Aerobic ammonium-oxidizing bacteria (AerAOB) and anoxic ammonium-oxidizing bacteria (AnAOB) cooperate in partial nitritation/anammox systems to remove ammonium from wastewater. In this process, large granular microbial aggregates enhance the performance, but little is known about granulation so far. In this study, three suspended-growth oxygen-limited autotrophic nitrification-denitrification (OLAND) reactors with different inoculation and operation (mixing and aeration) conditions, designated reactors A, B, and C, were used. The test objectives were (i) to quantify the AerAOB and AnAOB abundance and the activity balance for the different aggregate sizes and (ii) to relate aggregate morphology, size distribution, and architecture putatively to the inoculation and operation of the three reactors. A nitrite accumulation rate ratio (NARR) was defined as the net aerobic nitrite production rate divided by the anoxic nitrite consumption rate. The smallest reactor A, B, and C aggregates were nitrite sources (NARR, > 1.7). Large reactor A and C aggregates were granules capable of autonomous nitrogen removal (NARR, 0.6 to 1.1) with internal AnAOB zones surrounded by an AerAOB rim. Around 50% of the autotrophic space in these granules consisted of AerAOB- and AnAOB-specific extracellular polymeric substances. Large reactor B aggregates were thin film-like nitrite sinks (NARR, <0.5) in which AnAOB were not shielded by an AerAOB layer. Voids and channels occupied 13 to 17% of the anoxic zone of AnAOB-rich aggregates (reactors B and C). The hypothesized granulation pathways include granule replication by division and budding and are driven by growth and/or decay based on species-specific physiology and by hydrodynamic shear and mixing.