Cold Spray Coating Technique for Mitigation of Chlorine-Induced Stress Corrosion Cracking (CISCC) in Stainless-Steel Dry Storage Canister for Spent Nuclear Fuel

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1. Introduction

There are two types of interim storage facilities to store spent nuclear fuel. One type is a wet storage system, and the other one is a dry storage system. Because the current wet storage pool has limited spaces for storing spent nuclear fuels, spent nuclear fuels need to be stored in dry storage facility after several years of cooling in wet storage pool, cooled by air for several decades until disposal.

In the last quarter of 2023, 22,522 light water reactor spent nuclear fuels are storing in wet storage pools. This amount is almost 75.4% of storage capacity of 29,872 fuel assemblies [1]. Compared with the first quarter which usage was 73.8%, with 22,033 stored out of same capacity, it can be seen that its reserve capacity is rapidly shrinking.

Because there are no dry storage facilities for light water reactors except for Wolsung heavy water reactor in South Korea, securing storage capacity of spent nuclear fuels using dry storage system is an urgent issue for our society. However, for safe and long-term storage of spent nuclear fuel in a dry storage facility, a special type of corrosion called chlorine-induced stress corrosion cracking (CISCC) should be considered. This phenomenon impairs the sealing of the storage canister, and makes radioactive materials leak out to the environment. Yet, there is only little research of possibility of corrosion and sealing failure of the canister by chlorine-induced stress corrosion cracking in the environment of South Korea.



2. Chlorine-Induced Stress Corrosion Cracking

Fig. 1 The mechanism of CISCC [2].

Chlorine-induced stress corrosion cracking (CISCC) in austenitic stainless-steel 304/304L widely used for dry storage canister worldwide is mainly occurred by the combination of three conditions. First is the segregation

of chromium in the material to prevent formation of passivation film. This elemental segregation is induced by heat under melting temperature, and the major provider of this heat is a fusion welding. In the vicinity of welding regions, its temperature is slightly lower than melting temperature while this makes chromium to segregate from stainless-steel as chromium carbide, combining with carbon. It is called sensitization, and the area in which sensitization occurred is called Heat Affected Zone (HAZ). This sensitization depletes chromium from the material, suppressing the formation of chromium passivation film.

Second is the chlorine-rich, corrosive environment. Chlorine tends to take an electron from adjacent atoms and becomes chloride ion (Cl⁻) because of its high electronegativity. In other words, chlorine has a tendency of corrode other materials. Generally, stainless-steels have superior resistivity of corrosion due to formation of chromium passivation films. However, as mentioned, stainless-steel can lose the corrosion resistance by sensitization. As a result, the surface of stainless-steel can corrode accompanied by pitting corrosion. These pits will become the initiation point of CISCC.

The last one is a residual tensile stress. The material experiences inherent tensile stress during fusion welding. After welding, the molten stainless-steel will solidify again as it cools, and the shrinkage during phase transition will induce a high level of tensile stress between the welded sides. This residual tensile stress of the material will tear the pits and make it into cracks.

3. The mitigation of CISCC

To restrain the occurrence of CISCC, the suppression of at least one condition of the CISCC is required. However, there is still no treatment available that can restore sensitization of materials other than heat treatment. This sensitization is less likely to happen to some special austenitic stainless-steel such as 347 stainless-steels, which contains niobium that prevents the precipitation of chromium carbide. Nevertheless, these stainless-steel are not commonly used because of its cost and accessibility.

Therefore, preventing CISCC by restoring material sensitization is not reasonable. Instead, relieving residual tensile stress of the material or forming a physical barrier between substrate and corrosive environment can be reasonable options. The alleviation of residual tensile stress is achieved by inducing severe plastic deformation (SPD) on the material surface. SPD induces a new compressive stress to the material, which will compensate the residual tensile stress that pulls the material to each other. Certainly, residual tensile stress also exists at the internal of the material, but the effect is relatively small and less considered because there are fewer stress concentration points such as corrosion pits than the surface.

Forming a physical barrier for the material is a widely used method for preventing unwanted corrosion. Representatively, painting on the metal surface is a one of the examples. But for dry storage canister, long-term durability is an important criterion because its high heat and radiation limit further repair. For this harsh environment, there are several researches of various surface coatings such as ceramics, polymers, and metal coating [3].



Fig. 2 A diagram of cold spray coating.

Cold spray deposition is a promising technique that can not only be a protective coating, but also relieve residual tensile stress at once. This technique accelerates a feedstock metal powder via inert propulsion gas such as nitrogen or helium, and collides it with a substrate. By this, the micron-scaled feedstock metal particle deforms and sticks to the surface, forming a high density, solid coating.

Also, while in this process, the impact of particles hitting the substrate develops SPD which applies compressive force to the surface. As mentioned, this compressive force counteracts with the tensile residual stress which already exists on the material. As a result, two conditions of CISCC are removed at once through this technique.

Nonetheless, cold spray coating is not a cure-all for potential corrosion of a dry storage canister. An intrusive corrosion between cold spray coating layer and the substrate which is called crevice corrosion can be occurred when its coating edge doesn't perfectly blend with the material. In this situation, there is no any physical barrier to block deposition of chloride ions and the alleviation of residual tensile stress is insufficient to prevent pit-to-crack transition. As a result, chloride ions will deposit in the gap, causing severe oxidationreduction reaction which leads to crevice corrosion. It depends on the metal composition, but normally the gap size which crevice corrosion can be occurred (Crevice Critical Gap Size, CCGS) is between $0.1 \mu m$ to 0.1 mm [4].

4. Conclusion

The management of spent nuclear fuel is an emerging topic for sustainability of nuclear power plants. South Korea is now using wet storage pools for interim storage of spent nuclear fuels. However, because the capacity of the wet pool is not enough to store all the spent nuclear fuel produced near future and the construction of new wet pools is much harder than the dry storage facility, interim storage of spent nuclear fuels in dry storage facility is inevitable. Dry storage system is much easier to expand its capacity due to its structure, but it also has a concern of potential corrosion and cracks, represented by CISCC.

This stress corrosion cracking can be mitigated by suppressing one or more conditions. A promising mitigation technique of CISCC is cold spray coating, which accelerates small metal powders to supersonic speed and makes them collide with the surface. By this deposition, metal feedstock powders will stick to the surface and build a physical, rigid barrier that isolates the susceptible material from external corrosive environments. While the metal particle collides with surface severely deforms both the metal particle and the substrate, compressive stress counteracting with residual tensile stress introduces

This treatment is effective to preventing potential CISCC occurrence on HAZ, but more researches about crevice corrosion between coating edge and the substrate are required. Furthermore, investigation of the difference of salt composition between nuclear power plant sites is needed because it can be an important variable of CISCC initiation and propagation.

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