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Development of Ceramic Humidity Sensor for the Korean Next Generation Reactor

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Abstract

Leak-before-break(LBB) approach has been shown to be both cost and risk effective by reducing maintenance cost and occupational exposure when applied to high energy piping in nuclear power plants. For Korean Next Generation Reactor(KNGR) development, LBB is considered for the Main Steam Line(MSL) piping inside containment. Unlike the reactor coolant piping leakages which can be detected by particulate and gaseous radiation monitoring, main steam line leak detection systems must be based on principles that do not involve radioactivity. Ceramics are widely used as humidity sensor materials which can be further developed for nuclear applications. In this paper, we describe the progress in the development of ceramic humidity sensors for use with the main steam lines of KNGR.

I. Introduction

All nuclear power plants designed prior to 1983 were required to take into account the dynamic effects by postulating circumferential and/or longitudinal ruptures in main coolant loop(MCL) and branch line piping(BLP)[1]. As results, these piping systems were required to be installed with various protective measures such as pipe whip restraints, snubbers, jet deflectors, jet impingement shields, and oversized component supportors[1]. But these devices may develop the potential for unintended thermoelastic restraint to piping, the accessibility problems associated during in-service inspection and maintenance, the increased radiation exposure to workers, and the loss of plant thermal output. Furthermore, the protection structures involve components and space that are costly to install and maintain.

In early 1980's, the U.S. N.R.C. performed studies on the possibility of a double-ended guillotine break(DEGB) or equivalent of the primary piping systems, and formulated the LBB approaches and acceptance criteria which culminated by issuing NUREG-1061 Volume 3[2]. From this, LBB applications were made by utilities as the request for exemption from the DEGB requirement on a case by case basis.

According to NUREG-1061, the following criteria must be satisfied in the piping systems to qualify for **LBB** application[1,2] ;

(1) Three independent leak detection systems are required and each with three redundancy should be capable of detecting a leakage rate less than 1.0 gallon per minute for the primary system.

(2) Throughwall leak size cracks(LSC) which are large enough to leak 10 gpm(NUREG-1061 applies a safety factor of 10) must be stable for the postulated load equal to $\sqrt{2}$ times the sum of the normal operation and safe shutdown earthquake(SSE) loads.

(3) Throughwall cracks twice as long the LSC must be stable under the sum of normal operation and SSE loads.

It is clear that leak detection capability is a critical factor in order to apply the LBB concept successfully to a plant. To date, LBB has not been applied to MSL. Hence there is no established leak sensors for MSL. After reviewing various leak detection system which is applicable to MSL within a containment of nuclear power plants, we have determined to develop a ceramic humidity sensor, as described herein.

II. Review of leak detection system for LBB application

Current leak detection systm used in Korean nuclear power plant

In YGN 3&4, to detect main coolant leakage, several containment parameters are monitored. Three essential leakage detection systems are containment atmosphere particulate radioactivity monitoring system, containment sump level and flow monitoring system, the containment atmosphere gaseous radioactivity monitoring system. But radioactivity-based systems can not be applied to main steam line due to extremely low radioactivity in secondary water. Hence we need to develop two additional leak detection systems for MSL. Humidity sensors have been used for leak detection in nuclear power plants. Siemens-KWU has applied FLÜS, a humidity sensor with air sampling scheme to a nuclear plant in Slovak[3]. CANDU employs a humidity sensor, beetles, for pressure tube leak detection.[4]

LBB application criteria

a. LBB analysis

Analysis on LBB applicability takes the following steps in accordance with SRP 3.6.3[5] and NUREG-1061[2]. The first step is screening against material degradation mechanisms including creep, corrosion, stress corrosion cracking, indirect causes and cleavage. In the second step, pipe loads should be determined as the sum of normal operation and SSE loads. Permitted leak limit in LSC determination is 10 gpm in the case of the identified leak, and 1 gpm in the case of the unidentified leak[6]. The final step is the crack stability evaluation based on the elastic-plastic fracture mechanics, generally conducted by J-T method[5].

b. Leak detection system requirement for the main steam line

Main steam lines are operated at 73 atm. and 290℃ in KNGR design. The tentative requirements of the leak sensors for MSL are established as follows;

(1) Leak detection system should be able to detect 1 gpm in the identified leak, and 0.1 gpm in the unidentified leak.

(2) The leak detection systems must have three diversity, each with three redundancy.

Among the present systems, containment sump level could be shared for both MCL and MSL on a conservative basis. Therefore at least two additional detection systems are required to satisfy the requirements. The fact that humidity increases with leak rate has allowed the use of humidity sensors for both the leak identification and leak rate estimation with a good sensitivity. Hence we have identified and developed ceramic humidity sensor for MSL application.

III. Development of the ceramic humidity sensor

Sensor material survey

Functional ceramic materials, which are superior in thermal stability, physical strength and chemical resistance as well as the electrical properties, are widely used to satisfy diverse needs for sensing devices. Inside the containment of the KNGR, MSL is insulated with asbestos. Since the proximity of sensor to MSL is desired for high sensitivity, ceramic materials are assumed to be most suitable. We later found that certain types of ceramic humidity sensors are actually used in the Japanese nuclear power plant[7]. After reviewing of various ceramic materials with humidity-sensitive characteristic, $MgCr_2O_4$-TiO_2 is identified as a candidate material for the environment in the insulation of MSL.

Humidity detection principle

For a ceramic humidity sensor material, $MgCr_2O_4-TiO_2$, its electrical conductivity increases upon adsorbing water molecule[8]. The mechanism of conduction in $MgCr_2O_4-TiO_2$ involves several microprocesses. First, a few water vapor molecules chemisorb on the grain surface by a dissociative mechanism to form two surface hydroxyls per water molecule; one hydroxyl ion adsorbed on a surface metal ion and the other hydroxyl ion from the proton combined with an adjacent surface oxygen ion. The hydroxyl groups, then, dissociate to provide mobile protons. They can migrate by hopping from site to site across the surface. Chromium is believed to be the most active surface metal ion since the Cr^{3+} ion combines and dissociates easily, providing mobile protons from the transition of Cr^{3+} ion to Cr^{4+} in the p-type semiconductors. In addition, some water vapor can adsorb physically on the surface with mobile proton to form a H_3O^+. The hydromium ion can further increase the conductivity due to its mobility. When molecular water is abundant, however, H_3O^+ ion will be hydrated and stabilized leaving the proton as the dominant chage carrier.[8,9]

III. Experimental procedure

In the process of sintering $MgCr_2O_4-TiO_2$, raw materials used in preparing the specimens were MgO, Cr_2O_3, and TiO_2. The raw materials were milled with a ball mill in ethanol for 24 hours. After drying, calcination was carried out at 1100℃ for 10 hours. After calcination, the powder mixture was sieved through 150 μm and 250 μm mesh respectively. It is advantageous to have a range of granule size, since the smaller particles will tend to fill the interstice openings between the larger particles[10]. Then the powder mixture was pressed into a rectangular form at a pressure of 32 MPa, and with cold-isostatic pressing(CIP) at a pressure of 100 MPa. After forming, $MgCr_2O_4-TiO_2$ was sintered in air at 1360℃, 1400℃, 1440℃, respectively, for 10 hours.

Fig. 1 Fig. 2

Fig. 1. SEM photograph of as-sintered surface of $MgCr_2O_4$-TiO_2

Fig. 2. SEM photograph of fracture section of sintered $MgCr_2O_4$-TiO_2

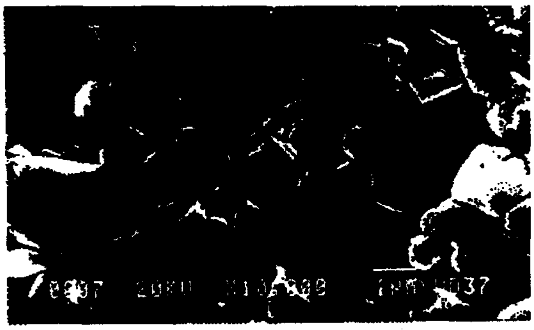

Fig. 3. SEM photograph of thermally-etched surface of sintered $MgCr_2O_4$-TiO_2

Results

The crystal structures of the sintered ceramic material were identified at room temperature by standard X-ray diffraction(XRD) techniques. XRD analyses showed that, the system $MgCr_2O_4$-TiO_2 was single phase with a pure $MgCr_2O_4$-type spinel structure. The density of the $MgCr_2O_4$-TiO_2 sintered at 1400℃ was measured to be 84.74±0.07 % of theoretical density by Archimedes method. The sintered compact shows a typical porous structure, as shown in Fig. 1. When the ceramic was mechanically fractured, its surface exhibits intergranular pores, as shown in Fig. 2. Thermally etched surface shows more tight microstructure, as shown in Fig. 3.

Preliminary measurements have been made to characterize the conductivity change with humidity. A static 1-liter autoclave was used as an environmental chamber for which temperature and humidity were controlled. Four-probe method is used with a Keithley Model 617 electrometer. The conductivity in vacuum at 100℃ was observed to be nearly constant over a range of applied current, as shown in Fig. 4. Fig. 5. shows the comparison of conductivity transient at 100℃ in vacuum, dry air, and humid condition. In the humid environment, the conductivity of the ceramic sensor material increased by a factor of about four.

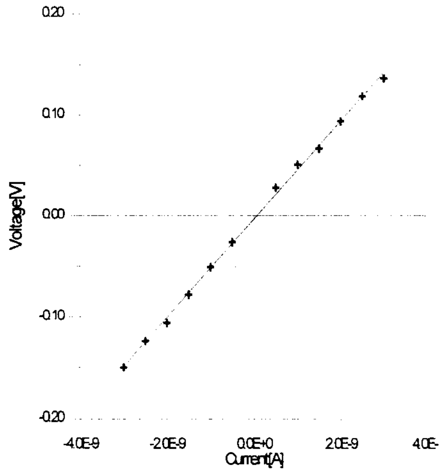

Fig. 4. Measured conductivity (dry air)

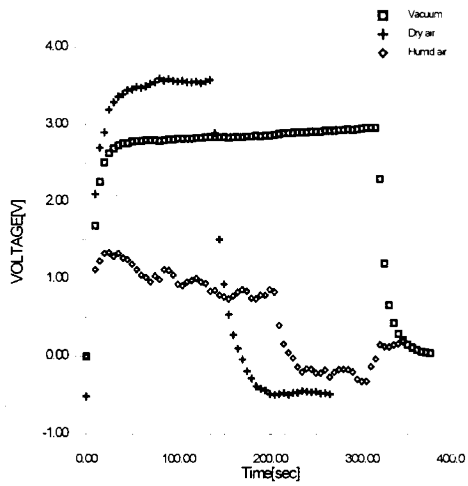

Fig. 5. Measured conductivity change (T=100C, I=1nA)

IV. Conclusion and future work

For LBB application to MSL of KNGR, ceramic-based humidity sensors are suggested as a leak detection system with adequate thermal, electrical, and mechanical properties. The developed humidity sensor material, $MgCr_2O_4$-TiO_2, exhibits a single-phase solid solution with a pure $MgCr_2O_4$-type spinel structure. The preliminary measurement of conductivity showed that the sensor has humidity-sensitive characteristic. But further characterization of the electrical properties of $MgCr_2O_4$-TiO_2 and humidity response are in progress.

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