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Review

Evaluating Nuclear Forensic Signatures for Advanced Reactor Deployment: A Research Priority Assessment

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Abstract: The development and deployment of a new generation of nuclear reactors necessitates a thorough evaluation of techniques used to characterize nuclear materials for nuclear forensic applications. Advanced fuels proposed for use in these reactors present both challenges and opportunities for the nuclear forensic field. Many efforts in pre-detonation nuclear forensics are currently focused on the analysis of uranium oxides, uranium ore concentrates, and fuel pellets since these materials have historically been found outside of regulatory control. The increasing use of TRISO particles, metal fuels, molten fuel salts, and novel ceramic fuels will require an expansion of the current nuclear forensic suite of signatures to accommodate the different physical dimensions, chemical compositions, and material properties of these advanced fuel forms. In this work, a semi-quantitative priority scoring system is introduced to identify the order in which the nuclear forensics community should pursue research and development on material signatures for advanced reactor designs. This scoring system was applied to propose the following priority ranking of six major advanced reactor categories: (1) molten salt reactor (MSR), (2) liquid metal-cooled reactor (LMR), (3) very-high-temperature reactor (VHTR), (4) fluoride-salt-cooled high-temperature reactor (FHR), (5) gas-cooled fast reactor (GFR), and (6) supercritical water-cooled reactor (SWCR).

Keywords: nuclear forensics; advanced reactors; fuel cycle; forensic signatures; priority score

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1. Introduction

Nuclear energy and nuclear security have been intertwined since the Manhattan Project, during which the first nuclear reactor, Chicago Pile 1, was constructed, thus enabling the technological and scientific advances that led to the use of atomic bombs in Hiroshima and Nagasaki [1]. These events spurred the need for international security and safeguards on special nuclear materials (SNMs) that could end up out of regulatory control by diversion from weapons programs. Once nuclear energy was recognized for its low carbon footprint and became an attractive solution to increasing demand in civilian energy [2], nuclear reactors became another potential source of SNM diversion.

According to the International Atomic Energy Agency (IAEA) [3], nuclear forensics is "the examination of nuclear or other radioactive material or evidence that is contaminated with radionuclides, in the context of legal proceedings under international or national law related to national security. The analysis of nuclear or other radioactive material seeks to identify what the materials are, how, when, or where the materials were made, and what their intended uses were". The birth of nuclear forensics coincides with the fall of the Soviet Union in the early 1990s, after which a slew of nuclear and radioactive materials were misplaced or stolen as a result of political turmoil [4]. Nuclear forensics is a key element of national security and traditionally has leveraged known relationships between material characteristics and process history to establish material provenance.

Throughout recent years, nuclear forensics research and development has grown quickly, as evidenced by several reviews, chapters, and books written on the subject since the turn of the century [4–12]. Recent advancements in the field include measurements of material properties such as morphology [13–17] and composition [18,19], as well as improvements in instrumental analysis techniques, including spatially resolved and high-precision measurements [20,21]. These developments are pursued with the goal of linking samples of nuclear forensic interest to people, places, and processes with unique, measurable characteristics known as *signatures*. Nuclear forensic signatures are therefore defined as characteristics, or combinations of characteristics, that help to address investigative questions.

To date, most nuclear forensics research and development has focused on signatures pertaining to material from the nuclear fuel cycle (e.g., ceramic UO_2 fuel pellets and uranium ore concentrate (UOC)) [22,23], as these are attractive materials for diversion [24]. However, around the turn of the 21st century, designs for advanced reactors began to rise in popularity within the nuclear community for their improved heat transfer, material properties, efficiency, and novel passive safety features [2]. The past two decades have seen advanced reactors mature to design, construction, and even operational phases around the world [25,26]. Each of these designs has updated the traditional light water reactor (LWR) in any number of ways, from increased core temperature to changes in fuel, coolant, neutron spectrum, or core design. With the rise of advanced reactors in the clean energy sector, so too comes a need for new safeguards and nuclear forensic signatures. With the fabrication of new SNM-bearing reactor fuels, entirely new ways to divert nuclear and radioactive materials are introduced and must be addressed before implementation [25,27–31]. Signatures that are currently used to identify the origin of nuclear materials out of regulatory control are not sufficient for analysis of advanced fuels that may take on entirely new compositions (e.g., Th-U vs. U-Pu) and forms (e.g., molten salts, tri-isostructural (TRISO), metal).

In order to assess the state of understanding of nuclear forensic characteristics and signatures of advanced reactor fuels, we have completed an evaluation of advanced reactor designs and the potential signatures of their proposed fuels. We provide a semi-quantitative assessment of six advanced reactor designs based on technical maturity, material attractiveness, and the degree of difficulty, or strain, involved in researching signatures for relevant advanced fuels. Each reactor design has been assigned a *priority number*, which suggests the order in which the nuclear forensics community should prioritize research and development of material signatures for the related fuel cycle.

2. Defining Priority for Advanced Reactors Signature Research

Nuclear forensics has long focused on developing the signatures of uranium and plutonium-based materials diverted from both commercial reactors and their respective fuel cycle facilities. Today, a majority of commercial reactors are LWRs, but there is growing interest in advanced reactors, evidenced by advanced reactor programs in the US [32] and internationally [26]. Just as with any other nuclear facility, advanced reactors are attractive targets for the diversion of nuclear materials [27–31,33]. To prepare for this vulnerability, the nuclear forensics community should pursue research on nuclear forensic signatures of materials that may be diverted from them.

This initiative first requires a compilation of knowledge on promising advanced reactor designs, including their deployment timelines, technical maturity, and the composition and form of their fuel. This evaluation is intended to inform the priority in which the nuclear forensics community could undertake research and development toward signatures associated with each advanced reactor design.

The Gen IV International Forum (GIF) identified six promising advanced reactor designs in its original (2002) Technology Roadmap [2], which was updated in 2014 to reflect the advancements of those systems during the previous decade [25]. The systems identified by GIF include the gas-cooled fast reactor (GFR), very-high-temperature reactor (VHTR), supercritical water-cooled reactor (SCWR), lead-cooled fast reactor (LFR), sodium-cooled fast reactor (SFR), and molten salt reactor (MSR). Although this is not an exhaustive list of advanced reactor designs, these six represent some of the more technologically mature designs and are a logical option to focus future safeguards and forensics research.

To assess advanced reactors in a nuclear forensic context, this work considers six slightly adjusted categories of advanced reactors. Figure 1 lays out the types and subtypes of present-day reactors and advanced reactors discussed in this review. While much of the categorization used here is based on the GIF Technology Roadmap update [25], input from the IAEA's status update on molten salt reactors [34] and the advanced reactor fact sheet from the US Department of Energy (DOE) [35] are considered as well. Due to similarities in their fuel cycles, SFRs and LFRs will be treated jointly as liquid-metal-cooled reactors (LMRs) Similarly, gas-cooled reactors (GCRs), high-temperature gas-cooled reactors (HTGRs), and VHTRs, will be treated jointly under the category of GCRs to reflect their similar fuel cycles. This adjustment is intended to simplify the complex field of advanced reactors into groups that can be more easily evaluated through a nuclear forensic lens. GFRs will be treated independently from GCRs since they are expected to use a different fuel type and neutron spectrum. Finally, fluoride-cooled high-temperature reactors (FHRs) and liquid-fueled MSRs will be analyzed independently to account for the major differences in fuel form (i.e., solid versus liquid fuel), which makes a large impact on nuclear forensic analysis.

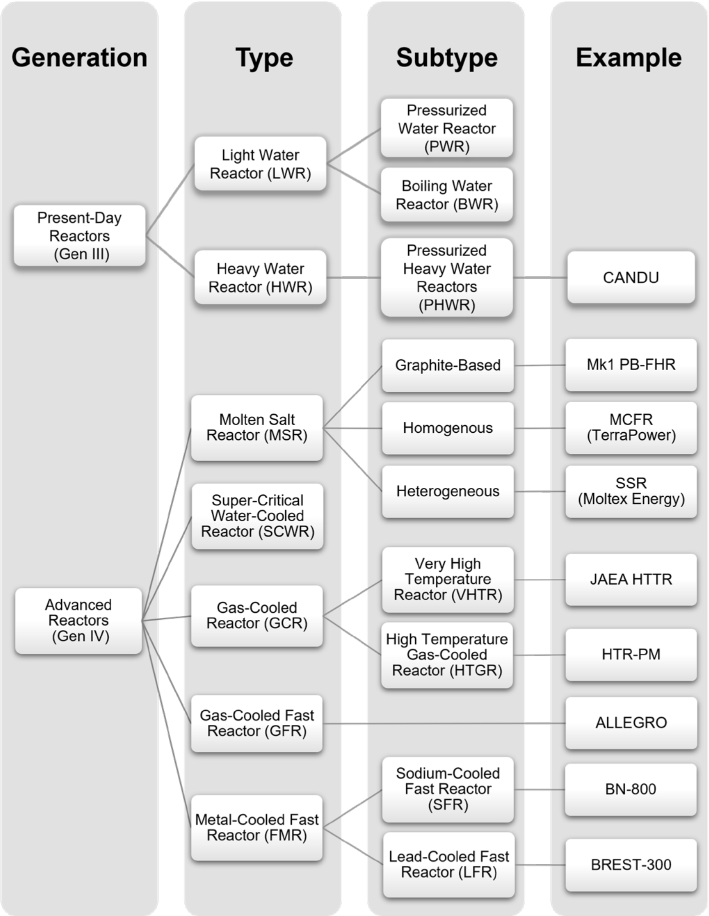

Figure 1. Reactor types and their hierarchical organization. Examples of operational or planned reactors are included to exemplify less common reactor subtypes.

2.1. Assessing Priority

The priority of research and development of material signatures for each advanced reactor design was evaluated using a novel semi-quantitative scoring system. A priority, 1 (highest priority) through 6 (lowest priority), was calculated for each reactor design based on the sum of three sub-scores: (1) technical maturity (TM), (2) material attractiveness (MA), and (3) signature research and development strain (RDS). The reactors were awarded a 0, 1, or 2 in each sub-score, with increased maturity or attractiveness yielding a higher sub-score. The sum of these three sub-scores was then sorted to reveal a priority order for further signatures research. For example, a reactor design that receives three sub-scores of the highest possibilities, 2, will have a total sum score of 2 + 2 + 2 = 6. A high sum score such as this would imply a high priority for the given design.

The sub-scores are intended to discuss the accessibility of attractive nuclear materials from the reactor in question, the level of threat posed by any amount of diverted fuel material, and the ease with which the source of diverted reactor materials could be attributed—given the current state of nuclear forensic signature knowledge. They are each evaluated on the same finite scale (0–2) to achieve equal weighting between these three factors. Further explanation of each sub-score and their assignment criteria are provided in the following sub-sections.

2.1.1. Technical Maturity (TM) Sub-Score

The technical maturity (TM) sub-score describes the accessibility of reactor materials and related fuel cycle materials by examining the total number of reactors on the market or in design stages. Table 1 provides a general description of assignment criteria for a TM sub-score of 0, 1, and 2. A higher sub-score corresponds to a more mature reactor technology. The TM of a reactor design was decided based on two criteria: (1) the estimated time until deployment of the first experimental or demonstration reactor, and (2) the number of reactors in design, construction, and operational states internationally. In general, less time until deployment and more planned reactors result in a higher TM sub-score.

Table 1. Technical maturity sub-score assignment criteria for 0 (least mature), 1 (average maturity), and 2 (most mature).

Technical Maturity Sub-Score	**0**	**1**	**2**
General description	**Least Mature** The most time until a large quantity of this reactor (and fuel) is on the market	**Average Maturity** An intermediate amount of time until this reactor (and fuel) is on the market	**Most Mature** A large quantity of the material used to fuel this reactor is, or will be, in commercial use very soon
Time to planned reactor	>10 years	5–10 years	0–5 years
Comparative number of planned reactors (weighted rank) [1]	1, 2	3, 4	5, 6

[1] See Table 2.

To factor in the number of reactors in various construction/operational phases, the reactor designs were assigned a rank, as shown in Table 2. The rank was determined primarily through data from the IAEA Advanced Reactor Information System (ARIS) database [26] and supplemented with other sources when indicated in the respective reactor design sections. The unweighted rank of each reactor design is based only on the total number of reactor projects in progress, i.e., the highest unweighted rank was awarded to the reactor design with the greatest total number of reactors. In the event of a tie in unweighted ranking, a weighted rank was determined by applying greater weight to commercial reactors, as opposed to demonstration or experimental reactors. Since commercial reactors require more fuel to operate, this weighted ranking system places the priority on reactor designs that may introduce a higher quantity of fuel into the market at a faster rate.

Table 2. Technical maturity ranking and sub-score based on the number of commercial, demonstration, and experimental reactors. Data retrieved from the IAEA ARIS database [26] and supplemental sources (see Section 2.2 text for details). Rank is assigned from 1 through 6 in order of increasing maturity. Color coding of the table highlights low TM (green), moderate TM (yellow), and high TM (red).

Reactor	Commercial	Demonstration	Experimental	Total Reactors	Rank (Unweighted)	Rank (Weighted)	TM Sub-Score
Gas-cooled fast reactor (GFR)	1	2	0	3	1	2	0
Liquid-metal-cooled reactors (LMR)	10	8	4	22	6	6	2
Molten salt reactor (MSR)	4	1	2	7	5	5	2
Supercritical water-cooled reactor (SCWR)	0	3	0	3	1	1	0
Gas-cooled reactor (GCR)	3	2	0	5	3	4	1
Fluoride-cooled High-temperature reactor (FHR)	2	2	1	5	3	3	1

In calculating the TM sub-scores in this work, it was found that the estimated time until a planned reactor was in operation for all advanced reactor designs was within 5 years [25,26,36,37]. Thus, they were indistinguishable by that criterion. As a result, the number of planned reactors was the main contributing criterion for determining the TM sub-scores. The reactors with the two highest ranks (5 and 6) were given the highest sub-score (TM = 2), those ranked 3 and 4 were given sub-scores of 1, and, finally, the reactors that were ranked lowest (1 and 2) were given sub-scores of 0. In this work, the sub-score assignments were not affected by using the weighted rank, as opposed to unweighted. However, any future use of this scoring system may reveal differences in scoring while using weighted ranking. At that time, a re-evaluation of the effect of the criterion “time to reactor operation” could be considered to strengthen the sub-score assignments.

2.1.2. Material Attractiveness (MA) Sub-Score

The material attractiveness (MA) sub-score describes the likelihood of diversion of nuclear material from a specific design. In general, an attractive material is one that has a low critical mass, does not require extensive reprocessing steps, and has a low dose rate. This sub-score is based on work by Bathke et al. (2012 and 2014) [24,38], which ranks several fuel cycle materials in terms of MA using a quantitative figure of merit. The figure of merit system provides a low, medium, or high MA result, which corresponds to a 0, 1, or 2 MA sub-score in this work, respectively. Some novel fuels may not fit perfectly into one of these levels of attractiveness. Mixed oxide fuels (MOX) are an example of one of these fuels that is not on Table 3 because there is not yet a standard composition of MOX. As a result, it is difficult to place it at any one level of attractiveness. Instead, the MA of MOX fuel will be assessed according to the isotopic composition expected for a specific reactor. Table 3 summarizes the criteria that are used to assign MA in this work.

Table 3. Material Attractiveness (MA) sub-score criteria. Many of these criteria are based on work by Bathke et al. (2012) [24]. All percentages are atomic percentages.

Material Attractiveness Sub-Score	**0**	**1**	**2**
General description	**Least Attractive** High critical mass Extensive reprocessing required High dose rate	**Average Attractiveness** Some combination of high/low critical mass and reprocessing needs	**Most Attractive** Low critical mass No or little reprocessing required Low dose rate
Typical uranium-based material	LEU (^{235}U < 5%)	HALEU (5–20% ^{235}U)	HEU (^{235}U > 20%)
Plutonium-based materials	Pu with > 18% ^{240}Pu * ^{238}Pu/^{239}Pu (>80%/<20%)	Pu with 7–18% ^{240}Pu 20–80% ^{238}Pu	Pu with 3–7% ^{240}Pu † <20% ^{238}Pu
Thorium breeder materials	Pre-irradiated thorium	^{233}U within used fuel	Separated ^{233}U (<10 ppm ^{232}U)
Other	Non-U-bearing Am and Cm	Transuraniucs (TRU)	

* Typically, high burn-up plutonium. † Also known as weapons-grade plutonium.

Additionally, this sub-score does not directly account for changes in MA that may occur throughout a fuel cycle. The attractiveness of material throughout the uranium fuel cycle, for example, is well understood and can be accounted for by noting changes in (1) enrichment in ^{235}U and (2) contamination with fission products. Generally, material within the uranium fuel cycle is most attractive before entering the reactor. In contrast, the MA of materials within the thorium fuel cycle is more difficult to understand and is complicated by the use of breeder reactors. There, attractiveness increases with the production of ^{233}U in the reactor and decreases with ^{232}U buildup. Overall, material within the thorium breeder fuel cycle is most attractive after irradiation in the reactor and separation of fertile ^{233}U in the decay tank. Thus, depending on the fuel cycle, material may be more attractive at different stages.

In an attempt to maintain a simple but accurate scoring system, the MA sub-score for each reactor design was determined based on the most attractive material found within the most commonly used fuel cycle for that design. A brief analysis of known reactors from ARIS and relevant company publications was used to determine the applicable fuel cycle and material for the MA sub-score assignment. Representative examples are shown in Table S1 in the supplementary information. Future iterations of this work could employ additional analyses of all possible reactor designs and fuel cycles to capture the changes in MA throughout fuel cycles.

2.1.3. Signature Research and Development Strain (RDS) Sub-Score

The final sub-score, signature research and development strain (RDS) describes the relative maturity of the research and development of signatures and related nuclear forensics analysis techniques for material related to each advanced reactor concept. In other words, a highly mature field of research would lead to low strain in the research of new or improved signatures. A well-developed signature is characterized by several well-established and peer-reviewed studies linking a particular material property (e.g., particle size of UOC grains) to relevant people, places, or processes of that material (e.g., a specific grinding process used only in the United States). In contrast, a less developed signature would be characterized by a lack of qualitative data linking the material property in question to any people, places, or processes related to the history of the material. This signature would not be acceptable for presentation in a legal context and thus not applicable for official nuclear forensic analysis of material out of regulatory control.

The basis of this sub-score is described in Table 4. Since the majority of research on signatures of reactor-related materials has been focused on UOCs and uranium fuel pellets for LWRs, an RDS sub-score of 0 was given to any reactor design that intends to use mainly ceramic UO_2 fuel pellets. On the other hand, a higher RDS sub-score was awarded to designs that intend to use fuel with less developed signatures. The difference between a sub-score of 1 and 2 is the ability to use a combination of physical measurements and databases to yield information about manufacturing and intended use. For example, liquid fuels can cool into any shape regardless of manufacturing and use; thus, analysis of those materials must focus on compositional signatures. Typically, signatures related to the isotopic, elemental, and chemical composition of the material are less developed, especially for novel fuels, so reactor designs intending to use liquid fuels are assigned an RDS sub-score of 2.

Table 4. Criteria for the signature research and development strain (RDS) sub-score.

Signature RDS Sub-Score	**0**	**1**	**2**
General description	Signatures = well characterized. Physical dimensions **can be** used to track the manufacturing process. Databases or standards exist for this type of fuel.	Signatures = not well characterized. Physical dimensions of the fuel can **in theory** be used to track the manufacturing process. Sufficient databases or standards do not yet exist.	Signatures = not well characterized. Physical dimensions **cannot be** used to determine the manufacturing history of this fuel. Sufficient databases or standards do not yet exist.
Fuel type examples	Ceramic UO_2 fuel pellets.	Any novel solid fuel (e.g., TRISO).	Liquid fuel (e.g., molten salt).

Admittedly, the construction of this final sub-score pointedly and qualitatively attributes higher priority to MSRs based solely on their liquid fuel form. Future work on this scoring system should develop a method of quantifying this sub-score. For this work, a qualitative sub-score for this category will suffice to indicate the challenge that is posed by the source attribution of liquid fuel as opposed to solid fuels.

3. Evaluation of Advanced Reactor Designs

In this section, each of the advanced reactor concepts is evaluated according to the priority scoring system. Due to the sheer variety of designs for each, reactors are assessed based on a few select designs for which sufficient information is publicly available to perform the evaluation. Further iterations of this work should strive to update this evaluation with additional reactor designs, if applicable.

3.1. Gas-Cooled Fast Reactors (GFRs)

Gas-cooled fast reactors (GFRs) are high-temperature helium-cooled reactors that use a fast neutron spectrum and typically operate with a closed fuel cycle [25,29]. Some advantages of GFRs include long-term sustainability, waste minimization via reprocessing, high thermal cycle efficiency, and a chemically inert, single-phase coolant. High-temperature transients require GFRs to use a dense fuel element, like cladded metallic or ceramic pellet fuels. The disadvantages of GFRs include rapid core heat up with the loss of forced cooling due to the low thermal inertia of the coolant and a coolant density that is too low for natural convective cooling [25].

The ALLEGRO experimental reactor is an example of a GFR demonstration reactor that is currently under design in the European Union (EU) [39]. The fuel for ALLEGRO will start as 35% mixed oxide (MOX) containing around 4.5% ^{235}U and 5% ^{239}Pu [39]. This starter fuel will slowly be replaced by experimental UPuC fuel with 29–35% plutonium content [39]. The EM2 reactor, a commercial reactor being developed by General Atomics in the United States, has been designed to use uranium carbide (UC) fuel with an enrichment of 7.7% ^{235}U [26]. Though the HALEU fuel planned for use by these two representative GFRs is only moderately attractive, the eventual plutonium-based fuel has the possibility of higher attractiveness to diversion.

At this point, the GFR will be assigned an MA sub-score of 2 to account for the uncertainty around plutonium content in these planned designs (see Table 3). UC and MOX/UO_2 fuels are novel solid fuel forms, which give GFRs a signature RDS sub-score of 1 (see Table 4). According to the IAEA ARIS database [26], three GFRs are in progress internationally: (1) ALLEGRO, (2) KAMADO FBR (in Japan), both demonstration reactors that are still under design, and (3) EM2, a commercial reactor. As compared to the others, there are relatively few planned reactors of this type, leading to a weighted technical maturity rank of 2 (see Table 2). This results in a TM sub-score of 0, the lowest. Thus, the sum score for the GFR concept is 3, which is average as compared to the other reactor concepts.

3.2. Gas-Cooled Reactors (GCRs), High-Temperature Gas-Cooled Reactors (HTGRs), and Very-High-Temperature Reactors (VHTRs)

Very-high-temperature reactors (VHTRs) are a subset of GCRs. They have a thermal neutron spectrum, are moderated by graphite, and use helium as a coolant. A VHTR is differentiated from a GCR and an HTGR based on the core temperature of the reactor. A VHTR aims for the highest core outlet temperature of the three concepts, 1000 °C, though recent designs achieve closer to 700–950 °C [26]. Such high temperatures allow for highly efficient electricity generation at 47–50% of thermal output [25]. Other advantages include the ability to produce hydrogen, a strong negative temperature coefficient of reactivity, and the high heat capacity of the graphite core [25]. The expected fuel for VHTRs is tri-structural isotropic (TRISO) particle fuel. Presently, more research and development on the fuel is required to ensure temperature stability at the extreme heat associated with VHTRs [25].

Since TRISO is a novel solid fuel, this reactor concept is assigned a signature RDS sub-score of 1. Due to the similarities between VHTR, GCR, and HTGR, the technical maturity score considers reactor designs from all three. As shown in Table 2, there are three commercial reactors: PBMR (South Africa), Prismatic HTR (GA, USA), and SC-HTGR (Framatome, USA), and two demonstration reactors, GTHTR300C (Japan) and HTR-PM (China) [26]. Compared to other advanced reactors, five total reactors that are in at least design stages give GCR/HTGR/VHTR a weighted technical maturity rank of 4 and a TM sub-score of 1. The range of enrichments planned for use in VHTR TRISO fuel is around 8–16% ^{235}U—the HTR-PM uses 8.5% ^{235}U, and the SC-HTGR uses 15.5% ^{235}U [26]. Thus, the MA score for VHTR is 1. The sum of all the sub-scores for GCR/HTGR/VHTR reactor concepts is 3, tied with GFRs.

3.3. Supercritical-Water-Cooled Reactor (SCWR)

Supercritical water-cooled reactors (SCWRs) are LWRs that operate under high temperatures and pressures above the critical point of water [25,39]. These reactors can operate with thermal or fast spectrum neutrons. Some of the advantages of SCWRs include increased thermal efficiency, elimination of coolant pumps, implementation of steam separators or steam generators, and a higher steam enthalpy, all of which contribute to lower operational costs [25]. However, the development of this advanced reactor still requires validation of transient heat transfer models, qualification of materials for cladding, and demonstration of passive safety systems [25].

According to the ARIS database, there are three demonstration SCWR reactors in the design stages: the CSR1000 in China, the HP-LWR in the US, and the JSCWR in Japan [26]. Since this is a low number of planned reactors as compared to the others, a weighted technical maturity rank of 1 is awarded, which corresponds to a low TM sub-score of 0.

SCWRs are designed to use the same ceramic UO_2 fuel pellets that are currently used in commercial LWRs but with higher enrichment—around 6.3% ^{235}U [26]. Thus, SCWRs receive an MA sub-score of 1 (for their HALEU level enrichment) and a signature RDS of 0 (for their well-studied fuel form). The sum score of the SCWR concept is 1, the lowest of the advanced reactor concepts.

3.4. Liquid-Metal-Cooled Reactor (LMR)

Liquid-metal-cooled fast reactors (LMRs) are fast spectrum reactors that utilize molten lead or molten sodium as a coolant [25,39]. The primary system can be operated at atmospheric pressure and high temperature, which can yield high power conversion efficiency [25]. Liquid metal coolant is attractive due to its high density, boiling point, heat of vaporization, and thermal capacity, which leads to less core voiding, more fuel dispersion, and better thermal inertia in case of loss of heat sink [25]. However, there are some corrosion effects with lead coolant at high temperatures. The weight of lead can be a problem for engineering seismically safe buildings, and the opacity can be an issue for inspection, monitoring, and handling. Alternatively, sodium coolant is quite reactive with air and water, which requires a sealed coolant system and complicates the safe handling of the material.

Both systems are relatively mature, and in total, there are four experimental, eight demonstration, and ten commercial reactors either operating or in design processes [26]. Thus, the LMR receives a weighted technical maturity rank of 6 and a TM sub-score of 2, the highest possible sub-score.

There are many proposed fuels for LMRs, though most designs use a solid MOX or nitride fuel [25,26]. Thus, the signature RDS sub-score is assigned a 1 (see Table 4). Two examples of LMR include the BREST-300, a lead-cooled reactor by Rosatom in Russia, and the Power Reactor for Innovative Small Molecule (PRISM), a sodium-cooled GE-Hitachi reactor design that is being used for the Natrium reactor and ARC-100 reactor [40]. These reactors use U-Pu nitride fuel with about 13.5% enrichment and a solid metallic alloy fuel of Zr, U, and 26% Pu, respectively [26]. The fuel enrichments assign LMR an MA sub-score of 1. Thus, the sum score of the LMR concept is 4, higher than GFR, GCR, and SCWR.

3.5. Molten Salt Reactor (MSR)

Molten salt reactors (MSRs) are defined by the use of liquid salts as a coolant, fuel, or both [25]. The IAEA [34] breaks MSRs into four classes: graphite-based, homogeneous (coolant and fuel are mixed in the core), heterogeneous (coolant and fuel are separate in the core), and other—as shown in Figure S1. Classes are then divided into families and further into types. Also included for each reactor type in Figure S1 is an example reactor that operated in the past, is currently operating, or is in the design stage. Note that the family "fluoride-salt-cooled high-temperature reactors (FHRs)" are distinguished by their use of solid fuel as opposed to molten salt fuel. Since the goal of this work is to identify the maturity of reactor designs based on fuel cycle materials, FHRs will be evaluated in the next section, separate from the rest of MSRs, due to this difference in fuel form. Considering only liquid-fueled MSRs, a signature RDS score of 2 is assigned to account for the less-developed research on halide salts in a nuclear forensic context (see Table 4).

In a liquid-fueled MSR, the fissile fuel is dissolved in molten halide salt (typically fluoride or chloride) [25,34]. MSRs can use either fast or thermal spectrum neutrons, though the use of fast spectrum allows for breeding fissile materials, resulting in more efficient resource utilization and waste minimization [25]. Other advantages of MSRs include low pressure, high boiling point, optical transparency, large negative temperature and void reactivity coefficients of molten salts, the possibility of using a closed thorium fuel cycle, and large-scale power generation with passive safety characteristics [25]. Research and development efforts on tools to limit corrosion on structural materials and further safety analysis are required before proceeding with construction on most designs.

According to ARIS, there are four commercial liquid-fueled MSR designs in progress: IMSR-400 (Canada), LFTR (Terrestrial Energy, Charlotte, NC, USA), MSR-FUJI (Japan), MSTW (Copenhagen, Denmark), and ThorCon (ThorCon, USA), as well as, one demonstration reactor: MSFR (France) [26]. The US Nuclear Regulatory Commission (NRC) also recognizes the TerraPower Molten Chloride Fast Reactor (MCFR), which is an experimental MSR being designed in the USA [41,42]. With six total reactors, many of which are slotted for commercial use, the technical maturity of MSRs is ranked a 5 and given a high TM sub-score of 2.

The fuel used in MSRs can vary widely by reactor, so to evaluate material attractiveness, consider the MCFR as a representative reactor. The fuel for MCFR is intended to be U-Pu-Th-LiF with an initial enrichment of 12% ^{235}U [43]. This value gives MSRs an MA score of 1, similar to most of the other advanced reactor concepts. Note, however, that fuel enrichment percentages are difficult to find for most liquid-fueled MSRs. This most likely stems from the complexity of closed and modified open fuel cycles, which flow liquid fuel and coolant throughout the reactor, as well as from the experimental and proprietary nature of MSR research today. Therefore, the MA sub-score of MSRs is likely to change from reactor to reactor and possibly over time. With this evaluation, the sum score for the MSR concept is 5, the highest of the advanced reactor systems.

3.6. Fluoride-Cooled High-Temperature Reactor (FHR)

The FHR is a family of MSRs that use solid fuel and a molten salt coolant [34]. Typically, the fuel used is TRISO [26], which gives FHR a signature RDS score of 1 (see Table 4). There are two FHRs cited in ARIS: the SmAHTR at Oak Ridge National Laboratory (ORNL) and the Mk1 FHR at UC Berkeley [26]. According to the World Nuclear Association, three additional FHRs are in progress but not listed in ARIS. These include (1) TMSR-SF1, the sister reactor to TMSR-LF (China), (2) KP-FHR (Kairos Power, Alameda, CA, USA), and (3) the test rector Herme (Kairos Power) [37]. A total of five reactors gives the FHR a weighted technical maturity rank of 3 and a respectable TM sub-score of 1. Using the Mk1 as a representative FHR, the enrichment of TRISO fuel used is planned to be 19.8% ^{235}U [26], which results in an MA score of 1 to account for this HALEU-level enrichment. Overall, the sum score for the FHR concept is 3, tied with GCR/HTGR/VHTR and GFRs.

4. Priority Assessment Results

A summary of the sub-scores and priority number assignments for each advanced reactor design can be found in Table 5. The results of this assessment show that liquid-fueled MSRs should be a top priority when considering future nuclear forensic signatures research due to the high technical maturity, intermediate material attractiveness, and high signature research and development strain of the MSR design. Following closely behind, at priority number 2 are LMRs, mostly due to their high number of planned reactors leading to a high TM sub-score. Three reactor designs are tied for priority number 3: GCRs, FHRs, and GFRs. Finally, priority number 6 is the SCWR design due to its low technical maturity and signature RDS.

Table 5. Results of the priority assessment summarizing the three sub-scores, their sum, and the assigned priority of each advanced reactor design.

Reactor Design	**Technical Maturity**		**Material Attractiveness**		**Signature R&D Strain**		**Sum Score**	**Priority Number**
Molten salt reactor (MSR)	2	+	1	+	2	=	5	1
Liquid-metal-cooled reactor (LMR)	2	+	1	+	1	=	4	2
Gas-cooled reactor (GCR)	1	+	1	+	1	=	3	3
Fluoride-cooled high-temperature reactor (FHR)	1	+	1	+	1	=	3	3
Gas-cooled fast reactor (GFR)	0	+	2	+	1	=	3	3
Supercritical water-cooled reactor (SCWR)	0	+	1	+	0	=	1	6

It is worth noting that while the technical maturity sub-scores of the MSR and LMR are equal, the weighted rank of LMR did score higher than that of the MSR. In this work, each sub-score was intentionally given equal weight by using a three-tiered score. The authors believe that this accurately portrays the importance of each sub-score in a nuclear forensic lens; however, in any future iterations or uses of this priority scoring system, experimenting with adjusted weighting would allow the application of different lenses. For example, the TM and MA sub-scores are also relevant for safeguards, and higher weighting on those categories could adjust the priority for a safeguards audience.

Additionally, note the three-way tie between GCR, GFR, and FHR. While these reactor concepts are found to be of the same priority, they arrived at a priority number of 3 in different ways. The GFR received the highest material attractiveness sub-score while receiving the lowest technical maturity of the three reactor concepts. This means there are fewer opportunities for material to be diverted from the GFR and fuel cycle at present, but as time progresses and the GFR design becomes more technically mature, its priority will likely increase, surpassing that of GCR, FHR, and potentially even LMR.

While this evaluation shows the MSR as a high priority for signature research and development, the rankings will most likely change over time as certain reactor designs become more (or less) popular. At present, it is vital to begin research on signatures of molten salts to maintain preparedness in the nuclear forensic community as advanced reactors begin to hit the commercial market. In the future, this evaluation should be updated to continue to inform the direction of signatures research.

5. Conclusions

Interest and investment in advanced reactors have increased in the past decade, and this exciting development for nuclear energy must be met with readiness by the nuclear forensic community. The deployment of advanced reactors and their novel fuel cycles may produce new sources of nuclear material diversion. In this work, a semi-quantitative research priority assessment was performed on six advanced reactor designs (GFR, LMR, GCR, FHR, SCWR, and MSR) based on the technical maturity of the design, the attractiveness of the fuel materials, and the status of research and development of relevant nuclear forensic signatures. Molten salt reactors were found to be the highest priority (priority number 1) for further signatures research, followed by LMRs. The designs GCR, FHR, and GFR tied for third, while SCWR was assigned the lowest priority (priority number 6). Signatures of advanced reactor materials are relatively unexplored, and the field is ripe with opportunities for discovery. With this in mind, future research efforts should initially focus on developing signatures for materials within MSR and LMR fuel cycles to maintain preparedness in a shifting nuclear energy landscape.

The assessment performed in this work provides insight into the current status of signature development for advanced reactor designs; however, the priority scoring method may be applied in future assessments as well. Additional developments of this method could involve the full quantification of sub-scores, for example, an in-depth analysis of relevant signatures for the signature RDS sub-score. Furthermore, this work assumes that selected design examples of each advanced reactor are representative of the average proposed design. As more information becomes publicly available, this assumption should be re-evaluated, and additional designs should be considered as necessary.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/jne5040032/s1, Figure S1: Molten salt reactor class hierarchy; Table S1: Advanced reactor fuel examples for material attractiveness sub-score. References [5,26,34,39,43] are cited in the Supplementary Materials.

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