Transactions, **SMiRT-26**
Berlin/Potsdam, Germany, July 10-15, 2022
Special Session: Performance assessment of spent fuel in storage and transportation

Cooperation in Advanced Technologies Associated with the Back End of Nuclear Fuel Cycle

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ABSTRACT

A Memorandum of Understanding (MOU) between Argonne National Laboratory (Argonne) and the German Bundesanstalt für Materialforschung und -prüfung (BAM) was signed in October 2014. Its objectives are to promote cooperation among scientists and specialists at Argonne and BAM and establish a framework for collaboration in advanced technologies associated with the back end of the nuclear fuel cycle. Collaborative activities involving Argonne and BAM may be implemented through the promotion of joint research activities and scientific workshops and conferences; exchange of technical information; and visits by scientists, specialists, and graduate, postgraduate, and Ph.D. students. The MOU was renewed in January 2020 for another five years. Highlights of the progress and achievements are provided in identified topical areas for collaboration on ageing management guidance, storage, transportation and disposal R&D, joint conference activities, and conclude with the benefits of the MOU in fostering Argonne and BAM collaboration.

INTRODUCTION

A Memorandum of Understanding (MOU) between Argonne National Laboratory (Argonne) and the Bundesanstalt für Materialforschung und-prüfung (BAM) was signed in October 2014. Its objectives are to promote cooperation among scientists and specialists at Argonne and BAM and establish a framework for collaboration on advanced technologies associated with the back end of the nuclear fuel cycle. Collaborative activities involving Argonne and BAM may be implemented through the promotion of joint research activities and scientific workshops and conferences; exchange of technical information; and visits by scientists, specialists, and graduate, postgraduate, and Ph.D. students. The MOU was renewed in January 2020 for another five years. Major topical areas of collaboration identified in the MOU are: (1) Guidance documents on aging management programs, (2) mechanical properties of high-burnup fuel claddings, (3) long-term performance of metallic and elastomeric O-ring seals, (4) polymeric neutron shielding materials, (5) modelling and simulations of structural and thermal performance of spent fuel and dry cask storage systems during storage and post-storage transportation, (6) advanced surveillance technologies for the monitoring and inspection of dry cask storage systems during storage and subsequent transportation, and (7) geological repository engineering technology (e.g., disposable containers), which is a new topical area included in the renewed MOU (2020–2025).

Highlights during the first MOU collaborations (2015–2019) can be found in Refs. [1, 2]. BAM and Argonne researchers have also published and shared information on the above topical areas (1) to (6) at the 2013, 2016, and 2019 International Conferences on Packaging and Transportation of Radioactive Materials (PATRAM) and at the Annual American Society for Mechanical Engineers (ASME) Pressure Vessels & Piping (PVP) Conferences, as well as through participation on the spent fuel management subcommittees and working groups sponsored by the United States Electric Power Research Institute

(EPRI) and the International Atomic Energy Agency (IAEA). Two books: *Safe and Secure Transport and Storage of Radioactive Materials* [3] and *The Ageing of Materials and Structures, towards Scientific Based Solutions for the Ageing of Our Assets* [4] also contain chapters contributed by Argonne and BAM authors.

Because of the pandemic in 2020 and 2021, all regular meetings and conferences were either cancelled, delayed, or held virtual online. The pandemic, however, has not stopped collaboration, as the virtual platform was found effective for project discussions involving researchers at different locations around the world. An example is the IAEA Cooperative Research Project (CRP) on Ageing Management Programs for Spent Fuel Dry Storage Systems. Two Consultancy Meetings and the Third Research Coordination Meeting were held online in 2020 and 2021, with participants from nine countries to discuss progress towards completion of the final report [5], which will be published by IAEA in 2022. Both Argonne and BAM researchers have contributed chapters and sections in the final report.

In this paper, progress and highlight achievements in selected topical areas are summarized and the plan for future collaboration is briefly described.

AGEING MANAGEMENT GUIDANCE

Argonne and BAM researchers have supported the development of guidance documents on aging management programs for spent fuel dry cask storage systems (DCSS) in both countries and Independent Spent Fuel Storage Installations (ISFSI) in the United States. Argonne prepared a topical report *Managing Aging Effects on Dry Cask Storage Systems for Extended Long-Term Storage and Transportation of Used Fuel* [6] for the U.S. Department of Energy (USDOE), which was referenced by the U.S. Nuclear Regulatory Commission (USNRC) in *Managing Aging Processes in Storage (MAPS) Report – Final Report* [7], providing the technical basis for the *Standard Review Plan for Renewal of Specific License and Licenses and Certificates of Compliance for Dry Storage of Spent Nuclear Fuel – Final Report* [8], the *Standard Review Plan for Spent Fuel Dry Storage Systems and Facilities – Final Report* [9], and the *Standard Review Plan for Transportation Packages for Spent Fuel and Radioactive Material – Final Report* [10].

The U.S. nuclear industry has incorporated the DOE/Argonne ageing management report [6], by reference, into NEI 14-03 [11], which was mentioned in NUREG-1927 [8], containing “tollgates” as periodic points within the period of extended storage when licensees would be required to evaluate aggregate feedback and perform and document a safety assessment that confirms the safe storage of spent fuel. Tollgates are described as an additional set of in-service assessments beyond the normal continual assessment of operating experience, research, monitoring, and inspections on the structure, system, and components (SSC) that are important to safety for DCSS and ISFSI during extended period of storage operation. NEI 14-03 also describes a framework for the aggregation and dissemination of operating experience across the industry through the use of an ageing-related operating experience “clearinghouse,” titled the ISFSI Aging Management Institute of Nuclear Power Operations Database (ISFSI AMID).

Two guidance documents issued by the German Nuclear Waste Management Commission (ESK) are: *Guidelines for Performance of Periodic Safety Reviews and on Technical Ageing Management for Storage Facilities for Spent Fuel and Heat-Generating Radioactive Waste* [12] and *Guidelines for Dry Cask Storage of Spent Fuel and Heat-Generating Waste* [13]. These two guidance documents address the technical concept and safety requirements for all dry cask storage systems for spent nuclear fuel and high-level waste in Germany, as well as the requirements and recommendations that are considered by the German licensing authority, the Federal Office for the Safety of Nuclear Waste Management (BASE). While the descriptions and details in the ageing management guidance documents may vary, the periodic safety review of the storage facility mentioned in Ref. [12] shares the same basic philosophy that applies to the periodic examination of industry and site-specific operating experience mentioned in the DOE/Argonne ageing management report [6], as well as in NUREG-1927 [8] and NEI 14-03 [11]. The IAEA document *Storage of Spent Nuclear Fuel, Safety Standards, Specific Safety Guide* SSG-15 [14] address the safety requirements and the design and review of dry storage systems and facilities for spent nuclear fuel. All of these guidance documents are generally reviewed and updated over time to incorporate changes in regulations, national standards and codes, and technology and advances in knowledge, so they should be followed for changes and updates as well.

STORAGE, TRANSPORTATION AND DISPOSAL R&D − SHARING INFORMATION

Each country's approach to the management of the backend of fuel cycle waste varies depending on the country's waste management policy, reprocessing or disposal; and on technology, wet or dry storage, and subsequent transportation via road, rail, or waterways. Choices of dry storage locations, at-reactor or away-from-reactor, and storage environment, indoor or outdoor, also vary, as reflected in each country's approach to guidance on ageing management programs. The need to store spent fuel in dry cask storage systems longer than initially intended is universally recognized, however; therefore, effective aging management programs are required to reduce risk, ensure public safety and health and protect the environment. There are more than a dozen U.S. dry cask storage systems (DCSS) designs that are of two general types: (1) self-contained shielded metallic casks with bolted closure without an overpack and (2) metallic canisters with a separate overpack to provide radiation shielding and physical protection [6]. In Germany, dual-purpose cask (DPC) design is used for storage and transport operations [12, 13], and there are differences in materials used to construct the casks (e.g., ductile cast iron or forged steel for DPCs and stainless steel and carbon steel, or concrete, for canisters, bolted casks, and overpacks in the DCSSs. There are also similarities in the functional materials used in the DPCs and DCSSs (e.g., metallic and elastomeric O-ring seals and polymeric neutron shielding materials). DPCs and DCSSs contain spent fuel assemblies and internal baskets and share the same concerns about maintaining the spent-fuel configuration, cladding integrity, and fuel retrievability during long-term storage and subsequent transportation. Whereas all of the DPCs in Germany are stored indoors (at the 12 on-site and 2 former central spent-fuel storage facilities as well as at Jülich Research Center for AVR-fuel and Interim Storage North for the Russian pressurized water reactor (Voda Ernergo Reactor [VVER]) fuel, the DCSSs in the United States are placed outdoors on concrete pads at 71 ISFSIs in 34 states. Two license applications for a Consolidated Interim Storage Facility (CISF) are under review by the USNRC since 2016 and 2017; both applications address concerns involving ageing management of canisters previously loaded at various ISFSIs across the U.S., transportation of these canisters to the CISF, and their continued interim dry storage at the CISF, the so called "72-71-72" issues in the U.S. for which inspection and monitoring of canisters and DCSS are the key to verifying the integrity of canisters in storage and for transportation. The IAEA Regulations for the Safe Transport of Radioactive Material SSR-6 [15] also includes requirements in Para 106, Para 503(e), Para 613A, Para 809 (f) and 809 (k) on ageing considerations for packages under "shipment after storage."

Among the topical areas identified for collaborative R&D between Argonne and BAM on storage, transport, and disposal, four areas have received continuing attention and activities and they are: (1) Mechanical properties of high-burnup (HBU) fuel cladding, (2) long-term performance of elastomeric O-ring seals, (3) modelling and simulations of structural and thermal performance, and (4) pressure and temperature monitoring for dry cask storage systems. Highlights in these areas are provided below referencing relevant and recently published literatures, as appropriate.

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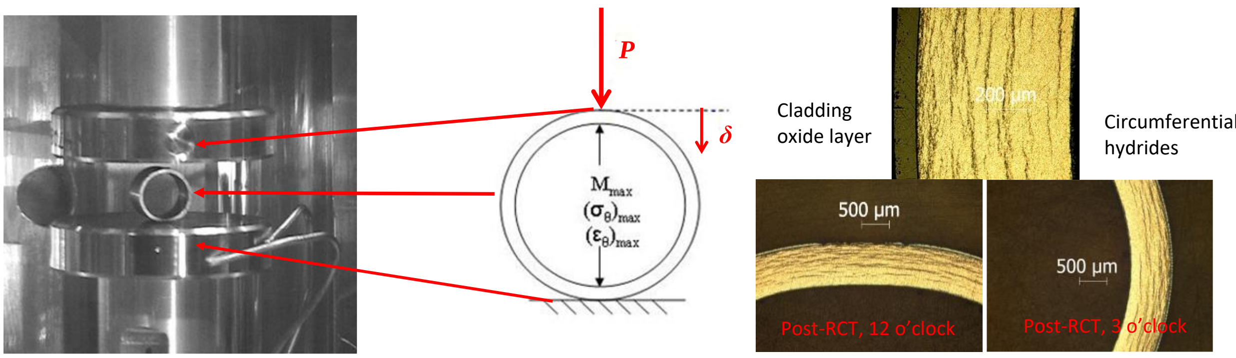
Mechanical properties of high-burnup fuel cladding

Structural analyses of high-burnup (HBU) fuel cladding require cladding mechanical properties and failure limits to assess its performance during long-term storage and subsequent transportation. Pre-storage drying and transfer operations, as well as early-stage storage, can subject the HBU cladding to higher temperatures and much higher hoop stresses than those associated with in-reactor operation and pool storage. Under these conditions, radial hydrides may precipitate during slow cooling that may result in embrittlement as the cladding temperature decreases below a ductile-to-brittle transition temperature

(DBTT). Argonne researchers have developed a test protocol for studying HBU cladding embrittlement that has been accepted by the USNRC. Experimentally, the protocol involves two steps: (1) radial-hydride treatment (RHT), during which the HBU cladding specimen is exposed to simulated drying-storage temperature and hoop stress conditions, and (2) ring compression testing (RCT), for which the HBU RHT-cladding ring specimens are subjected to hoop bending stresses, to determine strength and ductility as functions of test temperature. The RCT is used as a ductility screening test to simulate pinch-type loading on the HBU cladding, which occurs during normal conditions of cask transport and/or drop accidents [16-21]. Figure 1(a) shows the RCT of a HBU as-irradiated Zry-4 cladding ring specimen under loading P with displacement (δ)-control; M_{max} is the maximum bending moment, $(\sigma_\theta)_{max}$ and $(\varepsilon_\theta)_{max}$ are the maximum



(a) (b)

Figure 1. (a) Illustration of RCT of a HBU as-irradiated Zry-4 cladding ring specimen, and (b) cladding outer-surface oxide layer, circumferential hydrides, and post-RCT metallographic images at 12 o'clock and 3 o'clock.

hoop stress and hoop strain, respectively. Figure 1(b) shows metallographic images of pre- and post-RCT of a HBU Zry-4 cladding ring specimen with circumferential hydrides and a cladding outer-surface oxide layer. The RCT results showed that the HBU Zry-4 cladding with circumferential hydrides was ductile. The finite element structural analysis results showed that in the elastic regime, the outer surface oxide layer kept its integrity and was able to bear the compression load; whereas cracks were developed later in the oxide layer as indicated in the post-RCT images at 12 o'clock versus 3 o'clock in Fig. 1(b) [22].

The relationship between the hydride morphology, in particular the presence of radial hydrides, and the failure mechanisms associated to the RCT were investigated on a mock-up material by BAM and Universidad Politécnica de Madrid in Spain [23]. Samples of unirradiated ZIRLO® cladding were pre-hydrided and subjected to a RHT. A considerable fraction of radial hydrides was generated, the radial hydride continuity factor being around 80 to 90% of the wall thickness. The samples with reoriented hydrides were tested using the RCT at room temperature. Macroscopic brittle failure was observed with sudden load drops associated to unstable crack propagation events through the radial hydride network. It was found that the failure micro-mechanism is quasi-cleavage in the hydrides and micro-void nucleation, growth and coalescence in the zirconium matrix, with ductile tearing patches connecting neighbouring hydrides. The interaction between the location and continuity of radial hydrides and the hoop stress determines cladding failure in the presence of radial hydrides. These results are being used to describe the failure mechanisms of HBU cladding. The paper "Brittle Failure Analysis of High-Burnup PWR Fuel Cladding Alloys," by BAM and Argonne co-authors K. Simbruner, M. C. Billone, U. Zencker, Y.Y. Liu, and H. Völzke, has been accepted for presentation at SMiRT-26 [24].

Long-term performance of elastomeric O-ring seals

Elastomers are widely used as sealing materials in packages and containers for low and intermediate level radioactive wastes and as an additional component to metallic seals in bolted casks for spent fuel and high-level waste. Such "auxiliary" seals enable proper leakage rate testing of the metal barrier seals. Maintaining the confinement/containment boundary for the bolted dry cask storage systems during long-term storage and transportation requires understanding of the ageing mechanisms that affect the long-term performance of elastomeric O-ring seals concerning their function as auxiliary seals. The work conducted by the BAM researchers on elastomeric seals since 2016 can be found in Refs. [25-29]; the two most recent journal articles, both published in 2020, are "Erroneous or Arrhenius: A Degradation Rate-Based Model for EPDM during Homogeneous Ageing" [28] and "Insights for lifetime predictions of O-ring seals from five-year long term aging tests" [29]. Accelerated ageing tests of elastomeric seals under operation-relevant conditions, coupled with physics-based modelling simulation, are essential to the development of reliable prediction methods for long-term performance of elastomeric O-ring seals used for storage and transportation casks.

Modelling and simulations of structural and thermal performance

Modelling and simulations of structural and thermal performance of spent fuel and dry cask storage systems are important to the development of ageing management programs for long-term storage and subsequent transportation for storage and final disposal. Examples of modelling and simulations of structural performance of HBU fuel cladding have been provided by Han et. al [22], using finite-element code ABAQUS® that included the RCT machine stiffness, elastic and anisotropic plastic stress-strain properties, and large strain capability to simulate the HBU cladding ring specimens subjected to RCT hoop-bending loading. Examples of modelling and simulations of thermal performance of dry cask storage systems are provided by Li et. al [30-33], using both finite-element code ANSYS and computational fluid dynamics (CFD) code FLUENT to simulate temperature and flow fields in dry cask storage systems during vacuum drying and long-term storage. Another CFD code, STAR-CCM+, has been used by Argonne researchers since 2019 to simulate the temperature and gas flow fields in helium and air gas leakage experiments conducted by using a 1/4.5-scale model cask at the Central Research Institute of Electric Power Industry (CRIEPI), Japan. The STAR-CCM+ is implemented on a client-server environment, using high-performance computing resources at the Argonne National Laboratory Computing Resource Center.

Pressure and temperature monitoring

Pressure and temperature monitoring systems are employed to help augment inspections, or potentially reduce the frequency of (or eliminate) inspections. Continuous and periodic condition and/or performance monitoring would enable timely detection of ageing effects, such as pressure drop and/or leakage from the confinement boundary of bolted casks or welded canisters, thus allowing mitigation and repair to continue effective ageing management of dry cask storage systems and reduce risks to public safety, health and protect the environment. The primary monitoring systems that are in common usage today are pressure-monitoring systems for bolted-cask systems and temperature-monitoring systems for both bolted-cask and welded-canister systems. The pressure-monitoring system for bolted-casks is a surveillance measure to identify a hypothetical metal seal failure either of the inner primary lid system or the outer secondary lid system. The double-barrier design allows for monitoring the pressure level in the intermediate cavity and to send a signal in case a critical pressure loss may occur followed by inspection routines to identity which seal or the monitoring device itself has failed. Subsequent repair routines consider exchange of the monitoring device or a metal seal of the secondary lid system. Just in case of a primary seal failure a seal change cannot be performed without moving the cask to a hot cell or a nuclear power plant pool (which are usually not available in an interim storage facility) and a third repair lid has to be assembled on top of the secondary lid to re-establish a two-barrier-lid-system.

Temperature-monitoring systems are employed on bolted-cask and welded-canister systems to help augment walkdowns to detect blockage of inlet vents of dry cask storage systems. Vent-monitoring systems have been implemented and operational for several dry cask storage systems in the U.S. and natural convection monitoring of storage buildings in Germany for many years and have a robust history of performance. The monitoring methodology for detection of helium gas leakage from canisters based on changes in temperatures at different locations has been developed and demonstrated in the 1/4.5-scale model gas leakage experiments [34-38]; however, practical temperature-monitoring systems for actual industry canister helium-leakage detection have not been implemented to date.

Remote area modular monitoring for temperature measurement, RAMM-TM, is an innovative remote monitoring device that can detect gas leakage from canisters containing spent nuclear fuel by measuring changes in surface temperature differences between the top and bottom of the canister. The performance of RAMM-TM in detecting canister gas leakage from a small, simulated chloride-induced stress corrosion crack (CISCC) has been demonstrated in experiments using a 1/4.5-scale model cask. Both helium and air gas leakage from a canister were detected within hours after the start of the leakage. The change in surface temperatures at the top and bottom center of the canister (ΔTBT) during gas leakage (depressurization) triggered automatic alarms, providing a sound basis for early detection of gas leakage from the canister. Advanced data analytics and high-performance computing STAR-CCM+ CFD simulations are being developed for demonstration and application of RAMM-TM in actual dry cask storage systems [39]. When deployed as a distributed remote sensing and monitoring system, the RAMM-TM methodology would allow consequence management through the implementation of mitigatory actions to continue effective ageing management and to reduce risks to public safety, health, and protect the environment [40].

Geological repository engineering technology

Geological repository engineering technology is an additional topical area included for collaborative R&D between Argonne and BAM researchers. Disposable containers for spent fuel and high-level waste in a geological repository or deep borehole involve many facets of geological repository engineering technology, such as drilling, container emplacement, monitoring, thermal and structural analyses, sealant for borehole closure, to name a few. Argonne researchers were involved in a USDOE program on universal canister design for disposal of small waste forms in a deep borehole in crystalline rock. A universal canister-based system can be used for handling these wastes during the disposition process, covering storage, transfers, transportation, to facilitate the eventual disposal of these wastes [41]. BAM has performed a research study on "Requirements and Concepts for Containers for the Final Disposal of Heat-generating Radioactive Waste and Spent Fuel Assemblies in Rock Salt, Claystone and Crystalline Rock" (project KoBrA) together with BGE Technology GmbH and funded by the German Ministry for Economic Affairs and Climate Protection. Outcomes were published in a final report [42] and will be presented at the upcoming IGD-TP (Implementing Geological Disposal Technical Platform) Symposium in September in Zurich, Switzerland. In the KoBrA research project, requirements for high-level radioactive waste disposal containers were derived for the first time systematically and under consideration of the international state of the art. This included determination and summarizing the container-relevant boundary conditions and load parameters for disposal containers in the three potential host rocks rock salt, claystone, and crystalline rock. Based on this, the container functions necessary to fulfil the requirements under the given impacts and stresses were determined and considering a step-by-step approach from the general to the site-specific. Finally, potential suitable container concepts were derived and discussed. A major aspect in this context is the consideration of already existing DPCs and whether they can be upgraded for final disposal.

JOINT CONFERENCE ACTIVITIES

Argonne and BAM researchers regularly attend and contribute technical papers at international conferences, such as the annual ASME Pressure Vessel and Piping (PVP) Conference, the Institute of Nuclear Material Management Annual Meeting, the American Nuclear Society International High Level Radioactive Waste Management Conference, the IAEA conferences, and the International Symposium on Packaging and Transportation of Radioactive Materials, which is held once every 3 years. For the ASME PVP conference, Argonne and BAM researchers have been involved in the Operations, Applications, and Components Committee since 2011 serving as Technical Representatives, organized and chaired many technical sessions over the years. The Special Session on performance assessment of spent fuel in storage and transportation for SMiRT-26 is another example of joint conference activities by Argonne and BAM that also include contributions from Japan, Spain, and IAEA. Senior staff of Argonne and BAM also attend the Electric Power Research Institute (EPRI)'s Extended Storage Collaboration Program (ESCP) meetings, during which side meetings are held on these occasions to discuss progress and plan future collaborative activities. During the 2nd Research Coordination Meeting hosted by Argonne for the IAEA Cooperative Research Project on Ageing Management Programs for Spent Fuel Dry Storage Systems, April 29-May 3, 2019, the BAM and Argonne participants discussed collaboration on mechanical properties of HBU fuel cladding and modelling and simulation of structural performance using finite element analysis codes. These joint conference activities and collaborative work between Argonne and BAM are expected to continue into the future.

SUMMARY AND CONCLUSIONS

Six major topical areas have been identified for continuing collaboration between Argonne and BAM researchers on advanced technologies for the backend of the nuclear fuel cycle. Among them highlights of progress and achievements are provided in five topical areas on (1) Ageing management guidance, (2) Mechanical properties of HBU fuel cladding, (3) Long-term performance of elastomeric O-ring seals, (4) Modelling and simulations of structural and thermal performance, and (5) Pressure and temperature monitoring. Geological repository engineering technology is a new topical area identified for collaboration, which involves disposable containers for geological repository and deep boreholes for nuclear waste disposal.

Collaborations of scientists and specialists from Argonne and BAM are expected to yield benefits not only to their respective institutions and government sponsors, but also to other countries and the international community that shares concerns on ageing management programs for extended long-term storage of spent nuclear fuel and subsequent transportation. In 2009, EPRI established the ESCP in the United States. The EPRI ESCP international subcommittee, formed in 2010, held multiple biannual meetings between 2010 and November 2021, in-person as well as virtual in 2020 and 2021. There were over 400 ESCP members from more than 20 countries, and subcommittees covering international collaboration, concrete, chloride-induced stress corrosion cracking, fuels, HBU demonstration, non-destructive examinations, and repair and replacement. The primary goal of ESCP is to share information among its members to identify shared R&D needs and potential areas for "formal" collaboration outside ESCP. The MOU between Argonne and BAM on collaboration in advanced technologies associated with the back end of the fuel cycle is an example of an outgrowth of the ESCP activities that also enhanced the collaboration in the IAEA Cooperative Research Project on Ageing Management Programs for Spent Fuel Dry Storage Systems.

ACKNOWLEDGMENTS

The authors would like to acknowledge contributions from their colleagues at Argonne and BAM whose work are referenced in this paper. Argonne National Laboratory's work was sponsored by the U.S. Department of Energy under Contract No. DE-AC02-06CH11357. The U.S. Government retains for itself, and others acting on its behalf, a paid-up, nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.

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