Supporting Information for "Hydrate-based strategy for highly-efficient dewatering of waste-activated sludge through interfacial water reconfiguration and micro-pore-confined water reduction"

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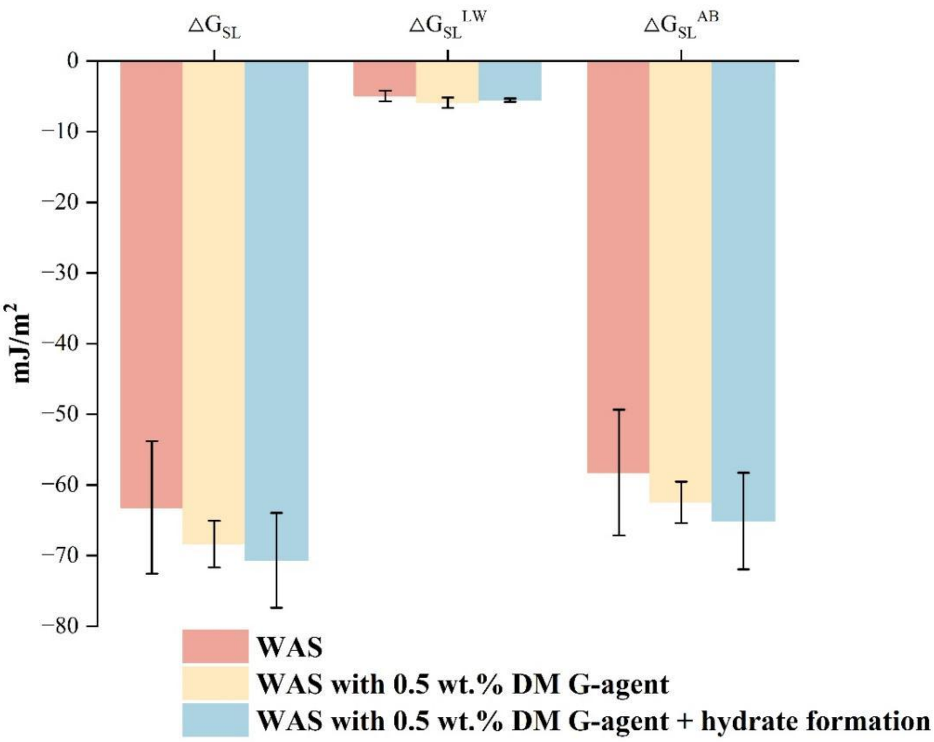

Figure S1. Interfacial free energy of raw WAS, WAS amended with 0.5 wt.% DM G-agent and G-agent-amended WAS with hydrate formation (water-to-hydrate conversion ratio of 100%). Experimental conditions: 200 mL WAS with initial water content of 98.0 wt.%; the headspace for gas pressurization was 800 mL; the initial pressure of CO_2 was 5 MPa; the temperature for CO_2 hydrate formation was kept at around 1.5 °C.

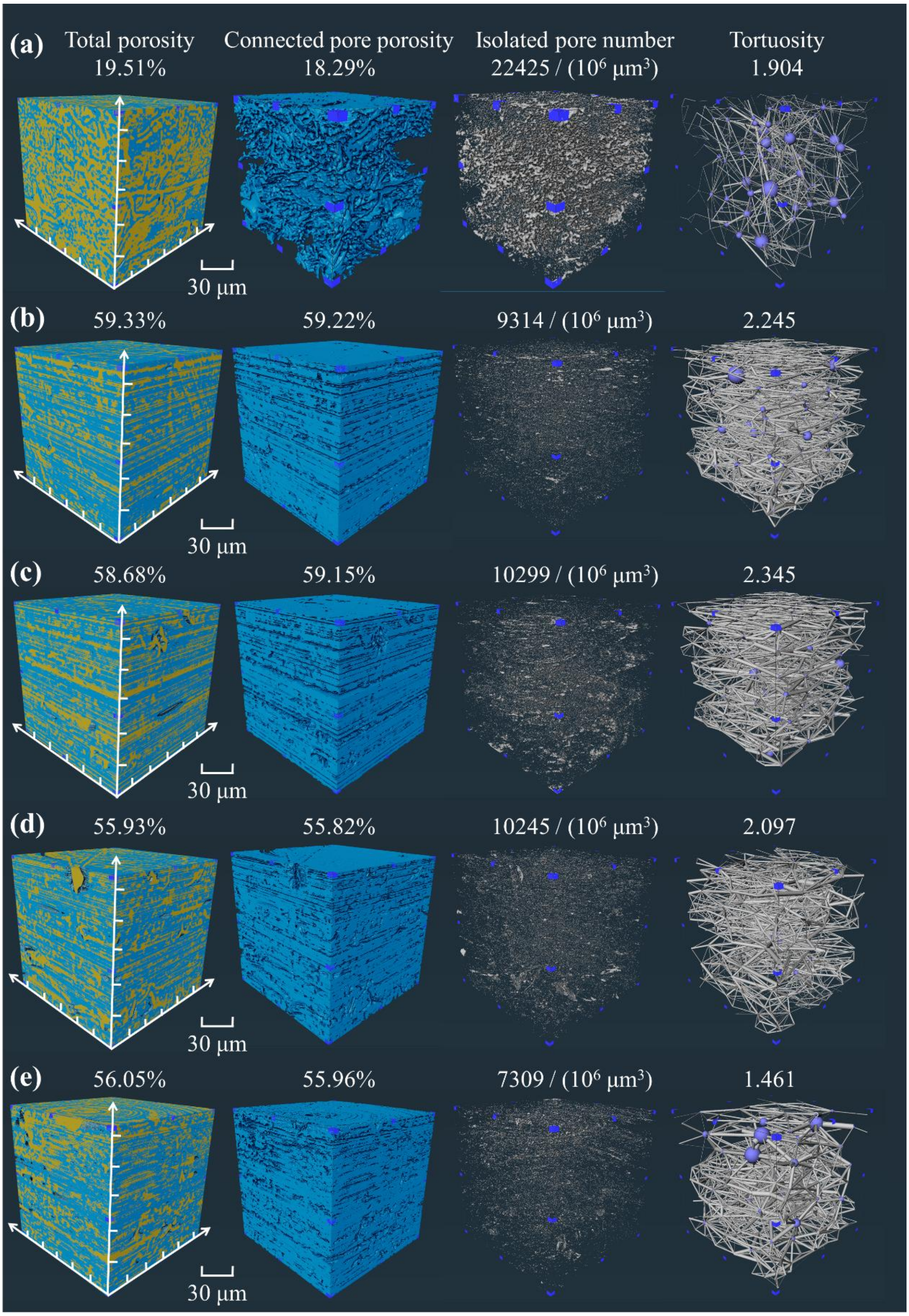

Figure S2. The reconstructed three-dimensional models of WAS with varying G-agent dosages: (a) raw sludge; (b) 0.3 wt.% DM G-agent; (c) 0.5 wt.% DM G-agent; (d) 0.7 wt.% DM G-agent; (e) 1 wt.% DM G-agent.

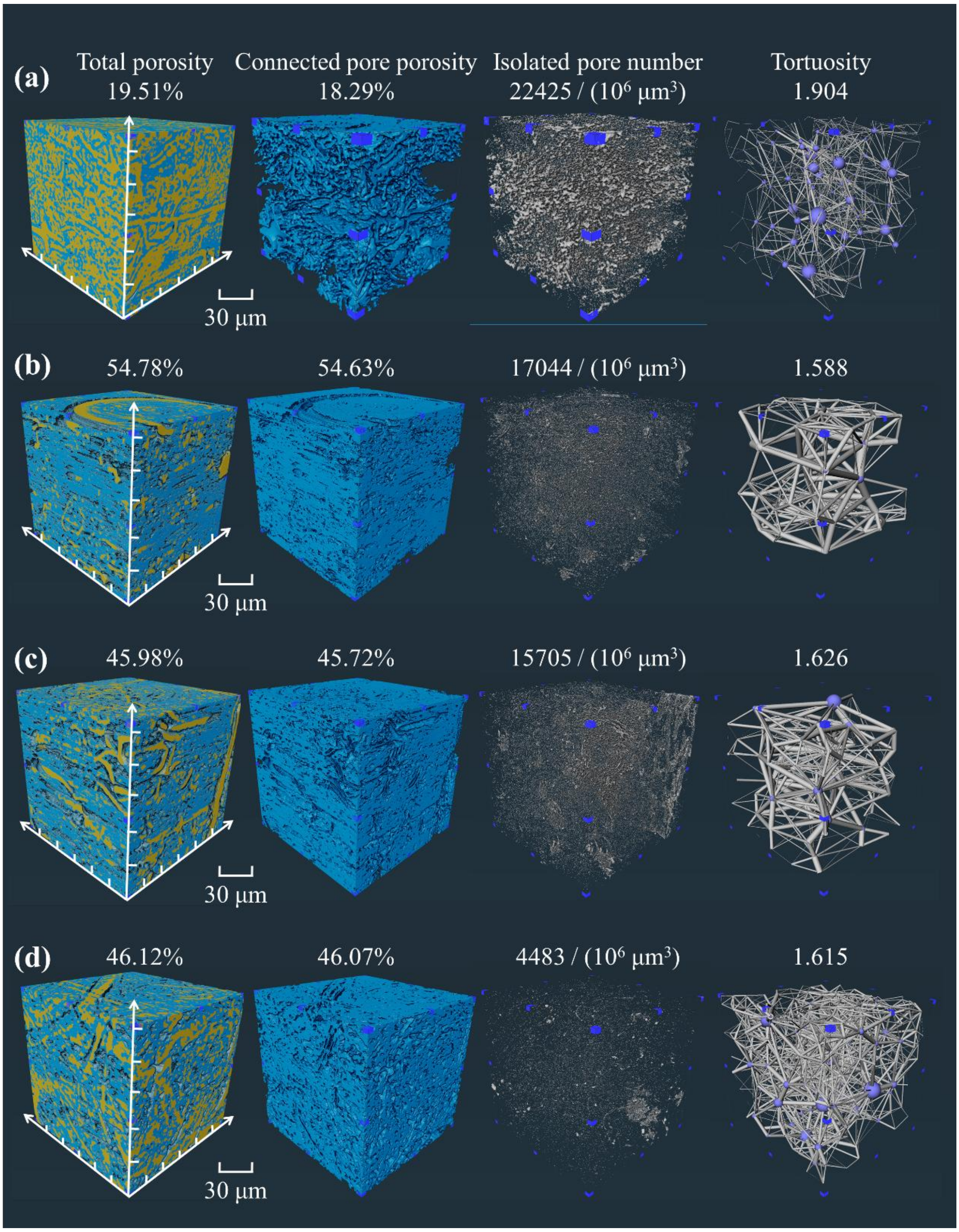

Figure S3. The reconstructed three-dimensional models of WAS with varying water-to-hydrate conversion ratios: (a) raw sludge; (b) 20% water conversion; (c) 60% water conversion; (d) 100% water conversion.

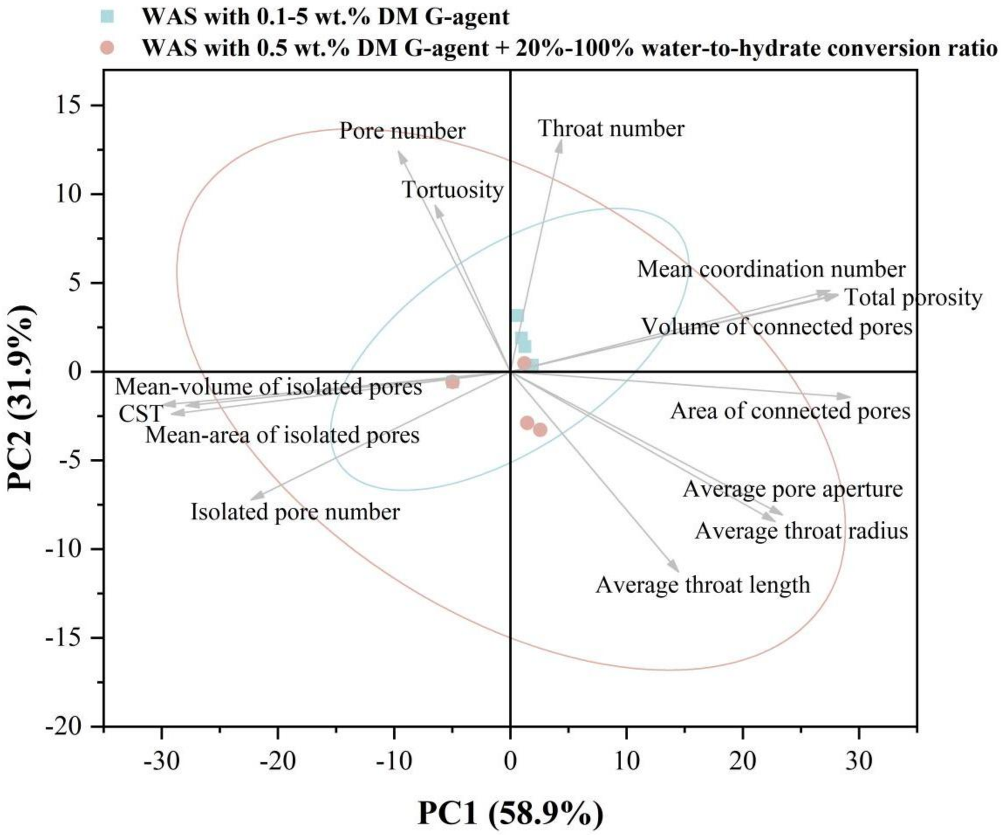

Figure S4. Principle component analysis (PCA) biplot of WAS dewaterability under different G-agent dosages and water-to-hydrate conversion ratios.

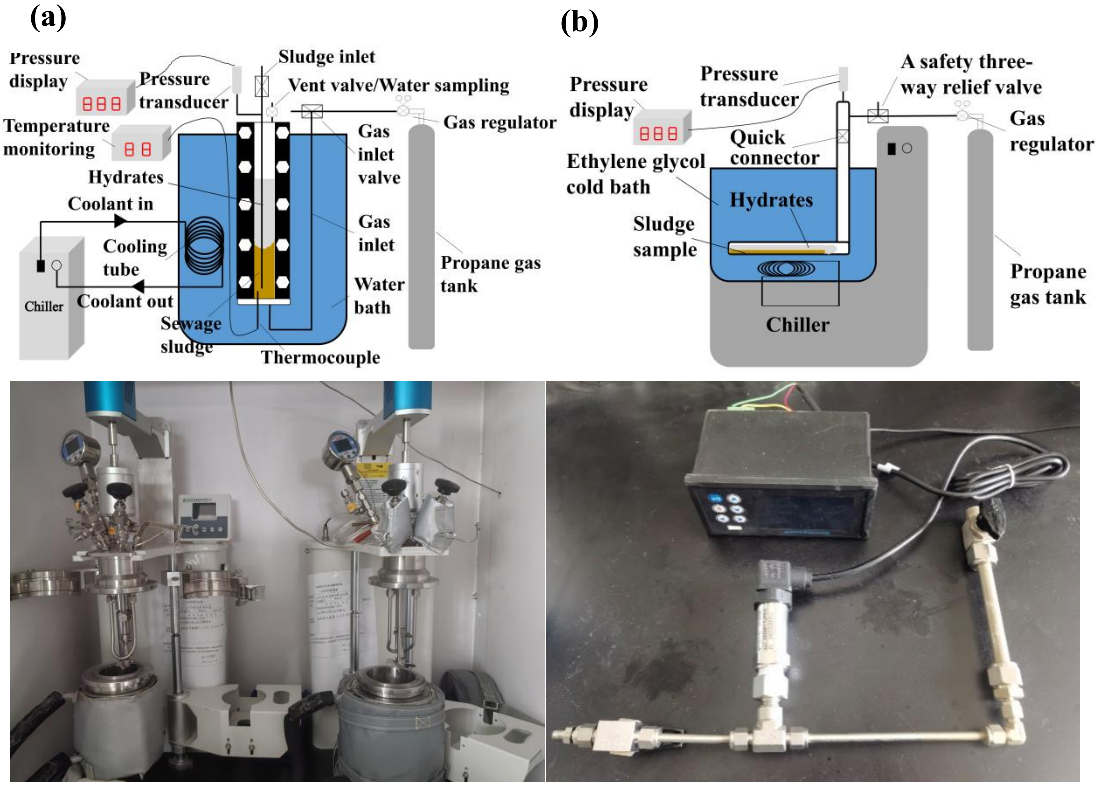

Figure S5. Set-up for propane hydrate formation: (a) vessel reactor (b) tube reactor.

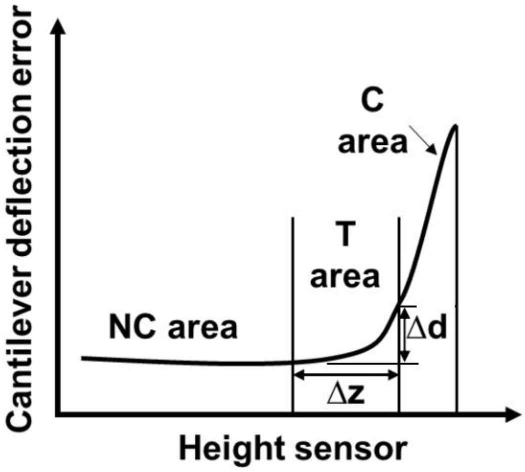

Figure S6. Sketch map of atomic force microscope (AFM) force curve measured on the solid samples with hydrated film[1].

Table S1. The basic properties of WAS samples and G-agent

Sample	Properties	Content
WAS samples	pH	6.84±0.11
	Total solids (TS, g/L)	15.50±0.20
	Volatile suspended solids (VSS, g/g TS)	0.52±0.08
	CST (s)	138.80±5.70
G-agent	Compositions	Dicyandiamide, poly(dimethyldiallylammonium chloride), dimethylallyl chloride

Table S2. Key porosity parameters of WAS samples obtained by the hydrate-based dewatering process, and WAS samples with varying water-to-hydrate conversion ratios and G-agent dosages

		Total porosity	Isolated pore			Connected pore	
			Number (per 10^6 μm^3)	Porosity	Mean-area (μm^2)	Porosity	Area (μm^2)
Hydrate-based dewatering process proposed in section 3.3.2[a]	**Step I**	19.51%	22425	0.13%	2.283×10^4	18.29%	1.357×10^6
	Step II	21.60%	6150	0.30%	4.797×10^4	20.80%	1.567×10^6
	Step III	67.63%	2081	0.03%	0.811×10^4	67.61%	1.691×10^6
	Step IV	78.36%	597	0.04%	1.159×10^4	78.35%	2.235×10^6
	Step V	63.38%	592	0.05%	1.179×10^4	63.37%	2.185×10^6
Sludge with varying G-agent dosages	**0 wt.%DM**	19.51%	22425	0.13%	2.283×10^4	18.29%	1.357×10^6
	0.3 wt.%DM	59.33%	9314	0.03%	0.770×10^4	59.22%	2.776×10^6
	0.5 wt.%DM	58.68%	10299	0.04%	1.121×10^4	59.15%	3.451×10^6
	0.7 wt.%DM	55.93%	10245	0.03%	0.826×10^4	55.82%	3.430×10^6
	1.0 wt.%DM	56.05%	7309	0.03%	0.962×10^4	55.96%	3.355×10^6
Sludge with varying water-to-hydrate conversion ratios	**20%**	54.78%	17044	0.02%	0.688×10^4	54.63%	4.021×10^6
	60%	45.98%	15705	0.04%	1.193×10^4	45.72%	3.370×10^6
	100%	46.12%	4483	0.03%	0.775×10^4	46.07%	2.769×10^6

[a] Steps of hydrate-based dewatering process proposed in section 3.3.2: Step I. Raw WAS (98.0 wt.% water content); Step II. WAS amended with 0.7 wt.% DM G-agent followed by centrifugation; Step III. WAS processed by hydrate formation (water-to-hydrate conversion ratio of 100%); Step IV. Hydrates-treated WAS further amended with 0.3 wt.% DM G-agent; Step V. WAS after pressure filtration.

Table S3. Key PNM parameters of WAS samples obtained by the hydrate-based dewatering process, and WAS samples with varying water-to-hydrate conversion ratios and G-agent dosages

		Pore Number (per 10^6 μm^3)	Average aperture (μm)	Mean coordination number	Throat Number (per 10^6 μm^3)	Average throat radius (μm)	Average throat length (μm)	Tortuosity
Hydrate-based dewatering process proposed in section 3.3.2[a]	**Step I**	115	11.336	5.087	293	1.446	28.538	1.904
	Step II	96	13.896	6.597	317	2.046	28.486	1.371
	Step III	186	17.058	10.652	989	3.900	23.752	1.688
	Step IV	176	18.564	12.085	1066	4.066	25.940	2.004
	Step V	238	15.552	11.297	1348	3.260	22.293	1.826
Sludge with varying G-agent dosages	**0 wt.%DM**	115	11.336	5.087	293	1.446	28.538	1.904
	0.3 wt.%DM	191	15.992	9.966	951	3.019	26.086	2.245
	0.5 wt.%DM	132	17.622	9.947	658	3.374	30.467	2.345
	0.7 wt.%DM	120	16.810	9.612	577	3.298	31.350	2.097
	1.0 wt.%DM	93	19.706	11.101	515	4.034	32.086	1.461
Sludge with varying water-to-hydrate conversion ratios	**20%**	19	31.092	8.585	82	7.720	51.353	1.588
	60%	23	27.776	9.122	104	6.133	50.982	1.626
	100%	88	18.448	10.677	470	3.865	31.197	1.615

[a] Steps of hydrate-based dewatering process proposed in section 3.3.2: Step I. Raw WAS (98.0 wt.% water content); Step II. WAS amended with 0.7 wt.% DM G-agent followed by centrifugation; Step III. WAS processed by hydrate formation (water-to-hydrate conversion ratio of 100%); Step IV. Hydrates-treated WAS further amended with 0.3 wt.% DM G-agent; Step V. WAS after pressure filtration.

References

1 Wu, B. *et al.* Crystallization-driven evolution of water occurrence states with implications on dewaterability improvement of waste-activated sludge. *Water Research* **244** (2023). https://doi.org/10.1016/j.watres.2023.120496