ROADMAPPING THE RESOLUTION OF GAS GENERATION ISSUES IN PACKAGES CONTAINING RADIOACTIVE WASTE/MATERIALS – A STATUS REPORT

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ABSTRACT

Gas generation issues, particularly hydrogen, have been an area of concern for the transport and storage of radioactive materials and waste in the Department of Energy (DOE) Complex. Potentially combustible gases can be generated through a variety of reactions, including chemical reactions and radiolytic decomposition of hydrogen-containing materials. Since transportation regulations prohibit shipment of explosives and radioactive materials together, it was decided that hydrogen generation was a problem that warranted the execution of a high-level roadmapping effort.

This paper discusses the major gas generation issues within the DOE Complex and the research that has been and is being conducted by the transuranic (TRU) waste, nuclear materials, and spent nuclear fuels (SNF) programs within DOE's Environmental Management (EM) organization to address gas generation concerns. This paper presents a "program level" roadmap that links technology development to program needs and identifies the probability of success in an effort to understand the programmatic risk associated with the issue of gas generation. This paper also presents the status of the roadmap and follow-up activities.

The "program level" roadmapping involved linking technology development (and deployment) efforts to the programs' needs and requirements for dispositioning the material/waste that generates gas through radiolysis and chemical decomposition. The roadmapping effort focused on needed technical support to the baselines (and to alternatives to the baselines) where the probability of success is low (high uncertainty) and the consequences of failure are relatively high (high programmatic risk). A second purpose for roadmapping was to provide the basis for coordinating sharing of "lessons learned" from research and development (R&D) efforts across DOE programs to increase efficiency and effectiveness in addressing gas generation issues.

The roadmap effort found that gas generation issues can adversely affect DOE milestones in a variety of programs at different sites and that gas generation issues represent a large risk to accomplishing DOE's environmental management mission to clean up DOE sites.

Some of the salient 'lessons learned' from preparing the roadmap include:

- Need to involve subject matter experts in a team setting to help the technologists and those having the needs to better determine the timing and specific requirements for the R&D activities. This results in better synergy, better communication, and speedier development of the roadmap and roadmapping activities
- For activities that have a high risk to their successful completion, it would be wise to develop some contingency planning, either of alternative technologies or alternative programmatic activities that have a possibility to solve the need(s)

It was concluded that continued use of roadmapping will be useful in keeping focused on the efforts necessary to mitigate the risk in the disposition pathways and to respond to the specific needs of the DOE programs and sites that have gas generation issues that affect shipping and storage of nuclear materials and waste.

The following is a brief summary of the status of some of the key activities on the roadmap:

- Nuclear Materials that generate hydrogen gas – Approval was given to use the 9975 shipping package for shipment of Nuclear Materials that meet the 3013 standard. The SAFEKEG shipping package will not be available prior to about June of this year. Shipments have not yet started to Savannah River Site (SRS). There is concern that there will be sufficient packages available for all the shipping needs, especially for Rocky Flats
- Orphan Nuclear Materials – There is concern that these materials will not be able to meet the standards needed for packaging and shipment. This could impact Rocky Flats closure schedules
- Gas getters – 'Getters' were demonstrated to have application to radioactive material packages, effectively mitigating hydrogen buildup within sealed environments. Progress was made in developing hydrogen getters for limited on-site transportation at SRS and for potential application in the TRUPACT-II for transporting TRU waste
- Reduce layers of confinement – Process was re-engineered, the concept was demonstrated, and now in process of developing a deployable package by the end of September 2002
- Permeable membrane R&D – Tests were done for higher temperatures and lower pressures. It was found that the membrane does not work well under lower pressures. Also, helium used for an inert atmosphere in the packaging clogs up the membrane

INTRODUCTION

To ensure that the various gas generation activities within the Office of Environmental Management were being properly integrated, Mr. David Huizenga, DOE EM-20 and Mr. Gerald Boyd, DOE EM-50, established a Task Group to develop an integrated gas generation research and development (R&D) plan (i.e., a "program level" roadmap). Members of the Task Group included DOE and contractor representatives from: the Nuclear Materials (NM) programs, which includes the Nuclear Material Focus Area (NMFA), the Nuclear Materials Stewardship Program, and the 94-1 Program; the National Transportation Program (NTP); the Spent Nuclear Fuel (SNF) program; the TRU Waste program, including the Transuranic and Mixed Waste Focus Area (TMFA); and site personnel associated with these programs. It is anticipated that future assessments will integrate data from the high-level radioactive waste (HLW) and mixed/low-level radioactive waste M/LLW programs to better determine if those programs also have gas generation issues.

The roadmap that was developed identifies the major gas generation programmatic issues within the DOE complex and the research that has been and is being conducted to address gas generation concerns.

This paper also presents the current status of key activities identified in the roadmapping.

PURPOSE

The "program level" roadmapping effort was intended to focus needed technical support to the baselines (i.e., the major steps for final disposition of waste/material) where the probability of success is low (high uncertainty) and the consequences of failure are relatively high (high

programmatic risk). The roadmap identified where emphasis is needed, i.e., areas where investments are large, the return on investment is high, or the timing is crucial for dispositioning the waste or material. A second purpose of the roadmapping was to provide the basis for coordinating sharing of "lessons learned" from R&D efforts across DOE programs to increase efficiency and effectiveness in addressing gas generation issues.

ROADMAPPING APPROACH AND METHODOLOGY

Development of the roadmap involved identifying the major steps needed for final disposition of the waste/material (or for storage pending disposition) and the associated R&D and certification activities required to ensure the viability of each step. In a typical disposition pathway, four major functions are needed: (1) treatment, (2) packaging, (3) transportation, and (4) disposal/storage. Each of these functions was examined to determine what technical support would be needed to make the function successful. The timing to have the technology in place was also captured to identify those areas where emphasis should be placed or where resources should be reallocated.

ROADMAP SUMMARY EXPLANATION

The gas generation roadmap summary (shown as Figures 1-4) show at a glance the functions involved to disposition a waste/material type, the R&D and also regulatory activities associated with successfully completing a function, plus the current condition of the functions and activities for each of the programs evaluated. This roadmap summary is designed to assist decision-makers by quickly showing how issue resolution activities are logically related to disposition functions and how resolution activities enable a function to begin or improve, which assists in directing decision-maker focus towards resolution of the issues.

The timeline column identifies four dimensions for an activity: (1) risk, (2) time, (3) type of activity, and (4) possibility for sharing across programs. Risk is depicted by the colors and time is shown by the location of an icon in relation to the timeline. The shape of the icon, as defined in the legend, shows the activity type. Figure 1 is marked up for this paper to highlight the view to show that a function and a milestone are in jeopardy.

It is essential the roadmap summary be colored. This added dimension provides significant information by identifying where emphasis and management involvement is needed. The colors indicate a level of risk as described by the following color definitions:

- *Red* indicates there is significant risk to achieving success. For example, the activity lacks funding, basic assumptions that provide the foundation for the activity or function are wrong, the regulator indicates that activity may be unacceptable, or the critical path appears to be unachievable unless heroic efforts are made

- *Yellow* designates moderate risk. For example, the activity is behind schedule, the success of the activity is suspect (funding, schedule, regulatory approval, politics), an aggressive assumption has not been proven, or some combination of factors

- *Green* indicates low risk. For example, an activity/function has funding, priority, and expects to meet its milestones and goals

- *Blue* describes where a process improvement happens. Examples are: an increase in process efficiency, reduction in cost, schedule improvements, and increased safety

- *Striping* shows that the function is 'at risk' unless, and until, the resolution activity is successfully completed. The color of the striping is indicative of the coloring of the resolution activity (indicating level of risk) feeding into the function

The Legend, located at the bottom of the roadmap summary, shows symbols for milestones, completion times for R&D activities, regulator approval times, and "at risk" activity flags. The flags are meant to be an added guide to direct decision-makers to focus on specific areas. An important symbol is the purple diamond surrounding an R&D icon. It designates a possible opportunity for programs to share information by virtue that they may be similar enough or have lessons-learned that another program can use for assistance. On the bottom row with the color definitions is a two-tier, two colored icon showing a performance improvement area. When a function has an increase in productivity or throughput because of an R&D activity, this icon shows it.

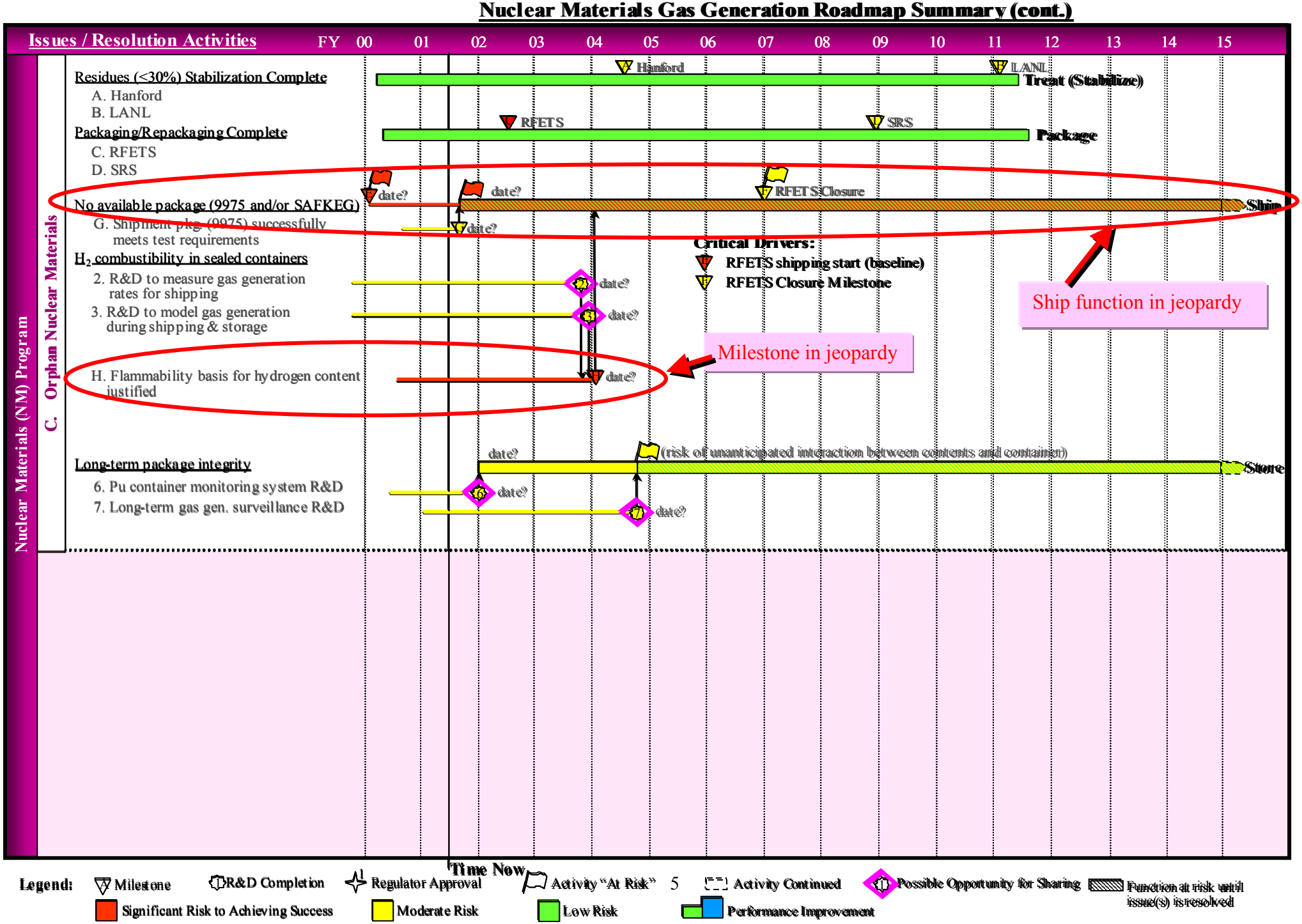

Fig. 1. Nuclear Materials Gas Generation Roadmap Summary (cont.)

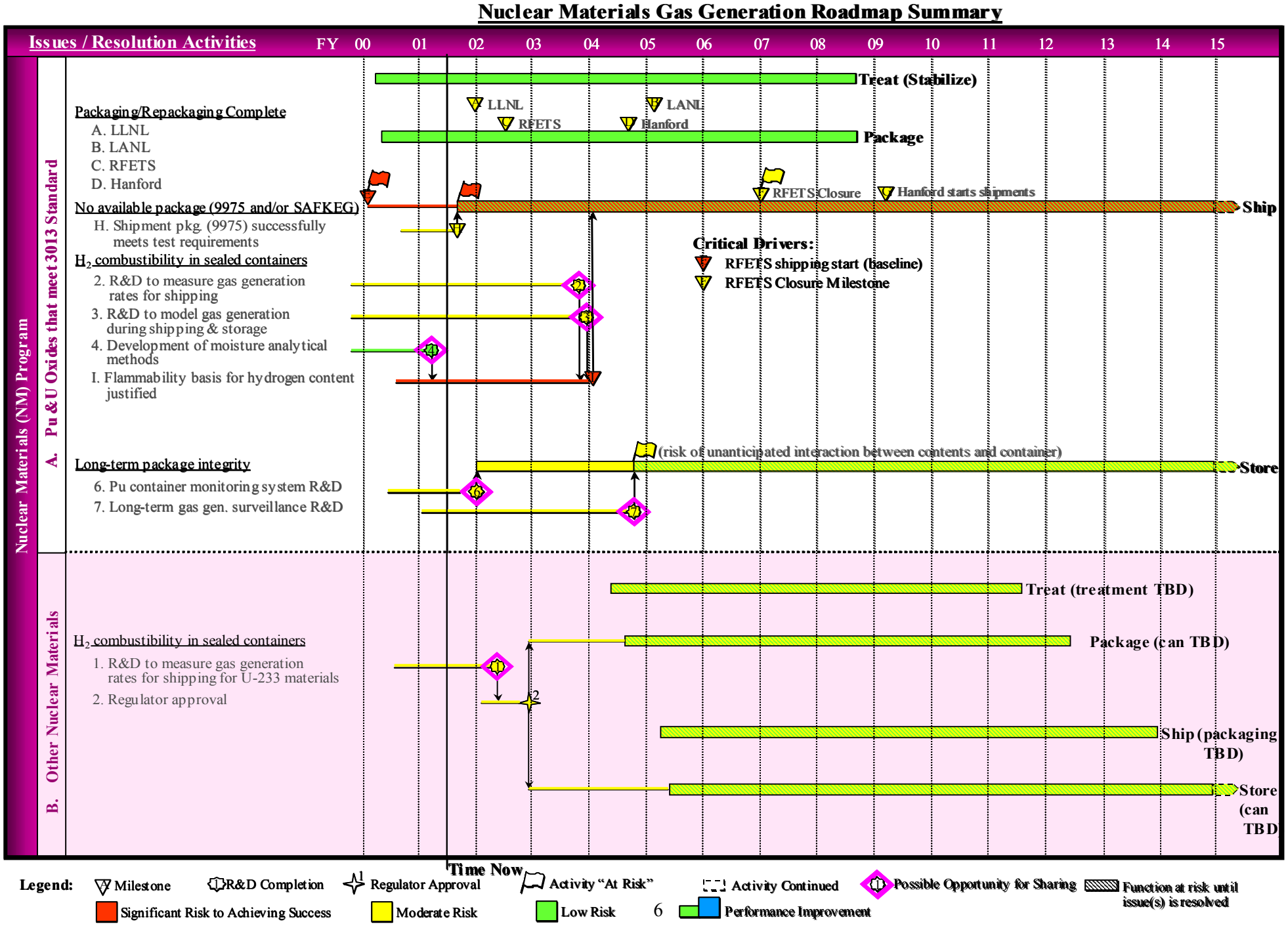

Fig. 2. Nuclear Materials Gas Generation Roadmap Summary (cont.)

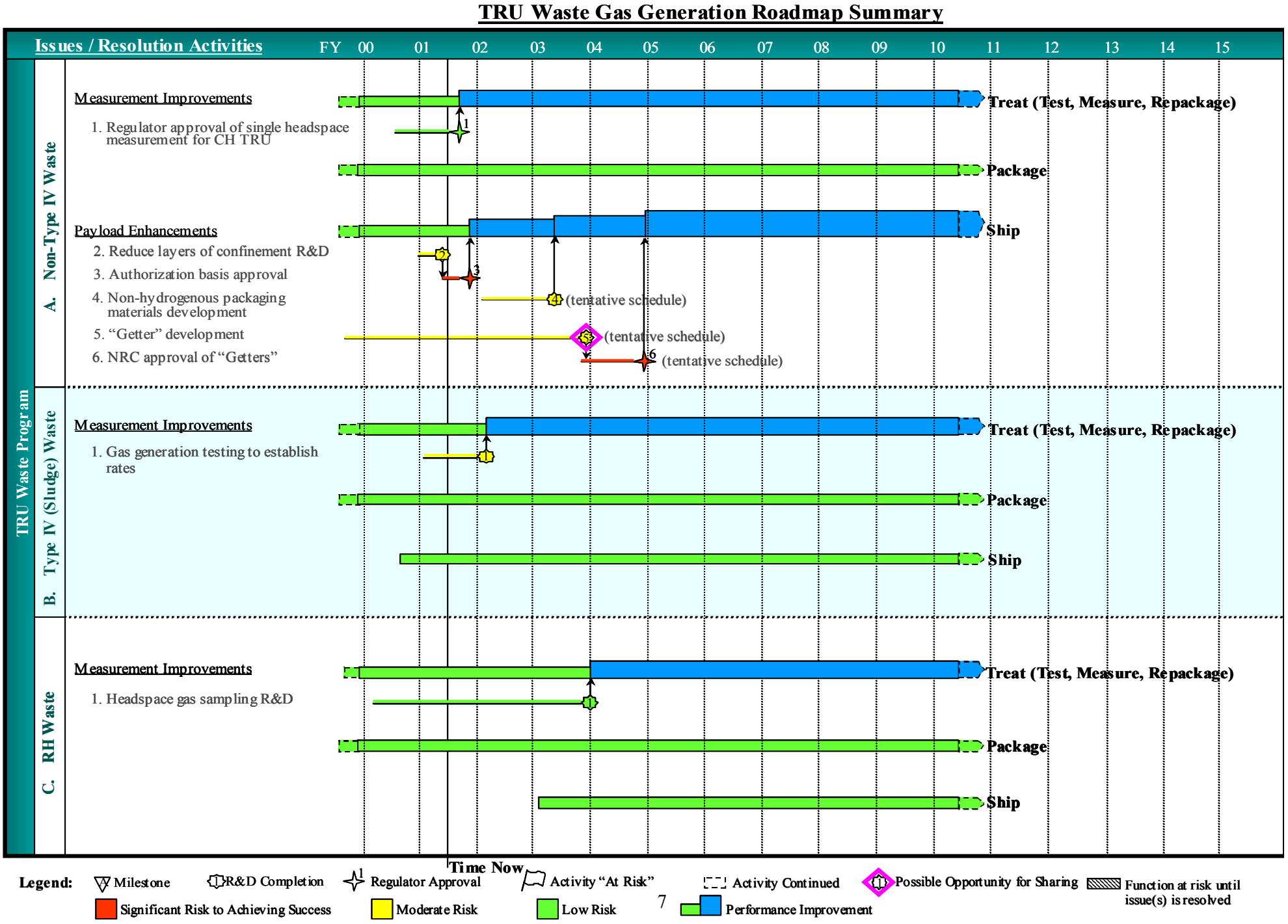

Fig. 3. TRU Waste Gas Generation Roadmap Summary

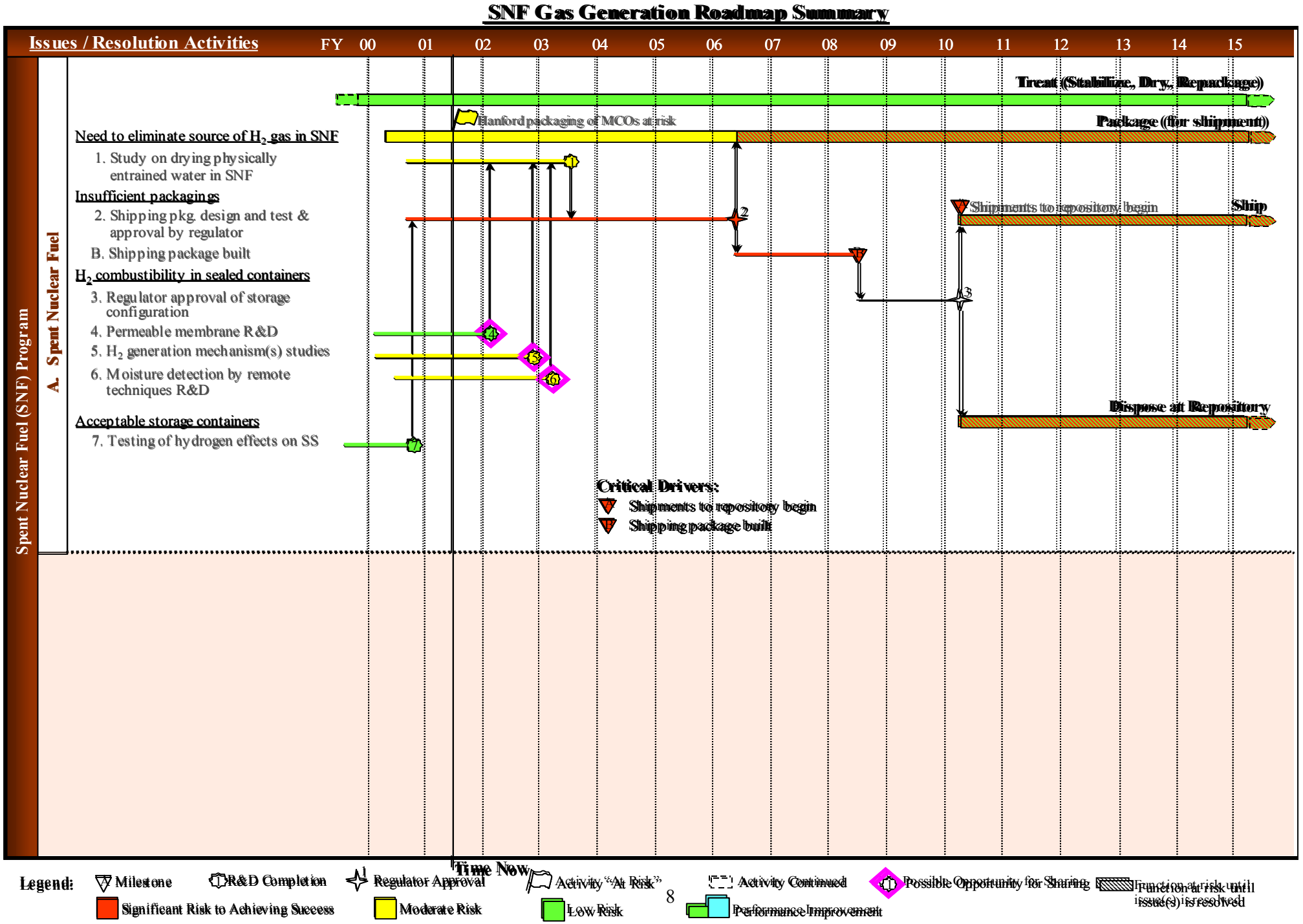

Fig. 4. SNF Gas Generation Roadmap Summary.

FINDINGS

By performing the roadmapping effort, the Task Group found that:

- There are no significant overlaps in planned R&D activities, nor appreciable duplication of R&D activities already performed
- There are many opportunities for sharing information about gas generation issues and potential solutions. Areas of greatest potential cooperation appear to be in sharing methods, approaches, and strategies
- Some nuclear materials, such as some impure oxides and residues, do not have a clear disposition pathway. It appears that these orphan nuclear materials need priority attention to make sure shipping packages can be certified, shipments can be approved and expedited, and pathways for disposition can be quickly developed (or the Rocky Flats Environmental Technology Site (RFETS) closure schedule needs to be revised)
- There are pathways that have significant risk, which may indicate more emphasis should be placed on contingency planning, for example, getters development and impure oxides and residues disposition
- For the RFETS shipping schedule to be met, both the SAFKEG and 9975 packagings must be certified, as they are both needed for transportation campaigns of nuclear material
- The baseline schedule for shipping of SNF to a repository is very much in jeopardy due to anticipated lack of funding for continued development of a transportation packaging (cask)
- The TRU program R&D activities being performed are to enhance performance of existing functions e.g., to allow greater amounts of waste in the shipping package thus decreasing transportation costs and schedule

LESSONS LEARNED

- Need to involve subject matter experts in a team setting to help the technologists and those having the needs to better determine the timing and specific requirements for the R&D activities. This results in better synergy, better communication, and speedier development of the roadmap and roadmapping activities

- For cross-program type activities, a lead needs to be specified and given authority to cause actions and recommendations to be carried out

- It was shown that 'programmatic' type activities are also important to address on a technology roadmap, since they are often an integral part of accomplishment and implementation of the R&D activity

- For activities that have a high risk to their successful completion, it would be wise to develop some contingency planning, either of alternative technologies or alternative programmatic activities that have a possibility to solve the need(s)

STATUS OF ACTIVITIES

TRU WASTE

TRU – Non-Type IV Waste

- Reduce layers of confinement – Process was re-engineered and the concept demonstrated last summer. Currently in process of developing a deployable package. It is planned to have this task done by end of September 2002 (about 1-1/2 years later than shown on the roadmap graphic). Participants are very optimistic that the process will work as planned

- Non-hydrogenous packaging materials development – Currently on hold. There are no plans to do anything on this task in the near future due to funding shortfalls and having a lower priority

- Getter development – This R&D task is currently in the bench-scale testing phase (Phase II). Completion is expected by April 2002. Phase III (large-scale) testing is expected to be complete by December 2002. The data will then be submitted to the NRC either as a proposed revision to the TRUPACT-II Safety Analysis Report (Rev. 21) or to begin discussions with the NRC on what additional information is needed to approve the use of getters. Testing so far on getter performance is very good. The schedule, as shown on the roadmap, looks to be on track

TRU – Type IV (Sludge) Waste

- Gas generation testing to establish rates – A peer review was held last fall. The task completion is projected to be sometime in FY-03, which is about 1 year later than that shown on the roadmap graphic

TRU – RH Waste

- Headspace gas sampling R&D – This task is complete, almost 2 years ahead of the projection

NUCLEAR MATERIALS

Pu & U Oxides that meet 3013 Standard

- Shipment packages (9975 and SAFKEG) meet test requirements and are available for shipping – Approval was given to use the 9975 shipping packaging for shipment of materials that meet the 3013 standard. Shipments have not yet started to SRS. The SAFKEG shipping packaging is currently undergoing certification review by AL/WSD. At this time EM-5 is no longer "in business." The target date for certification is April 2002. Confirmatory analyses by AL/WSD are complete and "Q0s" have been forwarded to LANL

- R&D to measure gas generation rates for shipping – (Unable to obtain current information)

- R&D to model gas generation during shipping & storage – Significant progress has been made in limiting the "theoretical" internal gas pressure in the 3013. At this time it appears that hydrogen gas pressures will be low, compared to the water vapor partial pressure caused by desorption of water from materials containing 0.5% moisture. A paper presenting the technical basis is in preparation. Related R&D is continuing, in part because the current results are still preliminary and incomplete

- Development of moisture analytical methods – This work is still on-going. However, TGA-MS (thermo-gravimetric analysis/ mass spectrometer) will likely be approved by April. Also, an evaluation of TGA-TFIR should be complete on roughly the same schedule. SCE (supercritical fluid extraction) is still under consideration and its applicability is expected to be better understood later this year. IGA (interstitial gas analysis) is also being considered. Completion is behind what was expected, but should not impact the milestones it supports

- Flammability basis for hydrogen content justified – This work is complete. It is currently believed that flammable mixtures cannot exist in the 3013 container when it is properly packaged

- Pu containers monitoring system and long-term gas gen. surveillance R&D – To date, approximately 3 years of data have been collected on some small-scale samples. Ten (10) of the "shelf-life" containers have been loaded and data collection begun. Such results as are available are being factored into gas generation considerations. Elements of this program will go on for as long as materials are stored in 3013 containers

Other Nuclear Materials

- R&D to measure gas generation rates for shipping for U-233 materials – This work is on-going, with no significant developments at this time

Orphan Nuclear Materials

There is no work going on with respect to orphan materials. It is likely that many of these will be sent to WIPP, for which definitive guidance is already available. For the rest, it is likely that their transportation will be accomplished on an "exception" basis, rather than attempting to amend SARPs to accommodate just a few packages of widely diverse materials.

The following is a list of the R&D activities shown on the roadmap graphic:

- Shipment packages (9975 and SAFKEG) meet test requirements and available for shipping

- R&D to measure gas generation rates for shipping

- R&D to model gas generation during shipping & storage

- Flammability basis for hydrogen content justified

- Pu container monitoring system R&D

- Long-term gas gen. surveillance R&D

SPENT NUCLEAR FUEL (SNF)

- Study on drying physically entrained water in SNF – This is on-going, no significant results, yet
- Shipping pkg. design and test & approval by regulator – Decision to transfer responsibility to RW for developing and procuring shipping packaging(s) for shipping SNF from DOE sites. Expecting to have packaging(s) supplied by commercial vendors. Timing for availability needs to be verified
- Shipping package built – See prior bullet
- Permeable membrane R&D – Done FY-01. The tests were done for higher temperatures and lower pressures. Found that the membrane does not work well under lower pressures. Helium used for an inert atmosphere also clogs up the membrane
- H2 generation mechanism(s) studies – This is on-going, no significant results, yet
- Moisture detection by remote techniques – This is on-going, no significant results, yet
- Testing of hydrogen effects on stainless steel – Done FY-00, no further work needed

OTHER

- The proposal to re-establish the ANSI 12.32 working group – Addressing the Standard for Gas Generation in Packages Used for the Storage or Transport of Radioactive Materials was discussed at the annual ANSI N14 management Committee meeting in April 2001. Moving forward with a standard for gas generation in packages was viewed as important, but not a high priority. Consequently, there has been no effort in developing a standard
- Sharing of technical information on gas generation in packaging – This was one of the recommendations from the initial Integrated Roadmap report. The National Transportation Program has planned a workshop as part of a DOE sponsored Packaging and Transportation symposium in January 2002 to address hydrogen generating plutonium dioxide contents. The gathering of DOE complex subject matter experts is an excellent opportunity to discuss pertinent issues
- Gas getters – Several approaches for elimination of gas generation or mitigation of impacts were pursued since the Integrated Roadmap report was released. Polymer getters (see above discussion of getters under TRU Waste section) were demonstrated to have application to radioactive material packages, effectively mitigating hydrogen buildup within sealed environments. Progress was made in developing hydrogen getters for limited on-site transportation at the Savannah River Site. Additionally, a polymer hydrogen getter material was extensively tested in FY-01 for potential application in the TRUPACT-II. Another approach for elimination of gas generation was the successful application, in the 9975 packaging, of using an inert atmosphere

RECOMMENDATIONS

The Task Group offered the following recommendations for consideration:

- There are four areas that have the most potential for sharing of information
  - Methods for measuring gas generation rates – Gas generation rates measurement methods, sampling methods, analysis methods, procedures, and even equipment for making these measurements are part of what can be shared among the programs in this area
  - Mechanisms for gas generation – Though gas generation mechanisms may differ due to the chemical and physical properties of the waste/materials in each program, the general mechanisms of radiolysis and some chemical interactions are the same or similar enough to warrant sharing of this information
  - Gas generation modeling methodologies – The approach used to develop a model for gas generation (or the structure of the model) may very well be applied to several different materials. A model would most likely require some modification as it is applied across different materials because gas generation mechanisms differ. Once developed, a model would require R&D data to demonstrate its applicability
  - Approaches for elimination of gas generation or mitigation of impacts:
    - Use of getters – If regulatory approval can be obtained, getters will allow greater quantities of TRU waste to be transported in each shipment. Although getters for materials other than TRU waste will likely be different because of the dissimilar environments in which they must operate, the processes for obtaining regulatory acceptance and the technical bases will likely be very much alike
    - Permeable membranes – If proven successful for "venting" of metal containers while maintaining their robustness, this technology is one that could potentially be shared by various programs within the DOE Complex
    - Drying/stabilization techniques – Drying/stabilization techniques are currently part of the planned or current baseline for preparing nuclear materials for long-term storage and transportation. These techniques, developed by the NM programs, should have applicability to the SNF program
    - Use of inert atmospheres – The NRC applies a simple 5% concentration limit to hydrogen in the container atmosphere. The DOE/EM regulator may be willing to consider instead a flammability limit for hydrogen content that considers limiting the oxygen concentration. The conditions that the regulator might impose, such as requiring the container to be opened in an inert atmosphere or requiring the package to survive more severe accidents would likely apply to different materials

The following were suggested ways of facilitating the sharing of gas generation information:

- Use of periodic symposium for sharing: Gathering the DOE Complex subject matter experts to openly discuss progress and issues are of immense value. Group dialogue most always leads to productive conclusions. This Task Group could be the nucleus of such a forum
- Reports 'clearinghouse' establishment: Charter an existing organization to serve as a 'clearinghouse' to collect and disseminate information on R&D and other activities that relate to gas generation throughout the DOE Complex to all entities that could gain from the sharing of this information

- Contingency planning

  There are pathways that are questionable single solution pathways. Therefore, the Task Group suggested that bottlenecks be defined and plans made as to how to mitigate them. Should a single pathway prove impassible, alternatives should be considered or put into place or the consequences will most likely be "missed" or "slipped" milestones. Therefore, it was recommended that the following should have contingency plans developed:

  - RFETS shipping schedule for impure Pu oxides and residues – The detailed schedules for shipment of nuclear materials from RFETS needs to be looked at in detail to determine if the baselines for shipping, development of the 9975 and SAFKEG shipping packagings, manufacture of the shipping packagings, and programmatic approvals, etc. are integrated and achievable. Other alternatives should be explored in case one of the links is delayed or unattainable
  - Use of getters to enhance shipping capabilities - Regulators do not currently accept getters in transportation packages. As a consequence, use of getters involves not only the R&D to identify, develop, demonstrate, and deploy them, but also involves convincing the regulator that they will perform as designed. Because of the real risk that the regulator may not approve the use of getters, the programs should plan for other contingencies
  - Use of permeable membranes – As with getters, the regulator may not accept the use of permeable membranes to vent hydrogen gas buildup in shipping and/or disposal containers. As with getters, the regulator acceptance of permeable membranes to vent hydrogen gas buildup in shipping and/or disposal containers is questionable since they do not currently allow any "vented" Type B packagings and may even question the applicability of "venting" to disposal. Contingency planning should be implemented to have alternatives in place if the regulators reject the concept or should the R&D prove to not work
  - SNF funding – Evaluate funding adequacy and timing of the SNF packagings to assure meeting shipping schedules and impacts of not meeting them

  The following were some suggested methods to help in identifying areas that are on critical pathways that need contingency planning:

  - Continue to use this roadmap as a planning tool for decision makers - The continued use of this roadmap would constitute good project management practices
  - Leverage the expertise and capability of the independent peer review group as mentioned in the following section
  - EM-20/EM-50 should assign responsibility to perform contingency planning to fold into the roadmap

- Establish an independent peer review group to:
  - Review proposed R&D and programmatic activities – This review would look at the details of the activities, including the schedule for development, to assure that the activity fully resolves the issue(s) or does so if combined with other activities
  - Perform quality checks on applications to regulators – Using experts in writing applications for packaging certification would streamline the now lengthy process of submittal to a regulatory agency, waiting for comments (usually in regards to issues that could be caught before handing the application to the regulator), and then responding to the issues and resubmitting. The expert group would provide invaluable advice that would most likely shorten the certification process

- Provide greater influence with the regulator(s) by facilitating submittals from the applicants to the regulators and by having a more consistent approach
- Assure life cycle planning – In some instances, dispositioning a waste/material included only either long-term storage or transportation. When designing and developing packagings, decision-makers should thoroughly develop the disposition path that takes the material/waste from current to end state. Once the disposition pathway has defined functions, a decision-making process should be used to ensure DOE resources are prudently employed

NEXT STEPS

Roadmapping efforts to date have proved very worthwhile. However, the roadmap will only continue to be relevant as it is used and updated with more detail. Programs will benefit from identifying issues and then tying them to research and development needs identified in the roadmap.

The roadmap effort found that gas generation issues can adversely affect DOE milestones in a variety of programs at different sites. It was also recognized that gas generation issues represent a large risk to accomplishing DOE's environmental management mission to clean up DOE sites.

The Task Group that created the roadmap recognized that current research efforts are based on identified needs, but that those efforts could be better coordinated to address the issues. The Task Group recommended that a group of experts from appropriate DOE programs continue to further develop the roadmap and prioritize issues and research efforts.