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Materials selection for nuclear applications: Challenges and opportunities

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ABSTRACT

We discuss the challenge of selecting materials for nuclear applications and outline the need for comprehensive databases to assist scientists and engineers in choosing materials that meet interdependent physical, chemical, and nuclear criteria. In conventional engineering, chemical and physical properties and the electronic structure of materials are typically the primary considerations; nuclear applications must also consider the nuclear physics characteristics of a material. Development of databases that correlate physical, chemical, and nuclear properties would accelerate and facilitate innovations in nuclear design.

1. Introduction: accelerating nuclear design by expanding traditional materials property maps

Selecting the most suitable material for a specific application is an intrinsic part of design and innovation. An engineering design can only be as good as the materials available to the engineer. Conventional applications are mostly interested in the chemical and physical properties of materials and their electronic structure; nuclear applications must also consider the nuclear physics characteristics of a material. This additional restriction in nuclear materials engineering poses tremendous challenges but also opportunities in material science.

The structural properties of materials must be understood in relation to the applications in which they will be used. Engineers often use materials property maps when selecting a suitable material for an application [1]. These maps describe fundamental material properties, plotted in two- (and sometimes three-) dimensional graphs. Property maps are based on Ashby's mechanism maps, which visualize the dominant deformation mechanism active under different stresses and at different temperatures, enabling the user to identify the active creep mechanism [2]. Databases of the physical and chemical properties of materials exist; databases of nuclear properties, known as cross section library, also exist, but the different databases have not been linked together in order evaluate the physical and nuclear properties concurrently.

The U.S. materials genome project was launched in 2011 to develop databases of the properties of a large number of materials. The fundamental physical data that these databases make available, combined with current computational efforts [3–5] to predict materials' properties, represent a significant advance. This combination of information sources allows designers to use modeling to screen materials, which is accelerating materials development and expanding design options. Although Ashby's original concept of property maps focused mainly on mechanical properties, it has been expanded to include chemical properties, such as the Gibbs free energy of formation for compounds.

However, as noted above, nuclear data have not been a focus in the development of materials databases even though nuclear properties have been determined for a wide range of isotopes and are available in discrete databases known as nuclear data libraries (e.g., ENDF/B-VII [6]). Nuclear cross-sections or attenuation coefficients are examples of fundamental properties that are similar to the physical property of density or the chemical property of free energy. These nuclear properties add another dimension or degree of freedom, namely, interaction with radiation, to the characterization of a material.

In many nuclear-engineering design and application challenges, the materials-selection challenge is to find a material that has the required combination of physical, chemical, and nuclear properties. A good example is selecting and designing fusion materials. The nuclear-fusion community has made significant effort to develop and characterize reduced-activation ferritic/martensitic steels (RAFM) [7,8]. The goal is to design ferritic/martensitic materials that have specific physical properties (high-temperature strength, radiation tolerance) and exhibit as little as possible activation and transmutation of alloying elements into radioactive isotopes because these effects make maintenance of a fusion plant difficult and increase the amount of radioactive waste that must be stored. Similarly, in first-wall design, low neutron activation, high melting point, and low sputter yield are desirable properties. Fig. 1 shows examples of sputter-yield property maps for 200-electronvolt (eV) helium (He) ions impinging onto various materials, in relation to a) melting point and b) boiling point (reproduced from data in [9]). Sputter-yield equations exist but are not currently integrated with

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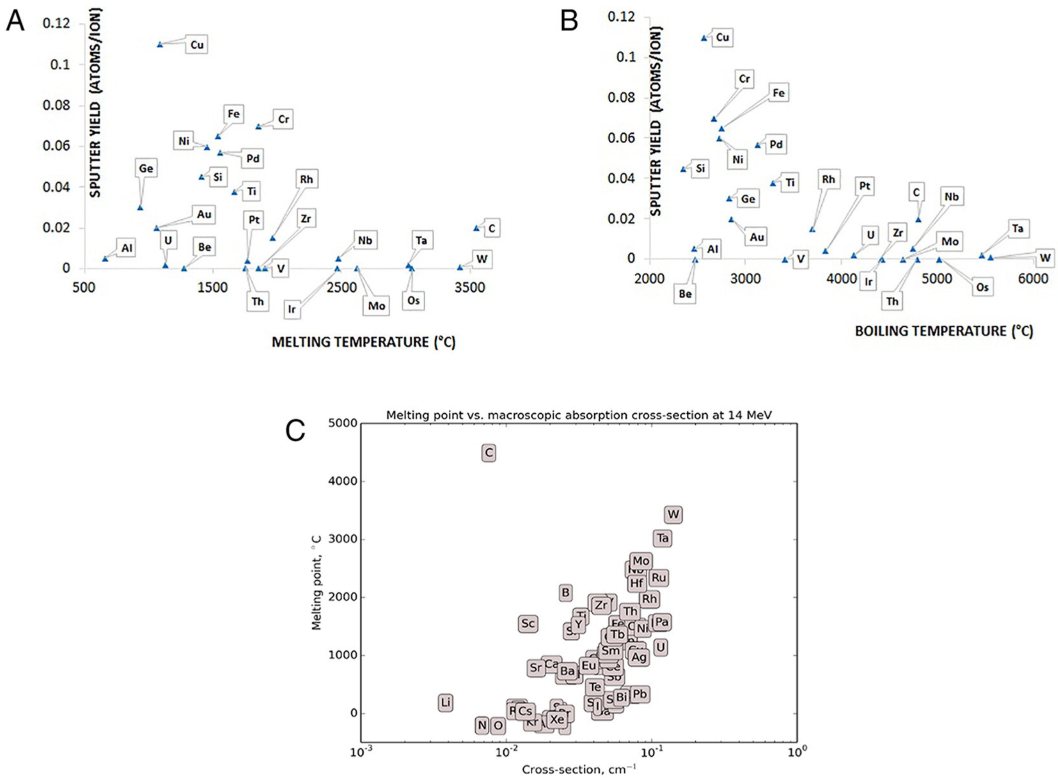

Fig. 1. Property map of selected elements: A) He sputter yield (atoms/ion) for 200 eV He ions versus melting temperature. B) He sputter yield (atoms/ion) for 200 eV He ions versus boiling temperature. C) Melting point versus macroscopic neutron absorption cross-section for 14 mega-electronvolt (MeV) neutrons.

other engineering data. Experimental data reveal the prime choices for fusion first-wall materials. For example, although carbon has a higher melting point than tungsten, its sputter yield is higher and its boiling point (at ambient pressure) is lower, making tungsten the better choice. One challenge evident in Fig. 1 is that fundamental data such as sputter yield or nuclear properties are based on pure elements, but actual engineering applications use alloys rather than elements. This reality is another reason why we need to develop nuclear materials property databases, as discussed in examples below.

In addition to understanding the nuclear properties of materials it would also be beneficial if one was able to evaluate the effect of irradiation on the material at the operating temperature of material. Irradiation damage in materials causes a wide range of changes in the material such as swelling, changes in dislocation structures, solute segregation, and radiation-enhanced and –induced precipitation. The effects of these different phenomenon in the material are embrittlement, hardening, void swelling and loss of ductility. Just as Ashby maps allow one to identify the type of creep mechanism activate at different temperatures and stress allowing to predict the possible physical/microstructural changes in the material over time in operation one could envision doing the same with irradiation damage. An example of this can be seen in Zinkle and Ghoniem [10] work on fusion materials. In this work the authors look at irradiation embrittlement and thermal creep properties and are able to define an operational temperature window for different fusion materials. Since the materials properties while in service are extremely important it would be helpful to manufacture these types' of plots for a range of materials that will allow an engineer to see quickly the materials performance and its microstructural evolution during operation or select materials that best fit the operation conditions. An initial (not complete) assessment can be the scattering cross section. If a material has high cross section one could expect that the material would experience a large amount of transmutations or displacement damage which would have an effect on the microstructure of the material and therefore the overall material proprieties. Adding temperature to this analysis would allow one to evaluate the materials ability to recover from the damage. Additionally dose rate can be an important parameter as well. Unfortunately the number of data that needs to be tracked and evaluated carefully grows rapidly making a compact database difficult.

The design of advanced fission reactors poses challenges similar to those discussed above for fusion. To select the best coolant to fulfill a reactor's physical and nuclear requirements, designers correlate properties such as density, heat capacity, boiling point, and melting point with nuclear cross-sections in the relevant spectrum. For example, fast reactor applications ideally use a coolant medium that is liquid at atmospheric pressure throughout the 150–800 °C range and has a low neutron cross-section in the fast neutron energy range. These criteria lead to the choice of liquid sodium, liquid lead, or lead bismuth. Liquid salts typically have a melting point that is higher than 150 °C, which puts them out of contention for fast-reactor applications unless the melting-point criterion is relaxed to higher temperatures [11]. However, when we consider additional parameters, such as chemical reactivity, sodium is eliminated from consideration because of its reactivity with water. This logical deduction leaves us with lead bismuth eutectic (LBE) as a theoretical better choice. However, the corrosion of steel by dissolution is a major issue in LBE systems. Thus, no coolant can intrinsically meet all of the engineering requirements listed above.

Another area where property maps would be of use is the increasing need for nuclear isotopes for cancer therapy and diagnostics. For this application, the proton-beam target windows need to withstand high-intensity radiation and high heat load and should also have low proton activation. Because of strength concerns, one of the most important parameters to consider is yield strength as a function of temperature [12], which results in the alloy HAVAR (heat-treatable Cobalt base alloy)

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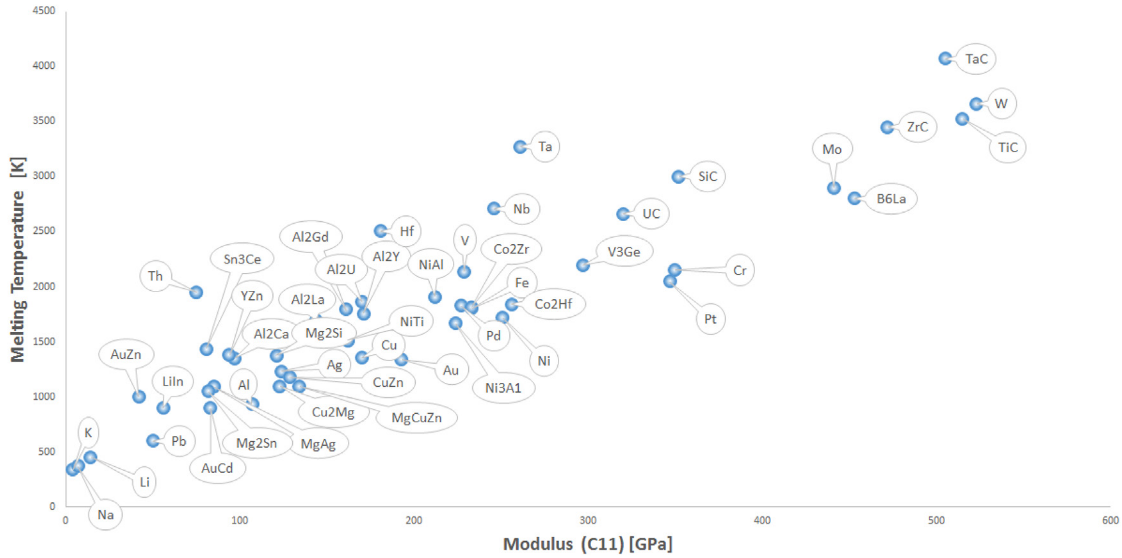

Fig. 2. Correlation between melting temperature versus elastic modulus (C11), plotted with data from [13,14].

being commonly chosen even though this material does not meet the activation criteria.

In the examples above, a comprehensive database of nuclear, physical, and chemical properties would accelerate materials design and selection. Because no database or property map currently combines classical materials properties with nuclear properties, selecting materials for applications such as the ones mentioned above requires significant, collaborative data mining by nuclear physicists, materials scientists, and mechanical engineers. Meanwhile, the need is increasing for new solutions for fusion, fission, accelerators, and medical applications. Current database-development efforts create an opportunity to introduce nuclear-materials property maps.

2. Data correlation

Many physical properties of materials show correlations. For example, melting point vs. elastic modulus or melting point vs. thermal expansion (Fig. 2) are related to the same fundamental property of inter-atomic potential and show similar trends. Correlations are also manifested in today's periodic table, where the elements are grouped in s-, p-, d- and f-blocks with similar chemical trends within each group.

Nuclear data are based on the properties of neutrons and protons rather than of electrons, so a property map including nuclear data will look fundamentally different from property maps and trends based on electron properties. The nuclear shell is a current model for understanding nuclei' nuclear properties [15]. In this model we fill the proton and neutron levels independently, so special conditions can result if neutron and proton behavior are affected separately. These cases mean that different isotopes of the same element can have vastly different neutronic properties. Such differences, say, in neutron absorption, can have significant implications for nuclear applications because they govern macroscopic phenomena, such as activation in this example.

The purpose of this commentary is to outline the need for a unifying database including neutronic properties, to facilitate the search for suitable materials or elements for nuclear applications. With maps such as the ones shown here, nuclear engineers would be able to optimize novel nuclear designs.

Next, we present case studies illustrating some nuclear engineering challenges that make clear the need for a comprehensive materials selection database and for improved data treatment. We do realize that the properties and requirements of each presented case study is larger than what is presented here. However, the issues raised do highlight

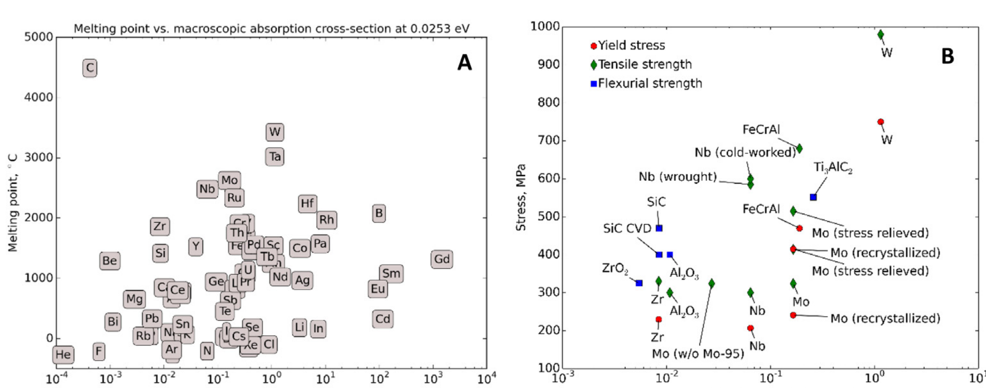

Fig. 3. A) Melting point as a function of the thermal-neutron macroscopic cross sections of the elements (natural isotopic abundance). (Right) Yield strength (for bulk) and flexural strength (for coatings) versus thermal-neutron macroscopic cross sections [26,27]. The macroscopic cross section is the microscopic cross section (cm^2) multiplied by the atomic number density (cm^{-3}), which gives it the units of cm^{-1}. In addition, 1/(macroscopic cross section) is the mean free path of the neutron.

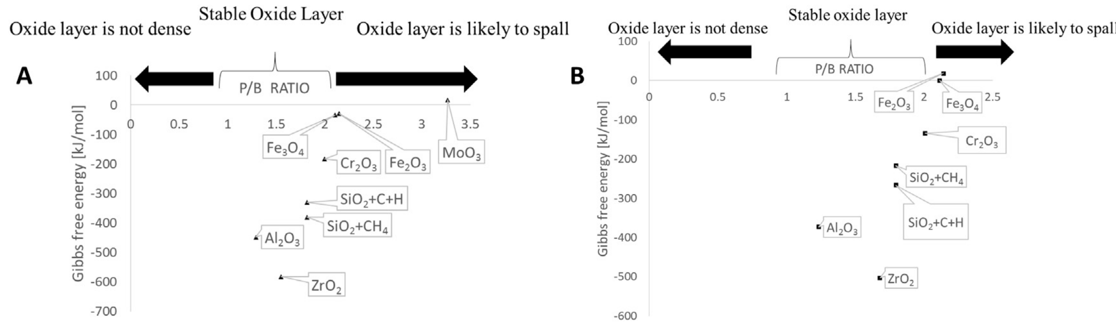

Fig. 4. A) Plot of Gibbs free energy versus P/B ratio at room temperature for selected oxides formed on metals. B) A plot of Gibbs free energy versus P/B ratio at 1000 °C [28–32].

the process of materials selection and illustrate the need for a unifying materials database including nuclear materials.

3. Case study: materials selection criteria for accident-tolerant fuel forms

The Fukushima Daiichi nuclear accident inspired research to develop accident-tolerant fuel forms to help prevent such disasters in the future. Currently, zirconium alloys are used as cladding materials in nuclear reactors. These alloys have a low Gibbs energy for oxide formation, which describes an exothermic reaction forming zirconium dioxide (ZrO_2) from the cooling water. Zirconium reduces the water in an exothermic reaction and releases hydrogen gas; therefore, plants must be designed to avoid temperatures of 1000 °C at which this reaction gets out of control. However, during a nuclear accident, lack of cooling and continuous generation of decay heat from the nuclear fuel can cause temperatures to exceed 1000 °C. Therefore, the nuclear materials community has been investigating alternative forms of fuel cladding that would avoid or at least slow down the risk of generating hydrogen and in turn an explosive hydrogen gas mixture [19]. The search for a suitable replacement material has been challenging because of the number of requirements that light-water-reactor nuclear fuel claddings must meet. Under normal power generating conditions, the material must resist corrosion by pressurized water, be inert to the uranium dioxide (UO_2) fuel and its fission products, have low fission gas permeation, have sufficient strength to hold internal pressure while providing low neutron absorption to allow an efficient chain reaction, and be cost effective and able to be manufactured. A performance evaluation light water reactor accident tolerant fuel has been discussed in [20]. Zirconium alloys meet all of these requirements during normal conditions but fail to meet them during accidents, in which the high temperature enables the runaway oxidation of the alloys by the cooling water, generating hydrogen as described above. Finding a material with all of the required properties under both normal and accident conditions has been a challenge. When no single material will fulfill all requirements, the engineer must find the best compromise. In this situation, the selection process entails weighing the importance of different material properties, such as the macroscopic absorption cross-section at operating conditions versus the hydrogen production in off-normal conditions and the relative difference between the choices.

Four systems have been studied: 1) silicon-carbide (SiC)-based composites [16–18], 2) iron-chromium-aluminum (Fe-Cr-Al) alloys [19,21–23], 3) coated zirconium [24], and 4) molybdenum (Mo) [25]. These four systems were chosen as balanced compromise among the application's requirements. However, because only mechanical and chemical property maps are available, it is difficult to assess performance based on the neutron absorption cross-section in correlation with the other properties. Considering only mechanical and chemical properties during the design process can have significant implications for the nuclear plant's maintenance and economics because higher neutron absorption in the cladding results in less energy being generated per unit mass of fuel with the same degree of enrichment. Plant redesign and operational changes may be needed to address this negative economic consequence. Figure 3 shows an example of a property map that would be useful for selecting accident-tolerant fuels; the map illustrates melting point versus thermal neutron macroscopic absorption cross-section. Zr, Si, and C are at the lower end of the cross-section spectrum but are in the middle of data points with regard to melting point and strength, which makes clear why they are the most common candidates for this application of accident-tolerant cladding. Both properties are readily available individually. Added value is generated when they are brought together in such maps.

Molybdenum has been suggested as a potential light-water reactor cladding material because of its high-temperature strength, which would prevent cladding deformation in an accident. However, examining Fig. 3, we can see that Mo has a macroscopic thermal-neutron absorption cross-section that is more than an order of magnitude higher than that of Zr. This significant increase in neutron absorption would have drastic effects on the neutronics of the core and could have major ramifications, as seen in [18]. Considering iron-based alloys, we observe that Fe and its traditional alloying elements (Cr, nickel (Ni), etc.) have greater macroscopic absorption compared to current Zr alloys but without the high melting-point advantage. This increase in cross-section of Mo- and Fe-based materials does not entirely rule out these materials for potential use as accident-tolerant cladding, but it underscores the need for thinner cladding and therefore weighs the materials selection toward yield strength. Therefore, yield strength versus macroscopic thermal neutron absorption cross-section would be another valuable property map for materials selection, as shown in Fig. 3B.

The question that we considered above for Zr (hydrogen production in water because of a runaway chemical reaction) can be addressed by considering the Gibbs free energy of oxide formation and the question of whether or not the formation of an oxide creates a passivating oxide scale on the cladding surface. Fig. 4 shows the Gibbs free energy of formation versus the Pilling Bedworth (P/B) ratio of a variety of oxides relevant to accident-tolerant cladding. The P/B ratio is one measure to assess qualitatively whether an oxide layer is under compressive stress and covers the surface. All of the listed materials fully cover the

Table 1
Comparing molybdenum isotopes [6].

Isotope	# of protons	Proton shell	# of neutrons	Neutron shell	Abundance [%]	Thermal neutron absorption σ (barns)
92-Mo	42	1g9/2	50	1g9/2	41.65	0.0153
94-Mo	42	1g9/2	52	2d5/2	9.19	0.1229
95-Mo	42	1g9/2	53	2d5/2	15.87	12.5800
96-Mo	42	1g9/2	54	2d5/2	16.67	0.6941
97-Mo	42	1g9/2	55	2d5/2	9.58	2.1341
98-Mo	42	1g9/2	56	2d5/2	24.29	0.4587
100-Mo	42	1g9/2	58	1g7/2	9.74	0.2120

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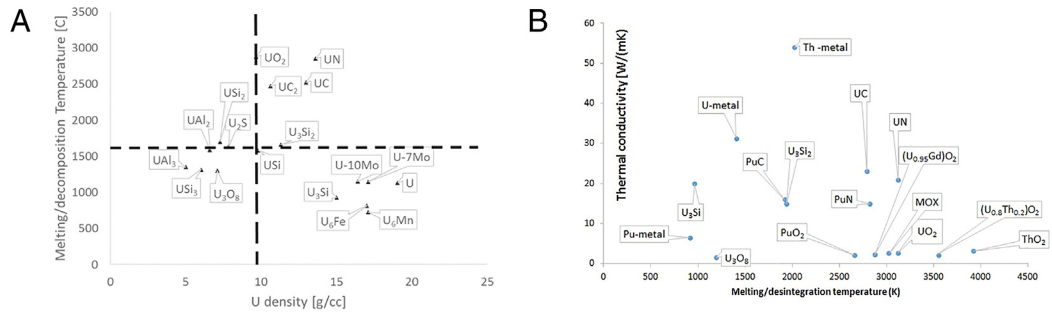

Fig. 5. A) Melting point versus uranium density for a variety of potential accident-tolerant fuel forms. In the plot, the vertical line represents maintaining at least the current density of UO_2, and the horizontal line is at 1600 °C which would be favorable because this melting point would exceed the melting points of the other core components [33–35]. B) The thermal conductivity of the fuel form versus the melting/disintegration temperature [35].

metal surface and are under compressive stress. Other parameters, such as oxidation rate and diffusion constants, must also be assessed to explain why oxides grow more rapidly on Zr alloys and Molybdenum base alloys than on steel alloys. The question of oxidation in LWR condition at elevated temperature is an important factor when performing a materials selection and effects all materials which are known to have a challenge in this area like for example Molybdenum. Extensive data mining is required for this assessment, which makes materials selection difficult. A unified database of materials properties would remedy this challenge.

Because the chemical and mechanical properties of materials depend on the electronic structure and bonding energies of the materials whereas nuclear properties are derived from the structure of the nucleus, different isotopes of the same element that have the same chemical and mechanical properties can have dramatically different nuclear properties. The nuclear properties have no effect on the chemical bonding of the material. This lack of interdependence of the properties means that we can manipulate one set of properties without interfering with the other. For example, we can revisit Mo for light-water reactor cladding and examine how one could influence the nuclear proprieties to improve the cladding's performance while maintaining its high melting point and good high-temperature strength.

Natural, elemental Mo has a valance-electron configuration of $4d^5 5s^1$. However, natural, elemental Mo is really comprised of seven Mo isotopes, which are listed in Table 1 along with their relative natural abundance. Although the chemical and mechanical properties are uniform across the different isotopes.

The rightmost column of Table 1 shows that the number of neutrons can greatly influence a material's nuclear properties: three orders of magnitude in the cross-section, in this example. We can envision that enriching one isotope over another will change the nuclear property significantly. For example, it can be seen that ^{95}Mo (15.87% natural abundance) is the largest contributor to the thermal neutron absorption cross-section. Therefore, possible enriching of the other isotopes[1] (removal of the ^{95}Mo) would result in the bulk Mo component maintaining its chemical and mechanical properties but changing its nuclear properties to a more favorable state (Fig. 3B) [18].

4. Case study: materials selection criteria for nuclear-fission fuel

UO_2 has been the main fuel form for most of the history of commercial nuclear power generation and is currently used in light-water reactors. Since the Fukushima accident, the use of UO_2 has come under increased scrutiny, and researchers are studying how to improve the accident tolerance of this fuel.

The fuel in a nuclear reactor has to survive in a harsh environment while maintaining its mechanical and chemical properties. Some fuel requirements are high melting point, low neutron cross-section of the non-fissile constituents of the uranium-based compound, chemical inertness in relation to coolant and cladding, a broad range of compositional homogeneity, stability under high temperature irradiation, high thermal conductivity, high density, and good mechanical strength. The current form of UO_2 fuel excels in a number of these areas; however, UO_2 has low thermal conductivity and poor mechanical strength. A variety of fuel forms have been suggested as possible replacements for UO_2, including uranium silicide (U_3Si_2), uranium nitride (UN), and uranium carbide (UC).

The melting point of the fuel is an important parameter because it is desirable for the fuel to remain solid at high temperatures and have the highest melting point of the materials in the core in the event of an accident. In addition, the heavy-metal density of the fuel is an important parameter; a new fuel would need at least to match the density of the current UO_2 fuel in order to avoid large changes in economics, fuel reactivity, or cycle length of the nuclear plant. Fig. 5A plots a variety of different fuels with their melting points versus their uranium density. The plotting of these properties allows one to eliminate quickly the elements that do not meet the criteria described above. Another essential relationship to plot is thermal conductivity versus melting point (Fig. 5B). Not surprisingly, the purely metal compounds exhibit the highest thermal conductivity, the oxides exhibit the lowest, and the nitrides and silicides are in between. Increasing the thermal conductivity of the fuel allows the fuel to have lower center-line temperatures, which means less heat stored in the fuel, the ability to transport heat to the coolant rapidly, lower fission gas release and fuel swelling; all these features must be considered in analyzing accident scenarios.

5. Case study: materials selection for fusion blanket coolants

Much research is focused on making commercial fusion energy a reality. Two key designs for fusion-based energy are magnetic and inertial confinement. A number of materials challenges need to be addressed [36–38]. We have discussed some aspects of the first wall; here, we discuss selecting materials for liquid-fusion blankets.

Viable coolant candidates for deuterium-tritium (D-T) fusion reactors must fulfill multiple requirements. In addition to suitable thermophysical properties, the blanket coolant is expected to collect the fusion energy from neutrons and breed enough tritium to make the system tritium self-sufficient. Two main figures of merit are typically used when evaluating the effectiveness of a fusion blanket: (1) tritium breeding ratio (TBR), defined as the number of tritium atoms generated per atom consumed at any given moment; and (2) energy multiplication (EM), defined as the ratio of the total amount of energy deposited in the blanket to the energy generated by the fusion source. Because the

[1] Isotopic enrichment has been often considered for various nuclear applications; for a complete assessment, other factors should be considered and in particular its cost.

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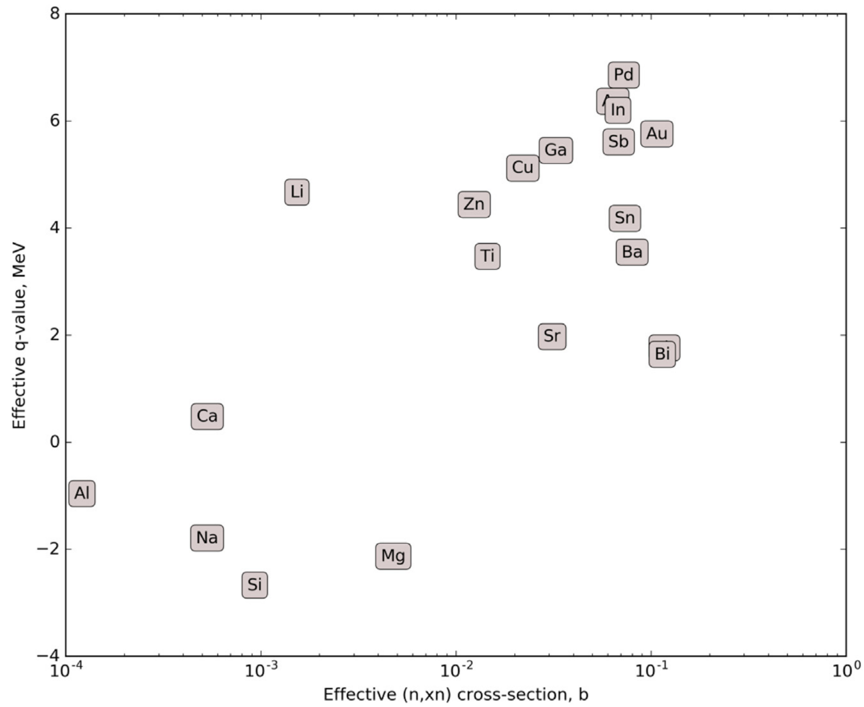

Fig. 6. Effective q-value versus effective (n,xn) cross section as calculated using a representative inertial confinement fusion blanket neutron spectrum [38] for candidate alloy components of blanket coolants.

coolant is one of the main components of the blanket, it is important to establish the coolant's contribution to these two parameters. Liquid lithium (Li) is a preferred choice for the blanket coolant because tritium is easily obtained by means of (n,α) reactions on Li-6. However, Li is chemically reactive. Alloys of Li that are less reactive would be preferable if they can provide acceptable tritium breeding and energy multiplication. Fig. 6 gives basic data that could guide the choice of an alloy alternative to pure Li, allowing the designer to infer the impact of each element on TBR and EM. The (n,xn) cross-section represents the capability of an element to multiply neutrons; the larger the cross-section, the larger the number of neutrons available for tritium breeding. The q-value is the average energy generated by all possible nuclear reactions for an element, weighted by the respective probability of the reaction. Because (n,xn) reactions are endothermic, and the energy is transferred to the produced neutrons, the energy generated (or lost) by these daughter neutrons is also considered. For simplicity, we assume that each neutron from (n,xn) reactions is eventually absorbed in Li. Both quantities, (n,xn) cross-sections and q-value, were determined using a representative fusion blanket spectrum; therefore, they are said to be the "effective" values. Detailed studies as performed by Jolodosky et al. [39] confirm that although some elements would be rejected based on other considerations, the elements in the top-right part of Fig. 6 are preferable as Li alloy components for fusion blankets.

6. Summary

The preceding case studies illustrate nuclear engineers' need to link together comprehensive databases of a large number of different materials properties, especially the heretofore-separated databases of nuclear properties and physical properties, to accelerate and enhance development in nuclear materials science and technology. The inclusion of nuclear properties in the combined database will effectively act like another parameter, or degree of freedom, in the design process, as compared with traditional, non-nuclear materials selection. Increasing capability in databases and computing power could support the creation of unified materials properties databases that would accelerate materials selection and engineering design for nuclear applications; non-nuclear applications will also benefit from this effort.

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