



Optimization of ZrO₂ coating deposited on carbon steel by Physical vapor Deposition (PVD) for protection in mildly saline environments.

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In the French concept of deep underground nuclear repository, the confinement of high-level nuclear waste is controlled by corrosion of a tight overpack made of low-alloy carbon steel (C-steel). The overpack dimensions are tailored to guarantee confinement during the repository thermal peak, but it may be interesting to increase the overpack lifetime by protective coatings. Due to their physico-chemical properties and corrosion resistance, ceramics such as zirconia (ZrO₂) can be candidates for such coatings. However, fabrication of oxide coatings has long relied on processes using hazardous or polluting solvents. This limitation can be overcome by techniques such as physical vapor deposition (PVD), which have been spurred by the recent development of High-power impulse magnetron sputtering (HiPIMS). Therefore, the goal of this study was to assess the protective efficiency of C-steel protection against corrosion by PVD-deposited zirconia coatings.

A ferrite-perlitic C-steel (P285GH) was used as a substrate. The surfaces were polished, degreased, and etched for 10 minutes in the PVD setup to clean the substrate. ZrO₂ ceramic coatings were fabricated by reactive PVD-HiPIMS using a zirconium target. The total deposition time, the duration of HiPIMS impulsion, the power density at the target surface and the target-to-substrate distance were varied. Characterization by X-ray diffraction, scanning electron microscopy and Raman microscopy showed that the coatings were homogeneous, relatively dense and composed of monoclinic zirconia forming columns. They were adherent over the entire surface, and displayed only few defects. Variations in the deposition parameters resulted in differences in crystalline properties and coatings thicknesses (between 5 and 10 μm).

The corrosion behavior of these coatings in reference (borate buffer) and mildly saline (40 mM NaCl) solutions were assessed in a three-electrode setup (Hg/HgSO₄ reference electrode, Pt mesh counter electrode) by monitoring of the open circuit potential (E_{OCP}) and by anodic polarization. Potentials are quoted against the Normal Hydrogen electrode (NHE). The E_{OCP} values measured were generally within ± 0.1 V of the value for bare steel (-0.56 V /NHE), but the corrosion currents were significantly lower, by one to three decades (0.2-5.8×10⁻⁸ versus 2.7×10⁻⁶ A/cm², respectively, in 40 mM NaCl). Immersion experiments over three weeks showed that significant variations in the corrosion current occurred, possibly due to corrosion and scaling within coating defects. Overall, however, the corrosion current remained lower than the value measured after one day. Additional experiments over longer timescales, at higher temperatures, and in synthetic clay porewater would aim to further assess the relevancy of such protective coatings.