The nuclear industry for electricity production generates 12,000 tons of nuclear-spent fuel annually. During the burning of nuclear fuel in reactors, more than 300 radionuclides are produced, corresponding to nearly 50 elements (from Ge to Dy and minor actinides). Reprocessing nuclear-spent fuel generates highly radioactive liquid wastes that must be isolated from the biosphere until their radiotoxicity level drops back to the initial uranium ore. Among them, long-lived radionuclides such as ^{135}Cs, ^{129}I, and ^{99}Tc demonstrated a lower contribution to long-term radiotoxicity than minor actinides but are highly mobile in the geological environment and soluble in groundwater. Numerous efforts are made to minimize their long-term impact on the environment. Existing concepts claim transformation of HLW into short-lived radionuclides, stable elements (transmutation), or immobilization in host matrices. Therefore, the selective separation and immobilization of hazardous radionuclides in highly durable matrices would reduce the risks of their migration to the biosphere during disposal. Various chemically durable materials with long-term stability and high resistance to radiation damage, such as glasses, ceramics, and glass-ceramics, have been considered for the immobilization of radioactive waste. Today, all elements in HLW are immobilized together in alkali aluminoborosilicate glasses. Other ways to dispose of HLW are still challenging and require improvements to become relevant and attractive for the industry.

Therefore, this project aims to develop a new and economically valuable method to immobilize radioactive waste by dissolution on the atomic scale in highly stable crystalline or partially crystallized matrices. Taking inspiration from nature, where the combination of $Ca^{2+}/Sr^{2+}/Ba^{2}$ and Ti^{4+}/Zr^{4+} or Al^{3+} and Ca^{2+} occurs in many minerals such as perovskite $CaTiO_3$, zirconolite $CaZrTi_2O_7$, calzirtite $Ca_2Zr_5Ti_2O_{16}$, tausonite $SrTiO_3$, or zeolites, we designed heterometallic compounds for application in nuclear waste immobilization. To safely and effectively dispose of radioactive waste, it is necessary to design appropriate matrices with high durability, stability, and resistance to various physical and chemical conditions, such as temperature, radiation, pressure, and acidity/alkalinity. As a possible solution to the problem of high fractions of poorly soluble HLW components, high content of heat-generating radionuclides, and medium waste loading, this project proposes the preparation of matrices using heterometallic precursors.

Heterometallic alkoxide clusters composed of two different metal cations and optional ligands containing B, Si, P form M-O-M′ or M–O–M′–O–B(Si,P) motifs that, after removal of volatile organic compounds, will be transformed into ceramic or glass-ceramic materials.

The main goal of the proposed research is to develop effective methods for immobilizing high-level radioactive waste (HLW) by creating insoluble solid waste that will remain stable for many thousands of years. An important aspect of this project is understanding the incorporation of HLW elements into wholly or partially crystalline matrices derived from the decomposition of heterometallic precursors. These results will be necessary to determine the influence of the precursor structure on the morphology and properties of formed composite materials, which will allow the determination of variables necessary for the effective immobilization of separated and non-separated nuclear waste.

This information will be used to find correlations between the structures of the heterometallic precursors, the material properties of the wasteforms, and the amounts of nuclear waste incorporated in the matrices. Laboratory studies of minor actinides or Pu wastes are impossible because of their high radioactivity. Therefore, lanthanide ions (Ln^{3+}/Ln^{4+}) will be used as substitutes for the transuranic elements present in HLW, i.e., Am^{3+}, Cm^{3+}, Pu^{4+}, or Np^{4+}. Other short- or long-lived radionuclides such as $^{135,137}Cs^{+}$, $^{87}Rb^{+}$, $^{90}Sr^{2+}$, $^{93}Zr^{4+}$, ^{129}I will be simulated by their stable isotopes.

The incorporation of model HLW elements into ceramic or glass-ceramic matrices will lead to the formation of materials with physicochemical or mechanical properties completely different from those currently available. This approach is attractive for the synthesis of materials with desired functionalities, i.e., controlled density, shrinkage, or morphology, which will be essential in further catalytic or electrochemical applications.