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Source Term Evaluation for a Spent Fuel Reprocessing Facility

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INTRODUCTION

Spent fuel reprocessing is a backend fuel cycle option that has been employed in several countries. In the U.S., there is no spent fuel reprocessing facility currently in operation. However, the U.S. has had two facilities that were built in the mid-1960s to early 1970s. The Atomic Energy Commission, the predecessor of Nuclear Regulatory Commission (NRC), used 10 CFR Part 50 to grant an operating license for the Nuclear Fuel Services reprocessing facility at West Valley, New York, in 1966 and to issue a construction permit for the Barnwell Nuclear Fuel Plant (BNFP) Separation Facility in 1970 [1]. Only the West Valley facility was operated for a period of time. The BNFP was built but never operated.

After more than forty years since the advent of the West Valley facility, the current regulation still requires that spent fuel reprocessing facilities must comply with 10 CFR Part 50 to obtain a construction or operating license [1]. 10 CFR Part 50.34, which is to ensure adequate protection of public health and safety, requires that applicants must demonstrate their designs meet general design criteria and are capable of mitigating a postulated set of accidents, known as “design-basis” accidents, and that radiological exposure limits for the public can be maintained.

Now, 40+ years after the last reprocessing plant was licensed, licensing, building, and operating a new reprocessing plant is being considered. To close this 40+ year gap, the NRC must prepare to review an applicant’s license submittal and to conduct an independent safety review.

Over the years since World War II, there have been a number of accidents, ranging from minor to extensive structural damage and large radioactive contamination at reprocessing facilities [1, 2, 3, 4]. The domestic and some of the nondomestic accidents were at defense reprocessing facilities; others were at commercial reprocessing facilities for spent nuclear fuel. The ability to estimate radioactive releases from a reprocessing facility is necessary for the NRC to determine that the source term is within limits identified by regulation in order to grant construction and operating licenses. The release of radioactive materials into the atmosphere is often referred to as a “source term,” and that terminology is used in this summary.

Aqueous extraction technology is well established at the commercial scale [1, 5, 6, 7]. In fact, most commercial spent fuel reprocessing facilities in the world are based on aqueous extraction technology. France and Japan focus on advanced aqueous extraction cycles that allow more refined separation of fission product types to reduce waste volumes. However, extra extraction steps may increase accident frequencies. Many of steps in these advanced cycles are either new or do not have sufficient operating histories to estimate accident frequencies, so there is some uncertainty in the risk of operating these advanced facilities.

Safety improvements in aqueous extraction based on operating years and the lessons learned from the past accidents have ranked this technology as safer than the pyro-processing technology, with the possible exception of the advanced aqueous extraction cycles that have less certain accident frequencies. The lessons learned allow regulators to impose strict requirements on facility designers and operators to reduce the frequency of accidents.

DEVELOPMENT OF A SOURCE-TERM TOOL

Potential Approaches to Evaluating Source Terms

Evaluation of source terms at a spent nuclear fuel reprocessing facility must first consider a number of accident types that can lead to releases within the confinement building. Each accident type has a distinct set of mechanisms that need to be evaluated in some manner to determine the characteristics of the release. For each accident type, an instantaneous or time-dependent insertion of mass and energy into a compartment or room of the reprocessing facility must be evaluated. The mass insertion can be in the form of vapor or aerosol particles or a combination of both. Furthermore, because aerosol transport depends on aerosol size and density, the aerosols need to be characterized with a size distribution. Energy insertion affects room or compartment temperature and pressure, which in turn affects the rates and pathways that vapors and aerosols may take in escaping from the facility into the atmosphere. Finally, if the accident type is initiated by a highly energetic event, walls, ventilation ducts, doors, and other building structures may be altered. This in turn affects the rates and pathways that vapors and aerosols may take in escaping into the environment.

In previous safety analyses for fuel reprocessing and other nuclear facilities at the Department of Energy (DOE), analysts estimated source terms using the DOE Handbook [8], an assumed aerosol size distribution, and a safety system code (e.g., typically MELCOR, which is included in the DOE toolbox of recommended codes) to estimate the fractional release into the environment, which is referred to as the leak path factor (LPF) [9]. DOE Handbook values were empirically determined from laboratory-scale experiments and simplified approaches in the 1970s and 1980s. Especially for explosions and fires, extrapolation from the small-scale laboratory experiments to a reprocessing facility may not be appropriate. When large-scale experimental data are not available, computational fluid dynamics (CFD) approaches may be more reasonable than extrapolating bench-scale experimental data. Smoothed particle hydrodynamics can be used to model the expulsion and breakup of liquid globules initially contained in a vessel when an explosion occurs. Data from accidents at reprocessing facilities may allow for model validation to improve confidence in CFD results.

A safety systems code (SSC) is needed to evaluate the fraction of the radionuclides released into a room or compartment of a facility that ultimately gets released into the atmosphere. The SSC must treat aerosol agglomeration, aerosol drop evaporation and condensation, aerosol deposition onto surfaces, and transport of the aerosols through building rooms and compartments and through a ventilation system. The SSC code needs to treat pressure and thermally driven flows, system features such as sprays, and filters on ventilation systems. DOE analysts have typically used MELCOR as the SSC because it has all of the features needed to evaluate LPF), which is essentially the fraction of the material released into an internal compartment of a facility that is ultimately released into the atmosphere outside the facility.

Significant Accident Types

Explosions – Chemical explosions are the most energetic events that can occur in a reprocessing plant, and thus tend to create the largest source terms. Important aspects of a chemical explosion for source terms are

- rapid release of chemical energy, generally from within a closed vessel,
- rupture of the vessel when the pressure pulse from the explosion is sufficient,
- expulsion of liquid solutions from within the vessel,
- creation of aerosols by breakup of the expelled liquid globules,
- change of phase,
- aerosol mechanics, including agglomeration, evaporation/condensation, and deposition of aerosols onto surfaces, and
- potential damage to walls, doors, and the ventilation system.

The basis used in previous analyses for the amount and size distribution of aerosols formed as the result of an explosion within a vessel are correlations taken from the DOE Handbook [8]. An alternative is to replace these correlations with one derived from the CFD approach that is briefly described above and elaborated in a companion paper [10]. A separate analysis or use of a correlation may be needed to determine whether the explosion breaks walls, opens doors, or reduces the integrity of the ventilation system. Once the initial mass and size distribution of aerosols and vapors has been determined, a SSC would be used to calculate the ultimate release into the atmosphere.

A side effect of an explosion is that a portion of the expelled liquids are likely to end up on the floor of a room or compartment of the facility. Depending on the conditions, the flammable liquids may catch fire. If that happens, a fire would also need to be modeled as described in the following paragraphs.

Fires – Fires can occur following an explosion or from a spill or pipe leakage. Fires can release a significant amount of energy but over a longer time period than a chemical explosion. Important phenomena that are expected to occur during a fire are

- change of phase of combustible fluids,
- achieving flammable conditions by mixing with air,
- release of energy, soot, and formation of radioactive aerosols during the fire, and
- agglomeration, evaporation/condensation, and deposition of the aerosols.

Traditionally, a fire code has been used to determine the rate of energy and soot insertion into a room or compartment as a function of time. Fission products dissolved in the solvent are assumed to be released into the room in proportion to the amount of solvent consumed by the fire. A similar approach is envisioned for the model being developed. A SSC code can then be used to estimate the source term into the atmosphere based on the mass and energy insertion into a compartment of the reprocessing facility.

A side effect of a fire in a compartment is that it may heat up a vessel to the point of inducing a chemical explosion. If this occurs, the explosion would need to be modeled as outlined above.

Nuclear excursion (Criticality) – In an aqueous extraction, the presence of both fissile and neutron moderators (such as hydrocarbons and water) can induce an inadvertent criticality event. Reprocessing facilities are designed to minimize the possibility of a criticality, but it is impossible to entirely eliminate this possibility under all possible off-normal conditions. If a criticality accident occurs, nuclear fission provides a source of energy that is inserted into the vessel containing the fissile materials. The number of fissions that occur along with other parameters defining the vessel characteristics determine whether the vessel containing the fissile material might fail. If the energy released by the criticality event is sufficient to fail the vessel, liquid can be expelled from the vessel. This situation would be treated similarly as an explosion. A lower-energy criticality event may vaporize some of the solvent, which would carry with it some of the fission products in solution. If this were to occur, some of the fission products could escape into a room or compartment of the facility. A SSC code would then be used to track the aerosol mechanics and transport from a room or compartment and into the atmosphere.

Spills – This accident type is usually associated with corrosion of a pipe or vessel that leads to a spill onto the floor of a room or compartment. Correlations from the DOE Handbook [8] can be used to estimate the quantity and size distribution of aerosols generated. CFD approaches, as outlined above could also be used to improve the estimation of the quantity and size distribution of aerosols. The mechanics and transport of the aerosols inserted into a room or compartment would be treated with a SSC code.

Spills of flammable liquids can also lead to fires, which would be treated as described above. As mentioned previously, fires can lead to explosions. This sequence of events, a spill leading to a fire leading to an explosion, can be treated by combining these individual events.

SUMMARY

Interest in building a reprocessing facility is reviving in the US. Licensing such a facility requires the NRC to perform independent reviews of the accidents and resulting consequences that could occur at a reprocessing facility. A significant part of the review is to evaluate potential source terms from a variety of accident types. The work described here lays out a framework for developing a tool that can be used to provide the NRC with a means to independently evaluate potential source terms from a reprocessing facility.

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