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Structural Features of Zirconium-Based Metal–Organic Frameworks Affecting Radiolytic Stability

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Metal–organic frameworks (MOFs) NU-1000 and UiO-66 are herein exposed to two different gamma irradiation doses and dose rates and analyzed to determine the structural features which affect their stability in these environments. MOFs have shown promise for the capture and sensing of off-gases at civilian nuclear energy reprocessing sites, nuclear waste repositories, and nuclear accident locations. However, little is understood about the structural features of MOFs which contribute to their stability levels under the ionizing radiation conditions present at such sites. This study is the first of its kind to explore the structural features of MOFs which contribute to their radiolytic stability. Both NU-1000 and UiO-66 are MOFs that contain Zr metal-centers with the same metal absorption cross section. However, the two MOFs exhibit different linker connectivity, linker aromaticity, node density, node connectivity, and inter-ligand separations. In this study, NU-1000 and UiO-66 were exposed to high (423.3 Gy/min, 23 min and 37 sec) and low

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(0.78 Gy/min, 4320 min) dose rates of ^{60}Co gamma irradiation. NU-1000 displayed insignificant radiation damage under both dose rates, due to its high linker connectivity, low node density, and low node connectivity. However, low radiation dose rates caused considerable damage to UiO-66, a framework with lower aromaticity and smaller inter-ligand separation. Results suggest that chronic, low radiation environments are more detrimental to Zr MOF stability than acute, high radiation conditions. Sandia National Laboratories is managed and operated by NTESS under DOE NNSA contract DE-NA0003525.