Radiation Damage and Activation of Nickel-Alloys in Molten Salt Reactor Core Environments

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1. Introduction

The development of Molten Salt Reactor (MSR) technology is actively underway over the world. As a variety of MSR design concepts are considered depending on the situation of each country, Korea is developing MSRs that use a liquid fuel in the form of chloride salt and fast neutron spectrum. The MSR attracts interest because it provides a number of benefits, including passive safety, atmospheric pressure operation, high thermal efficiency, *etc*. However, the corrosion of structural materials in molten chloride is recognized as a critical concern about the success of MSRs. The oxide surface layer for corrosion protection is generally unstable in molten salts. Because of the harsh service environment for structural materials in MSRs, qualified alloys for a vessel are currently scarce.

During the MSRE (Molten Salt Reactor Experiment) program, which was operated by Oak Ridge National Laboratory in 1962, Hastelloy N (Ni-based alloy) and its variants were tested in the fluoride salts, which was found that there was significant surface cracking [1,2]. In order to overcome material problems in the MSR environments, we are developing a series of new Ni-alloys, which are being tested in chloride salts. In parallel with the corrosion tests, we analytically evaluated the radiation effects of the new alloys. First, the neutron spectra were calculated in the MSR core by employing the transport code. Then using the spectra, the amount of radiation damage and activation of the alloys was estimated. Although the current issue is focused on corrosion of MSR structural materials, the degradation of Ni-alloys by neutron irradiation should not be overlooked.

2. Methods

In estimating the amount of radiation damage and neutron activation of Ni-based alloys, we began to perform the neutron transport calculation to obtain the neutron flux. The flux is a major input to the SPECTER [3] and the NAC code [4]. The materials of our interest are listed in Table 1, where their chemical compositions and weight fractions are given. Two reference alloys - Hastelloy N and modified Hastelloy N, are included in the list, which are commercially available.

Table 1: Chemical compositions of Ni-based alloys (unit: w/o)

Ni-Alloys	Al	Si	Ti	Cr	Mn	Fe	Ni	Nb	Mo	Hf	Ta
M1	-	0.3	0.8	1d	0.3	1.5	66.2	0.8	2d	-	-
A1	1d	0.3	0.4	2d	0.3	2.5	75.2	0.4	1d	-	0.4
Hastelloy N	0.20	0.7	0.2	7.0	0.7	4.0	71.1	-	16.0	-	-
Mod Hastelloy N	0.20	0.1	0.2	7.0	0.2	-	78.7	1.5	12.0	-	-
MW2	0.50	-	1.0	2d	0.3	-	68.0	0.1	1d	0.1	-
MW4	0.50	-	1.0	2d	0.3	-	66.9	0.1	2d	0.1	-

(1d: one digit, 2d: two digits)

2.1 Neutron Transport Calculation

We performed neutron transport calculation using the Serpent Monte Carlo code to obtain the spectrum for the core region of MSR [5]. The core is composed of the tertiary salts of $NaCl-KCl-UCl_3$, which are contained in BeO and MgO reflectors. Figs. 1 (a) and (b) show the geometrical structure of a MSR core – views from the side and top, respectively. Based on this model, the 238-group neutron flux in the core was calculated. The temperature of the fuel salt is assumed to be 900K and the ENDF/B-VII libraries are applied to the calculation.

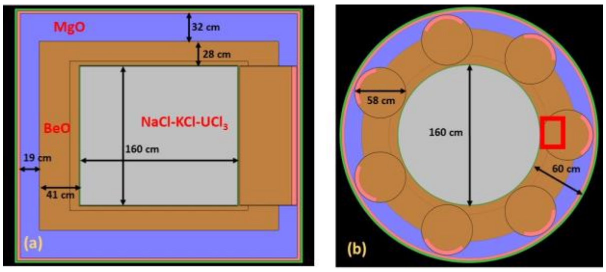

Fig. 1. Cross-sectional views of MSR core: (a) side, (b) top

2.2 Neutron Damage Evaluation

The SPECTER code was employed to obtain neutron damage parameters for a given neutron spectrum. The spectrum is the only input to the code that the user needs to define. The primary damage parameters from the SPECTER calculation include atomic displacement (displacement per atom, *dpa*), He/H production and averaged primary knock-on atom energy spectra. Although the parameter of *dpa* does not indicate the final and permanent damage, it is useful in quantifying the displacement damage without regard to radiation types.

2.3 Neutron Activation Calculation

The process of neutron absorption often results in the formation of an unstable activation product. As the activated products can be gamma-ray source, it is necessary to treat them with care. Since the element Ni is prone to neutron activation for fast neutrons, the NAC code was used to estimate the activity level for Ni-alloys.

3. Results

Neutron spectrum for an active core in MSR is shown in Fig. 2, where that for a baffle in pressurized-water reactor (PWR) was included for comparison. The fast flux is dominant in a MSR core, whereas the thermal flux is negligible. The MSR spectrum is composed of 238-group energy ranging from 1 x 10^{-10} to 20 MeV.

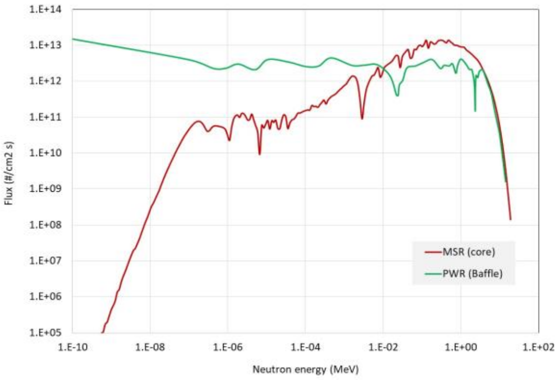

Fig. 2. Neutron spectra for a core region in MSR and PWR

The amount of primary damage due to neutron irradiation was obtained from the SPECTER calculation by assuming that the Ni-alloys were irradiated by the MSR flux (Fig. 2) for 20 years without interruption. The damage parameters are listed in Table 2, including *dpa*, He and H production in units of *appm* (atomic parts per million). It is found that the production of transmutation gas is primarily caused by Ni.

Table 2: Neutron damage parameters of Ni-alloys resulting from MSR neutron flux

	M1	A1	Hastelloy N	Mod Hastelloy N	MW2	MW4
dpa	103.7	119.0	106.7	106.5	104.6	105.2
He (appm)	195.1	196.4	202.1	219.2	199.7	195.1
He(Ni)/Total He, %	*98.4*	*94.9*	*97.2*	*99.1*	*98.3*	*98.3*
H (appm)	2828.2	2788.7	2909.8	3194.0	2907.6	2840.9
H(Ni)/Total H, %	*99.1*	*97.6*	*98.6*	*99.3*	*98.7*	*98.6*

The activity level of Ni-alloys was evaluated using the NAC code. Fig. 3 shows the changes in radioactivity as a function of time right after 20 years of neutron irradiation. After about 30 days of cooling time, the radioactive cobalt elements become a major radiation source, which was derived mostly from the neutron absorption reaction with Ni.

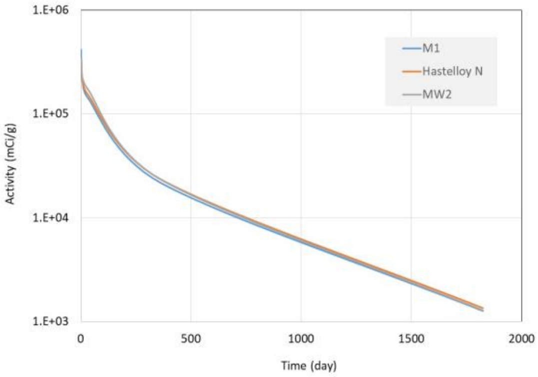

Fig. 3. Changes in radioactivity of activated Ni-alloys as function of cooling time

4. Conclusions

Ni-based alloys are candidate structural materials for MSR, for which resistance to corrosion by molten salt is prime requirement. For this reason, we are currently developing Ni-alloys for the application of a MSR in chloride salt. In parallel with the corrosion test, it is required to evaluate the radiation effect on the Ni-alloys under the MSR core environments. Using the computer codes, we calculated the neutron spectra for the MSR core, and evaluated the radiation damage and the level of activation of Ni-alloys. Although the benefits of Ni elements are being highlighted, their disadvantages must be considered. The followings could be represented from the calculation results.

- As the new alloys contain 66 to 79 w/o of Ni, most of radiation damage and radioactivity after neutron irradiation are determined by the amount of Ni.
- The transmutation gases - H and He from Ni-alloys are primarily produced from the Ni absorption reactions, which is over 90%.
- After one year of cooling, the major gamma sources emitted from the activated Ni-alloys are ^{57}Co and ^{60}Co, which were originated from the reactions with Ni.

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