

INVESTIGATION ON THE USE OF YELLOW TUFF IN ACIDACTIVATED GEOPOLYMERS FOR RADIONUCLIDES IMMOBILISATION

F. Fattori¹, E. Mossini^{1*}, G. Magugliani¹, C. Milanese², S. Paraboschi², G. D. Gatta³, D. Comboni³, A. Santi¹, F. Galluccio¹, E. Macerata¹, M. Mariani¹

¹*Politecnico di Milano, Department of Energy, Piazza Leonardo da Vinci 32, 20133, Milano, Italy*

²*Università di Pavia, Department of Chemistry, Viale Taramelli 16, 27100, Pavia, Italy*

³*Università degli Studi di Milano, Department of Earth Sciences, Via Sandro Botticelli 23, 20133, Milano, Italy*

* Corresponding author email: F. Fattori: fabio.fattori@polimi.it, E. Mossini: eros.mossini@polimi.it, G. Magugliani: gabriele.magugliani@polimi.it, C. Milanese: chiara.milanese@unipv.it, S. Paraboschi: sara.paraboschi01@universitadipavia.it, G. D. Gatta: diego.gatta@unimi.it, D. Comboni: davide.comboni@unimi.it, A. Santi: andrea.santi@polimi.it, F. Galluccio: francesco.galluccio@polimi.it, E. Macerata: elena.macerata@polimi.it, M. Mariani: mario.mariani@polimi.it

Radioactive waste (RW) conditioning is a crucial step to produce stable and durable waste forms, ensuring radionuclides immobilisation. In recent years, geopolymers have emerged as promising alternatives to traditional cementitious materials for the conditioning of low-to-intermediate level waste. Among them, alkaline geopolymers have demonstrated excellent retention performances for several cations, with particular regard to highly mobile fission products such as $^{137}\text{Cs}^+$. However, serious concerns arise when dealing with long lived isotopes such as ^{99}Tc , ^{93}Mo , ^{79}Se , ^{37}Cl or ^{129}I , which may be present as anionic species. In this context, phosphate-based geopolymers (PGP), typically activated using phosphoric acid, have demonstrated a higher efficiency in immobilising anions, according to preliminary literature studies. The objective of this research is to develop a PGP matrix capable of immobilising a broader range of contaminants, both cations and anions of interest for the nuclear industry. To this aim, novel PGP formulations using exclusively yellow tuff (YT) as precursor have been tested. YT is rich in zeolite chabazite (65% w/w), a crystalline aluminosilicate with high cation-exchange capacity, exhibiting high selectivity especially for Cs^+ . Furthermore, the use of YT as precursor offers significant environmental advantages as it is a natural and cost-effective material that requires minimal processing, primarily limited to grinding. Although already investigated in alkaline geopolymers, as to authors' knowledge, no works have been performed regarding YT activation in PGPs. A set of samples has been prepared, varying key parameters such as activator solution concentration, liquid-to-solid ratio, and curing temperature. The phase composition and morphology of the samples have been characterised using techniques such as powder X-ray diffraction, Fourier-transform infrared spectroscopy, and scanning electron microscopy - energy dispersive X-ray spectroscopy analyses. Although differences emerged between samples, in particular for those subjected to thermal curing, the stability of the zeolite chabazite phase remained largely unaffected under the investigated conditions, suggesting the preservation of Cs -retention capacity. To shine a light on the leaching behaviour of the new PGP formulation, stable isotopes of radionuclides commonly found in RWs, such as Cs , Sr , I , and Se , have been investigated, providing promising insights into the immobilisation behaviour of both cations and anions. The results were compared with the standards set by national 2 regulatory authorities. The findings from this work can be useful for the development of future tuff-based PGP formulations, paving the way for more effective and environmentally sustainable RW management strategies.

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