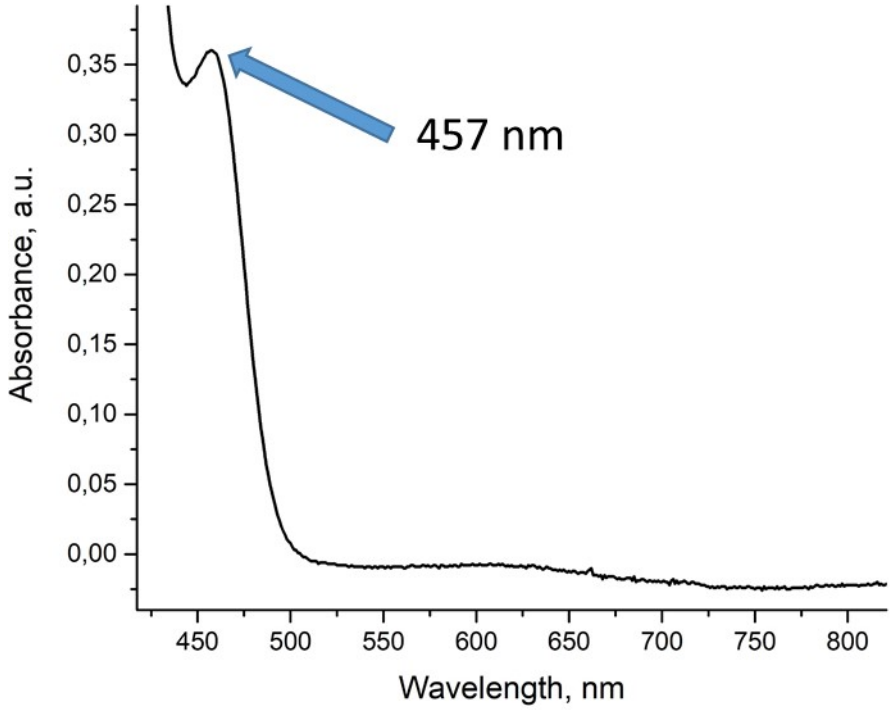

Fig. S2. UV-vis spectrum of *p*-dimethylaminobenzaldehyde and U(VI) hydrazinate, dissolved in hydrochloric acid.

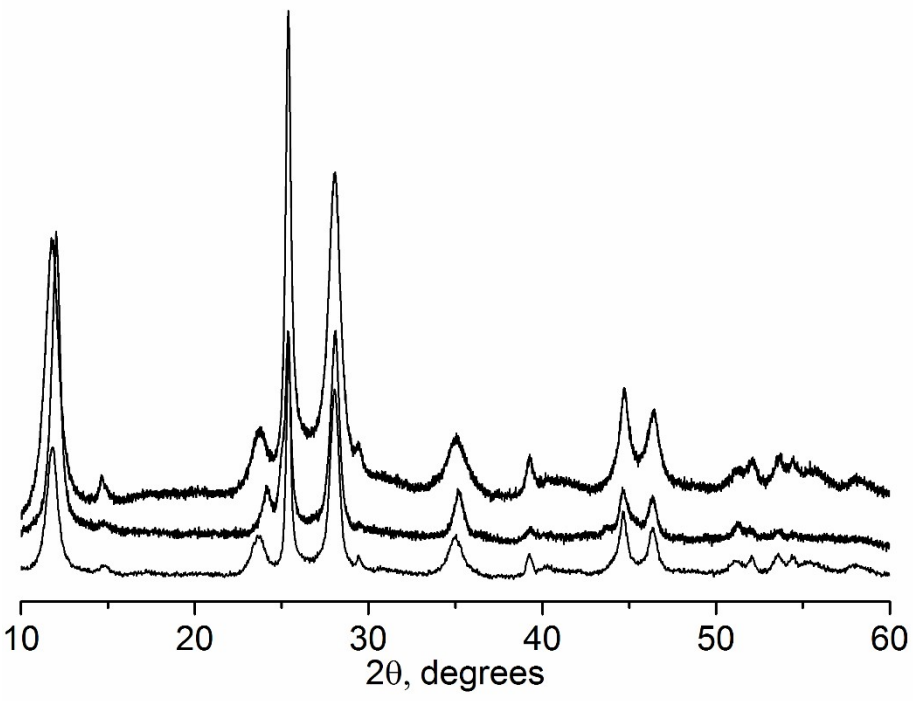

Fig. S3. Diffraction patterns of $xUO_3 \cdot yN_2H_4 \cdot zH_2O$ obtained from individual experiments to prove reproducibility.