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Development of Advanced Structural Materials for Future Nuclear Systems in Korea

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Abstract

Korea has put in a huge effort to develop the future nuclear systems such as a sodium-cooled fast reactor (SFR), a very high temperature reactor (VHTR) and fusion reactor. As the realization of such nuclear systems relies heavily on performance of materials, Korea has made steady and persistent effort to develop structural materials that can withstand the extremely harsh operating conditions. The present paper summarizes the current status of the Korean R&D activities on advanced nuclear materials including oxide dispersion strengthened (ODS) steels, Ni-base alloys and their ODS variations, reduced-activation ferritic-martensitic (RAFM) steels, and SiC composites.

1. Introduction

Future fission and proposed fusion reactor systems will be dependent increasingly on advanced structural materials to reliably deliver high performance with favorable safety attributes and acceptable economic cost. Currently, Korea has a plan to develop SFR and VHTR as the Gen-IV reactor systems and the corresponding structural materials are under development. The R&D program on the SFR technology development was initiated in 2007. The proto-type SFR (100 MWe) is scheduled to operate from 2028. The R&D program on a VHTR technology development was initiated in 2006, and the demo-type VHTR is planned in the future. During that time, researches on Fe-base ODS alloys, Ni-base ODS alloys, Ni-base alloys and SiC composites are vigorously carried out and manufacturing and joining technologies are simultaneously studied. Korea also joined the international thermonuclear experimental reactor (ITER) program for the nuclear fusion system development and has started
RAFM steel development for TBM application in 2012. The representative alloy is named as advanced reduced-activation alloy (ARAA) and the development will be completed by 2016. Here, the current status of high temperature alloy development for Gen-IV and ITER applications in Korea is summarized.

2. Development of Fe-Base ODS alloys and Joining Technology

Fe-base ODS alloys are being considered a prospective candidate material of in-core structural components such as cladding tube, wire and duct in the SFR [1,2]. Fe-base ODS alloys with a ferritic-martensitic structure have an excellent irradiation resistance to a void swelling as well as superior creep strength at elevated temperatures. We have developed the Fe10Cr-base ODS alloys and their out-of-pile tests are in progress. Shown in Fig. 1 are the tensile properties of the Fe10Cr-base ODS alloys at room temperature and 700°C. The yield strength of the alloy at room temperature varies depending on the heat treatment, the minimum value being ~630 MPa. At 700°C, the yield strength drastically decreases but is still higher than 240 MPa with an elongation of ~10%. Such properties are regarded to be adequate for SFR applications, and an evaluation of mechanical materials characterization and the out-of-pile test for the tube are under progress.

Only few efforts have been made for improving joining technologies. It is well known that uniform nano-oxide dispersoids act as pinning points to obstruct dislocation and grain boundary sliding in Fe-base ODS alloys. However, such advantages will disappear due to the coarsening and segregation of oxide particles when Fe-base ODS alloy is exposed to the temperatures higher than its melting point during the welding [3,4]. Therefore, we currently consider two different solid-state joining process for Fe-base ODS alloys, friction stir welding (FSW) and magnetic pulse welding (MPW). During the FSW, extremely large deformation and frictional heat are generated by the rotational behavior of the tool and accordingly plastic deformation and recrystallization simultaneously affect soundness of welds. During MPW, an eddy current is generated by a high-density magnetic flux and, as a result, strong repulsive force is created around the weldments. This force can drive the materials together at an extremely high rate of speed and creates an explosive or impact type of weld. The results of joining between Fe-base ODS alloy cladding tube and ferritic-martensitic steel end-plug are shown in Fig. 2. The outer diameter and the wall thickness of the tube are 7 mm and 0.5 mm, respectively, and those small pieces were successfully joined by FSW. It was confirmed that sound welds have been made with no defect, the uniform oxides dispersion have been preserved in the materials after joining.

3. Development of Ni-Base ODS alloys

A preliminary work has been performed to evaluate the feasibility of developing Ni-base ODS alloys as structural materials for the VHTR applications, in which mechanical properties and microstructure of the conventional Ni-base alloys (Alloy 617 and Alloy XR) and the ODS counterparts are compared. For both Ni-base ODS alloys, yttria (Y₂O₃) particles are used as dispersion strengthening medium. The Ni-base ODS alloys were produced by mechanical milling of the elemental powders with target compositions (Ni-22Cr-12Co-9Mo-1Al-0.6Y₂O₃ and Ni-22Cr-18Fe-9Mo-0.6Y₂O₃ for Alloy 617 and Alloy XR, respectively), mechanical alloying, hot isostatic pressing (HIP) at 1200°C, followed by hot rolling at 1050°C to 60% reduction in thickness. In both alloys, addition of yttria particle results in a significant refinement of grain structure (Fig. 3), which is attributed to the pinning of grain boundaries by the finely dispersed yttria particles. This in turn leads to a remarkable enhancement of yield strength. For example, the yield strengths of the ODS Alloy 617 at room temperature and 700°C are 650 MPa and 480 MPa, respectively, which are ~106% and 84% enhancements compared with those of conventional alloy. These results suggest a possibility of application of Ni-base ODS alloy for high temperature applications.

4. Development of Ni-Base alloys

In order to improve the high temperature performance of the commercial Ni-base alloys, an attempt was made to develop new Ni-base alloy. The NiCrCoMo-base alloys were designed and produced by conventional procedures including vacuum induction melting, homogenization heat treatment, hot rolling and solution treatment. The solution treated plate were subjected to an additional heat treatment to strengthen the grain boundaries through
carbide formation along the grain boundary. Presented in Fig. 4 are the stress-strain curves of the NiCrCoMo-base alloys, which shows the effects of additional heat treatments on the tensile properties at 950°C. In the case of additionally heat treated specimens, the yield and tensile strengths are similar, irrespective of the heat treatment condition. The ductility increased from 15% to 48% with the heat treatment temperature and then drastically decreased at 1140°C almost to the ductility of the solution annealed specimen. From the experimental results, it is reasonable that the grain boundary is effectively strengthened up to 1110°C. It seems that the abrupt decrease at 1140°C is related to a loss of carbide stability at the grain boundary. Mo was beneficial to high-temperature ductility while Cr was detrimental to high-temperature ductility. This was differentiated from the carbide composition. Co modified carbide composition providing the synergic effect of Mo and Co on mechanical property at 950°C. A high-temperature ductility of 76% was achieved by combination of alloying element and heat treatment without a significant loss of yield strength or tensile strength, which is comparable to commercial Ni-base alloy. However, it was found that there is a drawback on the corrosion rate, which should be improved.

5. Development of Structural Materials for Fusion Reactor Applications

An effort has been made to develop structural materials for the helium cooled ceramic reflector (HCCR) test blanket module (TBM) application in ITER under the framework of ITER TBM program. The program aims specifically at developing the Korean RAFM steel (named as ARAA) with mechanical properties (esp., impact and creep resistance) comparable or better than those of the conventional RAFM steels such as EUROFER97 and F82H [5,6]. The program is now on its first stage (2012-2013) during which alloy design, fabrication and evaluation of physical (elastic modulus, thermal conductivity, etc.) and mechanical properties (tensile, impact, creep and fatigue, etc.) of unirradiated materials are scheduled to be complete. Alloy design with two different strategies has been made: one is modification of the amounts of alloying elements such as C, Cr, W, V, Ta, N and Ti, and the other is to examine the potential role of alloying elements, e.g., Zr, that have rarely used in the conventional RAFM steels. With these strategies, a total of 73 alloys were designed and fabricated, and the mechanical properties of which has been evaluated. Process design has been made to optimize heat treatment (i.e., normalizing and tempering) conditions for new alloys. An attempt has also been made to employ a thermo-mechanical treatment during normalizing for further improvement of mechanical properties.

Shown in Fig. 5 is an example of mechanical properties, represented by creep rupture time and the ductile-brittle transition temperature (DBTT), of the program alloys. It is noted that alloys with the trace amounts (below 0.02 wt.%) of Zr, when tempered under the optimized condition, exhibit superior resistance to both creep and impact, compared with those of conventional RAFM steels. In the second stage of the program (2014-2016), two or more of the program alloys will be selected, for which 5-ton scale heats will be prepared and fabricated into plates with various thicknesses by POSCO Specialty Steel (Changwon, Korea). The plates thus prepared will be used to optimize the processing variables that vary depending on the size of products, and to evaluate mechanical properties for assessing reproducibility. A part of the tempered plates will be utilized for irradiation and post-irradiation evaluation (PIE): currently neutron-irradiation up to 3 dpa in High-Flux Advanced Neutron Application Reactor (HANARO) is scheduled. Based on the results of the irradiation tests, the final candidate will be chosen and produced on a large scale, which in turn will be utilized for fabrication of the HCCR TBM.

6. Development of Fabrication Technology of SiC Composite

The triple-layered (triplex) structure of SiC fiber-reinforced SiC matrix (SiCf/SiC) composite tube is one of the concepts of advanced nuclear fuel claddings with a superior accident tolerance, being currently of great interest worldwide [7,8]. The innermost layer is a high-density monolithic chemically vapor deposited (CVD) SiC to insure a gas tightness and the second layer consists of SiCf/SiC composite to increase a mechanical property and prevent a brittle fracture of the composite tube. Another CVD SiC is finally coated to protect the relatively porous composite layer containing a pyrolytic carbon (PyC) interphase from corrosion. Fig. 6 shows the SEM micrographs of the CVD SiC tube coated on the graphite rod substrate and the cross-section of the triplex SiC composite tube. The CVD SiC was coated with a very uniform thickness throughout the whole length and the triplex composite tube was successfully fabricated.

The stoichiometry of SiC is an important parameter to insure the irradiation tolerance of SiC ceramics. The
CVD SiC had a stoichiometric composition without any presence of free Si or free C as shown in Fig. 7. The Raman micro spectroscopy analysis shows only SiC peaks, transverse optical (TO), longitudinal optical (LO), and second order SiC bands. The X-ray photoelectron spectroscopy (XPS) analysis also confirms a stoichiometric SiC except for the Si-O peaks originated from the native oxide layer. Fig. 8 shows the cross-sectional microstructure of the CVD SiC and the nano-indentation hardness along the growth direction. The CVD SiC has a random texture with a small grain size at the early stage of the CVD process and a columnar growth occurs with an (111) preferred orientation as the thickness increases. The hardness increases as the preferred texture develops because the hardness shows a maximum value when the SiC has the (111) preferred orientation [9].

7. Summary

The R&D activities on high temperature alloys for future nuclear applications could be summarized to realize the Gen-IV reactor systems with enhanced economics and improved safety. The overall road map for the development of advanced nuclear materials coincides well with the R&D programs for Gen-IV reactor systems. Main target materials are ODS steels as in-core structural materials for SFR and Ni-base alloys and their ODS counterparts for VHTR, and SiC composites for general high-temperature structural applications. A long-term program to develop structural materials for ITER TBM applications has been formulated, and now the representative alloy, named as ARAA, has been developed, for which a large-scale production and the out-of-pile test are in progress.

Reference


Figures

![Fig. 1 Tensile properties of the Fe10Cr-base ODS alloy as a function of heat treatment condition at (a) room temperature and (b) 700°C.](image-url)
Fig. 2 Appearance of the FSW joint between the Fe10Cr-base ODS alloy and ferritic-martensitic steel end-plug.

Fig. 3. SEM micrographs of solution treated (a) conventional Alloy 617 and (b) the ODS counterpart. Finely dispersed yttria particles, a TEM micrograph of which is inserted in (b), leads to a significant refinement of grain structure.

Fig. 4. Stress-strain curves of the NiCrCoMo-base alloy at 950°C in air obtained for heat treated specimens after solution annealing as a function of subsequent heat treatment temperature.
Fig. 5. Plots of short-term creep rupture time against the DBTT of the Korean RAFM steels. The effects of tempering time on the properties are also seen from the plot. Note that all the alloys shown contain Zr and highlighted yellow is the properties of conventional RAFM steels reported in literature.

Fig. 6. SEM micrographs of (a) the CVD SiC inner tube and (b) the cross-section of the triplex SiC composite tube.

Fig. 7. Results of (a) Raman micro spectroscopy and (b) X-ray photoelectron spectroscopy analyses on the CVD SiC tube.
Fig. 8. Microstructure and nanoindentation hardness of the CVD SiC tube.