High-throughput design of low-activation, high-strength creep-resistant steels for nuclear-reactor applications

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ABSTRACT

Reduced-activation ferritic/martensitic steels are prime candidate materials for structural applications in nuclear power reactors. However, their creep strength is much lower than that of creep-resistant steel developed for conventional fossil-fired power plants as alloying elements with a high neutron activation cannot be used. To improve the creep strength and to maintain a low activation, a high-throughput computational alloy design model coupling thermodynamics, precipitate-coarsening kinetics and an optimization genetic algorithm, is developed. Twelve relevant alloying elements with either low or high activation are considered simultaneously. The activity levels at 0–10 year after the end of irradiation are taken as optimization parameter. The creep-strength values (after exposure for 10 years at 650 °C) are estimated on the basis of the solid-solution strengthening and the precipitation hardening (taking into account precipitate coarsening). Potential alloy compositions leading to a high austenite fraction or a high percentage of undesirable second phase particles are rejected automatically in the optimization cycle. The newly identified alloys have a much higher precipitation hardening and solid-solution strengthening at the same activity level as existing reduced-activation ferritic/martensitic steels.

1. Introduction

Reduced-activation ferritic/martensitic steels belonging to the so-called 9Cr family are potentially suitable materials for first wall and blanket structures of fusion power reactors [1,2]. For nuclear applications, the composition of these steels are to be modified from existing creep-resistant steels used in fossil-fired power plant applications. In addition to the harsh thermal and mechanical conditions in fossil-fired power plants, materials in critical nuclear-power-plant applications also suffer from the effects of neutron fluxes [3], which will activate elements directly or induce radio-activity by transmutation of alloying elements [4]. This causes a great problem in the disposal of waste materials at the end of their working life. To create reduced-activation ferritic/martensitic steels which are either not activated or if activated decay in a short time, low-activation elements are selected to replace the high-activation elements in the conventional ferritic/martensitic steels for non-nuclear applications. For instance, Mo is replaced by W; Nb is replaced by Ta, and alloying elements with a very high activation such as Cu, Ni and Co are avoided [5–9].

The lowering of the concentrations of the alloying elements contributing to the solid-solution and precipitation hardening however will have a strong and negative effect on the creep strength of such steels. The common strengthening precipitates in existing 9Cr reduced-activation ferritic/martensitic steels are $M_{23}C_6$, Laves phase and MX carbonitrides (TaCN and VCN). The $M_{23}C_6$ and Laves phase precipitates have much higher coarsening rates than the MX carbonitrides [10–14]. A high coarsening rate causes a relatively fast and strong decrease in precipitation hardening with time and is detrimental to the creep behaviour. Hence MX carbonitrides with their low coarsening rate are the main strengthening precipitate in reduced-activation ferritic creep-resistant steels. Recent work on the stability of MX carbonitrides showed that TaC has the highest stability of all MX precipitates followed by TaN and VN [15,16]. Therefore, TaC can be a promising candidate precipitate for nuclear materials. In contrast to precipitation hardening, solid-solution strengthening in high-temperature steels is in principle time-independent once the matrix reaches its equilibrium composition. W is the most common solid-solution-

strengthening element in reduced-activation steels and shows significant strengthening effects [17] in the initial stage. However, W originally in solid solution will be consumed in the formation of Laves phase precipitates (Fe_2W), lowering the solid-solution-strengthening effect. New alloying elements with a high solid-solution-strengthening factor and a low activation are required to replace W.

For the reasons outlined above, existing 9Cr reduced-activation ferritic/martensitic steels, such as F82H, Eurofer, ORNL 9CR-2VTa and CLAM, have a lower creep strength than high-end creep steels, e.g. P91 and P92, developed for conventional power plants [5]. It remains a challenge to obtain an optimal composition leading to a low activation and a decent creep strength at the same time. In previous studies, we have developed a computational alloy design approach which optimizes both solid-solution strengthening and precipitation strengthening in creep-resistant steels for fossil-fuel-power-plant applications by tuning more than 8 alloying element simultaneously [18–20] and taking into account the intended use temperature. In the present study this approach is further extended by considering the activation as a new optimization parameter in the design of new reduced-activation steel for nuclear applications with optimal combination of precipitation hardening and solid-solution strengthening. The optimization is done for a fixed high use temperature of 650 °C, in line with the targets set in the design of new power plants. The differences in composition and properties of the conventional and the reduced-activation steels are discussed.

2. Model description

As described in the introduction, a high creep strength can be achieved by a uniform distribution of fine TaC with an extremely low coarsening rate in a solid-solution-strengthened martensitic matrix. In addition, it is of great importance to have decent corrosion and oxidation resistances, which can be realized by ensuring a sufficient Cr content in the matrix upon the completion of the precipitation reactions. These features can all be achieved by using the computational alloy-design approach coupling a genetic optimization framework.

The alloy-design approach is summarized as follows. First, the genetic algorithm generates random 'solutions', i.e. combinations of composition and heat treatment parameters selected from the search space defined in Table 1. Per element or heat treatment temperature the search space is defined at 32 equidistant levels between the lowest and the highest value indicated.

Then, the algorithm checks whether for this candidate solution the desired microstructure can be obtained, using six go/no-go criteria defined in the order of the thermal treatments to which such steels intended for long life-high load applications are to be subjected. The typical heat treatment for martensitic creep-resistant steels starts with an austenisation/solution treatment, followed by quenching and a tempering treatment to form desirable precipitates in the martensitic matrix. In the present study the tempering temperature is set equal to the service temperature (650 °C) since the installations in which the steels are used have a very long service time and thus reach the equilibrium at the service temperature. At the (variable) austenisation temperature, thermodynamic calculations are performed and three go/no-go criteria

are defined: (1) the equilibrium volume fraction of austenite should be no less than 99%; (2) the amount of primary carbides should be no more than 0.5% in volume; (3) liquid and delta phase should be absent. To ensure that the austenite transforms completely to martensite during quenching to room temperature, a fourth go/no-go criterion is imposed: (4) the Martensite start (T_{Ms}) temperature should be higher than 250 °C. After quenching, the alloy will not experience any higher temperatures than the service/ageing temperature. Two additional go/nogo criteria at this use temperature are enforced: (5) the amount of precipitates other than TaX carbonitrides should be less than 1 vol%; (6) The equilibrium Cr concentration in the matrix should at least be 9 wt.% to assure decent corrosion, oxidation resistance and a high toughness at low temperature [21].

Solutions that meet the six go/no-go criteria will be further compared and ranked using three optimization criteria, which are activity, Precipitation Hardening (PH), Solid Solution Strengthening (SSS) respectively. These optimization factors are introduced below.

2.1. Activation

The activation R for each alloying element was calculated using Ref. [22].

$$R = \phi\sigma = \phi_0\sigma_0 + \phi_e I_0 + \phi_f\sigma_f \tag{1}$$

where R is the probability of an atom capturing a neutron per second, ϕ is the overall neutron flux, σ is the effective capture cross section, ϕ_0 is the flux of thermal neutrons, ϕ_e is the flux of epithermal neutrons and ϕ_f is the flux of fast neutrons. σ_0, I_0 and σ_f are the capture cross section for fluxes of thermal, epithermal and fast neutrons, respectively. The three flux levels (values used in the Reactor in Delft [22]) and the irradiation time t_{ir} are listed in Table 2. Other values could have been chosen, but they do not affect the final composition corresponding to a minimum activity much.

The values of σ_0, I_0 and σ_f depends on the neutron activation types, such as (n,γ), (n,α), (n,ρ), (n,n′) and so on and are taken from Ref. [23]. For one alloying element, all reactions may happen simultaneously, but only the one with the highest activity is recorded in this study. The activities also relate to the time scales after irradiation as a result of trans-mutated elements. Their values are calculated by multiplying a factor $e^{(-\ln 2*t_{ir}/t_{1/2})}$ with the immediate activation value, in which t_{ir} is irradiation time and $t_{1/2}$ is half-life. Four times (cooling periods) are chosen in ranging from time scales relevant to maintenance (0 year) to storage and disposal times (1, 5 and 10 years). The activities after each of the four times per gram are calculated according to Equation (1) and considering the transmutation, and results are listed in Table 3 for all the elements. C and N are considered to be non-activated for any of the

Table 2
The set of irradiation conditions imposed.

Thermal neutron flux (ϕ_0)	(m^{-2} s^{-1})	5.0E+17
Epithermal flux (ϕ_e)	(m^{-2} s^{-1})	2.5E+16
Fast neutron flux (ϕ_f)	(m^{-2} s^{-1})	3.0E+16
Irradiation time (t_{ir})	(s)	3.6E+08

Table 1
Search ranges (in wt.%) for 12 chemical elements and the austenisation temperature T_{aus}.

	C	Cr	Ni	Ta	Mo	W	Co	Nb	N	V	Mn	Si	Fe	T_{aus}
Min.	0	8.00	0	0	0	0	0	0	0	0	0	0	Bal.	1173 K
Max.	0.15	16.00	5.00	1.00	10.00	10.00	10.00	1.00	0.15	1.00	2.00	2.00		1523 K