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Characterization Strategy Report for the Criticality Safety Issue

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June 1997

Prepared for the U.S. Department of Energy under Contract DE-AC06-76RLO 1830

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Hanford Tank Characterization and Safety Issue Resolution Project

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Summary

High-level radioactive waste from nuclear fuels processing is stored in underground waste storage tanks located in the tank farms on the Hanford Site. Waste in tank storage contains low concentrations of fissile isotopes, primarily U-235 and Pu-239. The composition and the distribution of the waste components within the storage environment is highly complex and not subject to easy investigation. An important safety concern is the preclusion of a self-sustaining neutron chain reaction, also known as a nuclear criticality. A thorough technical evaluation of processes, phenomena, and conditions is required to make sure that subcriticality will be ensured for both current and future tank operations. Subcriticality limits must be based on considerations of tank processes and take into account all chemical and geometrical phenomena that are occurring in the tanks. The important chemical and physical phenomena are those capable of influencing the mixing of fissile material and neutron absorbers such that the degree of subcriticality could be adversely impacted.

This report describes a logical approach to resolving the criticality safety issues in the Hanford waste tanks. The approach uses a structured logic diagram (SLD) to identify the characterization needed to quantify risk. The scope of this section of the report is limited to those branches of logic needed to quantify the risk associated with a criticality event occurring. The process is linked to a conceptual model that depicts key modes of failure which are linked to the SLD. Data that are needed include adequate knowledge of the chemical and geometric form of the materials of interest. This information is used to determine how much energy the waste would release in the various domains of the tank, the toxicity of the region associated with a criticality event, and the probability of the initiating criticality event. Different characterization options are identified, each providing a different uncertainty in the risk calculation. Recommendations include processing existing data through the SLD to estimate risk, developing models needed to link characterization information to risk, and examining correlations between the existing characterization approach and data needs. Particularly if the user determines that the criticality risk is unacceptable or cannot be sufficiently determined due to data uncertainty, the user has the option to utilize the SLD to identify places where the type and level of sophistication of additional characterization work or model development may be needed to provide the adequate decision-making information.

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1.0 Scope

The purpose of this document is to describe a structured logic diagram (SLD) and associated support information (e.g., characterization and modeling needs). The SLD addresses the resolution of risks resulting from a criticality event occurring in the Hanford waste tanks. This waste has been stored for over forty years in Hanford single-shell and double-shell tanks.

Criticality is just one of the six issues (criticality, flammable gases, organic solvents, organic complexants, tank integrity, and ferrocyanide) that were part of this project. These issues were identified for examination to determine whether they could contribute to an unacceptable risk. Information from this report has been integrated with the findings reported for the other five issues to assess overall tank safety due to waste contents. This report will also

- describe a scientifically defensible process to assess and resolve criticality risk
- identify the significant parameters requiring measurement to quantify the magnitude of criticality disruptive events. These events must be identified for risk assessment
- identify scenarios for significant parameter determinations. These scenarios should optimize characterization needs and resolve risk acceptability.

2.0 Introduction

Since 1944, high-level radioactive mixed waste has been accumulated in 177 underground storage tanks (149 single shell and 28 double shell) at Hanford. Much of the 61 million gallons of waste in these tanks came from the large quantities of chemical agents being manipulated in day-to-day operations associated with special nuclear materials production, support activities and recovery of radioactive elements, and special nuclear materials from spent fuels. As processes were applied, modified, or as new technology came on line, the composition of the wastes disposed to the tanks changed over decades of operation. Wastes from site laboratory operations and facility and equipment decontamination activities were also disposed of in the tanks. The composition (chemical, radiological) and distribution of these wastes were further confounded by periodic inter-tank transfers of waste, waste cascading, tank leakage, and waste aging (chemical and radiolytic decomposition and radioactive decay). Waste cascading was an operation where supernate from one tank was overflowed to a second tank as more slurry was added to the first tank. As a result of these various processes, individual tank waste composition can vary from simple (relatively homogeneous domains and simple chemistry) to extremely complex (multiple heterogeneous domains and highly complex chemistry that includes starting materials and degradation products).

3.0 Approach

The approach to construct the criticality structured logic diagram and identify the supporting information consisted of four steps. In step one, a team of scientists (see Appendix A for the list of team participants and their associated expertise) developed the branch of the structured logic diagram which asserts criticality is physically impossible.

Next, the team defined actions that are taken on the waste which have an effect on the reactivity of the waste tank, and the associated parameters that affect criticality. In general, the nine parameters affecting criticality are mass, absorbers, geometry, interaction, concentration, moderation, enrichment, reflection, and volume. For the waste tanks, the sources and quantities of significant materials (i.e., fuels, water, poisons, etc.) are identified as key parameters for determining the probability of initiating and sustaining a criticality event. Mitigation or intervention activities were identified as paths that could be taken to reduce the risk due to criticality to an acceptable level.

The third step involved determining the probability of a criticality event. This was determined to be related to the probability of a chemical form change event and the probability of a geometric form change event, which results in a critical configuration.

The final step examined the physical consequences associated with a criticality event occurring. The physical consequence would not only include direct injury, but an increase in radiation dose and exposure to toxic materials. The energy released in the event volume would not only include the energy density of the initiating criticality event, it would also include the energy released from a secondary event. Criticality can result in an internal initiating event for the other safety issues, likewise, other safety issues can result in an internal initiating event to a criticality in the tank. There is a connection between the criticality SLD and other task SLDs.

The team also recognized the complex chemistry (radionuclides, organics, and inorganics), physical form (gases, liquids, and solids), stratification (number and sequence of domains and their homogeneity), and their effects on initiating a criticality event. Important properties of the materials to consider in addressing the criticality safety issue include their chemical and geometric forms.

The team used these four elements to develop the SLD. A simplified version of the SLD is depicted in Figure 3.1. Output at the top of the logic diagram is information that can be used to determine whether the contents of a specific tank pose an unacceptable criticality safety risk. The user will then be able to evaluate the risk based on the accuracy and precision of the data, taking into account uncertainty of data used at different tier levels in the scheme and error propagation. If the risk is unacceptable or cannot sufficiently be determined based on the uncertainty in data accuracy and precision, then the user has the option to use the SLD to identify places where the type and level of sophistication of additional characterization work or model development may be needed to provide the adequate decision-making information.

Information on risk is derived from the application of a risk model that utilizes information on radionuclide/chemical dose to humans and the environment and the probability/frequency of external events and other internal events playing a role in initiating the criticality event. The radionuclide/chemical dose information is derived from exercising a pathway model using radionuclide/chemical event release data.

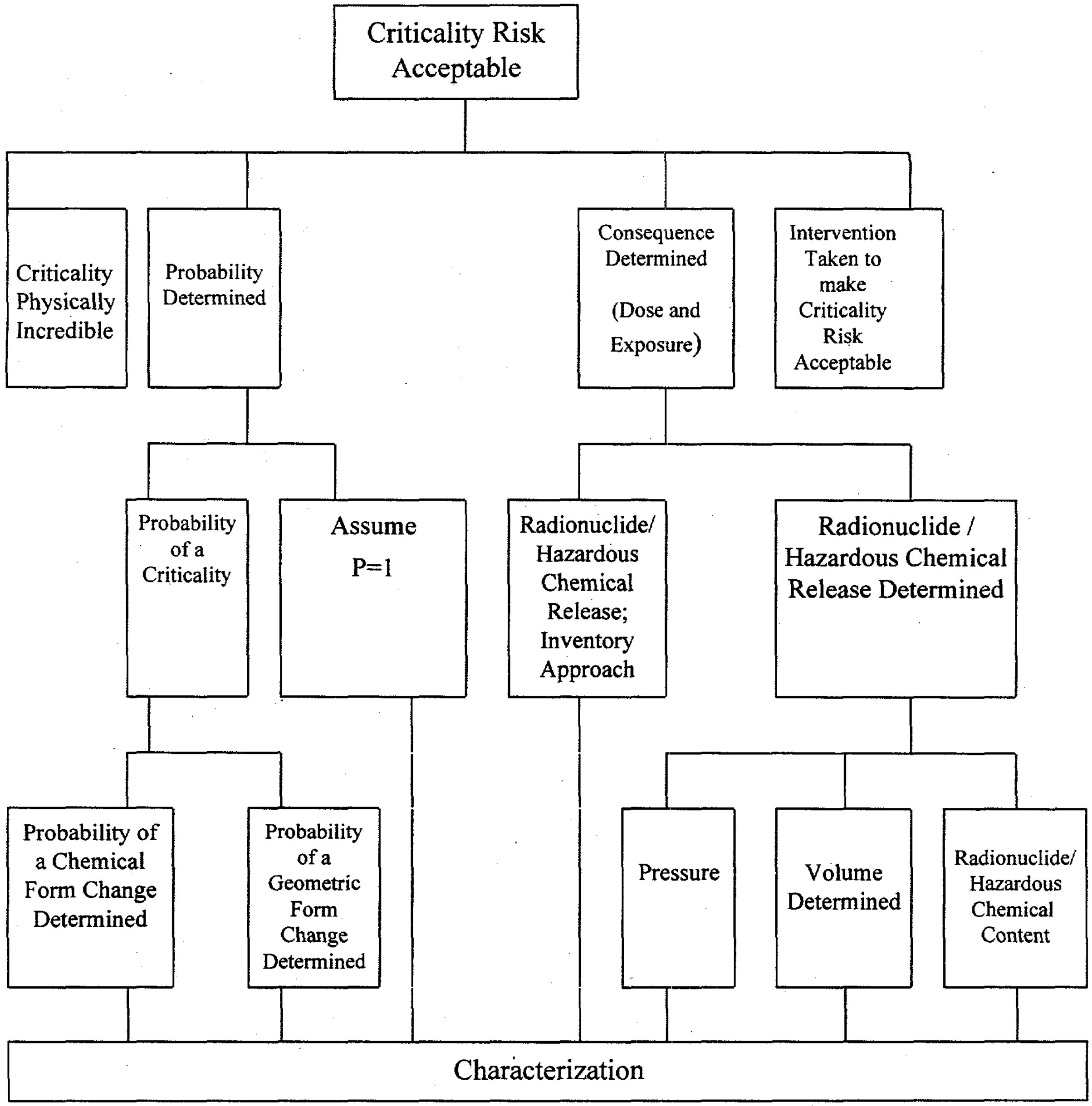


Figure 3.1. Simplified Criticality Structured Logic Diagram

The team determined that changes in geometric (including physical) form and chemical form of the material were significant parameters to be determined in predicting the probability of a criticality event occurring and the magnitude and consequence of the event. Two scenarios for determining these parameters were developed. In case 1, known as the bounding case, the range of applicable data such as total fissile and other materials content is assumed to be known and a bounding model is applied that provides an estimate of risk. In case two, a more rigorous explicit analysis is performed where data for chemical and geometric forms are derived from the available information.

A more detailed discussion of the specific elements of the logic scheme is described in Section 3. Figure 3.1 shows an overview of the criticality structured logic diagram associated with this report. A more detailed structured logic diagram is shown in Appendix B. This diagram indicates there are three paths illustrating that the criticality risk in a Hanford tank could be acceptable. Those are either to determine that criticality is physically impossible, the risk associated with the present tank is acceptable, or intervention makes the risk acceptable. In the detailed diagram, this is defined more precisely as a risk, agreed upon by regulators, managers and the general public, which is acceptable. When this risk level is defined in mathematical terms, this diagram (Figure 3.1) can be most useful and quantitative. Without a numerical risk factor, the logic in this diagram becomes largely qualitative.

Both radionuclide and toxic releases have similar logic associated with them. That is, the risks may be different, but the chances of either being released are related to similar phenomenon. For both types of releases, one could assume that all the contents of the tank could be released. This provides a bounding case. Certainly, if releasing all the contents of a tank were acceptable, then no one would need to calculate what a release might be in a real event. However, if the release of all the contents is unacceptable, then one would need to calculate the probability of releasing some portion of the tank contents and calculate the expected release.

The more detailed structured logic diagram shown in Appendix B is described more clearly in the following sections. Data needs are at the bottom of each branch of the logic diagram. These needs are defined in sufficient detail to agree with the logic. For example, one data need may be to determine the total mass of the radionuclides and fissile material in the tank. The group recognizes such a task could either be enormous or very simple. Determining the radionuclides in a tank could involve reviewing process data or obtaining screening information. For these examples, both the data and their uncertainties should be processed through the SLD.

The criticality SLD in Appendix B is described in detail in the following sections. Each section of the text (**C1-C10**) refers to continuation symbols on the criticality SLD in Appendix B.

3.1 Criticality Risk

The statement "The risk associated with having a criticality event in tank waste is acceptable" was the starting point of this task. The goal was to determine the data needed to be able to safely make this statement. Below the risk statement is a choice of three options. Either criticality is physically impossible, the risk is less than or equal to what is acceptable, or intervention is needed.

3.2 Risk to Workers/ Coworkers / Public / Environment (Section C1)

The risk gate (Risk is less than or equal to Risk $_{\text{acceptable}}$) is connected to a "conditional and" statement. This suggests that three things need to take place in the specified order. First, the probability and frequency of an event must be determined, then the consequences of an event must be determined, and finally, a model that calculates risk must be applied.

A criticality event could result in a disruptive event and a release of hazardous and/or toxic material to the environment. Examples of nondisruptive criticality events include localized increases in temperature and radiation that do not compromise the integrity of the tank.

3.3 Probability of a Criticality Calculationally Determined (Section C7)

The probability and frequency of an event occurring are related to the probability and frequency of an initiating event taking place, resulting in a change in either or both the chemical and geometric form of the fissile material region. These exact determinations can be avoided by assuming the probability of an initiating change that results in a criticality is unity. Initiating changes in the chemical or geometric form can either be from internal or external sources. External initiating events are brought from outside the tank boundaries. Examples include lightning and human activities.

3.4 Physical Consequences Calculated

The safety consequences of a criticality event can be determined if the consequences of the toxic and radioactivity exposure are determined. The greatest concern is the exposure from releases of radioactive and toxic materials from a criticality event. As evidenced from previous accidents, exposure after an event can have longer-lasting effects than the initial safety impact alone. Other consequences of a toxic materials and radioactivity release event might include damage to nearby buildings or tanks that may or may not cause an immediate safety concern.

The SLD branches associated with determining the radiation dose and the exposure to the toxic material are nearly identical. Therefore, this discussion will address one, not both. The logic gate below "radioactive material release determined" provides two options. Either the total tank contents can be assumed to be released, or the amount of release can be calculated by applying a radiactivity release model. The easiest option is to assume all material is released, and calculate whether this resulting risk is acceptable in the logic diagram. However, the more realistic case, that only part of the tank contents is released, requires a more detailed model calculation.

When the radioactivity release is calculated, not only does the amount of radioactivity in the source term in the event volume need to be determined, but so does the containment failure mode. These pieces of information can be entered into a fluid dynamics model to calculate how much material is released. The possibility of a pressure failure of the dome can be calculated if the mechanical properties, tank wall (dome) temperature, and dome pressure are determined. These pieces of information can be entered into a tank structural/mechanical failure model to identify whether a pressure failure will occur.

3.5 Intervention (Section C2)

In the event that the risk from criticality is found not to be acceptable, there are proposed mitigation or intervention activities that could be performed on the waste in the tanks to reduce the risk from criticality. They include adding or removing material from the tank and tank mixing. On any proposed intervention activity, the reduction in risk associated with criticality would need to be determined.

3.6 Chemical Form of the Fissile Material Determined (Section C3)

To calculate the amount of radioactive and toxic material that could be released in the event of a criticality, the chemical form of the material and the potential changes to the chemical form must be determined. The specific data needs are the total mass and volume of waste in the tank; the initial chemical form (speciation) of the fissile material and neutron absorbers in the solids, liquids, and gas phases in the tank; the oxidation-reduction potential (redox state); the temperature, thermodynamic, and kinetic data for species formation and interconversion; and the absorption properties for the sludge solids in regards to sequestering fissile material and neutron absorbers. Utilizing these types of data and applying the calculational model, the initial chemical form and the potential chemical form change can be determined. Some of the data for chemical form will change as a function of position and time, particularly if some action is taken on the waste.

3.7 Geometric Form of the Fissile Material Determined (Section C4)

To calculate the amount of radioactive and toxic material that would be released in a criticality event, the geometric form, physical state (solid, liquid, gases), and location of that material are important. The geometrical data needs are the total fuel mass and volume of the tank; the density and particle sizes; the fluid dynamics data, viscosity, tortuosity, porosity and temperature; the mechanical energy data, enthalpy, diffusion coefficient, and thermal conductivities. Utilizing these data and applying the calculational model, the initial geometric form and potential geometric form change can be determined. Some of the data for geometric form will change as a function of position and time. This change may occur with or without outside action being initiated.

The location of the event is important, because if the criticality event takes place well below the surface, then not only the material involved in the general location of the criticality, but also that material above the critical region is considered part of the "event volume." To determine the event volume, the compositions of both the localized region, which is critical, and the subcritical portions of the event volume need to be determined. In some cases, the primary event may trigger a secondary event, so both would need to be determined. Several models would be applied to this level to determine the actual concentration of radionuclides released in the case of an event.

3.8 Energy Release in Event Volume Determined (Section C5)

The energy released in the event volume is calculated based on the total number of fissions generated by the criticality event. To determine the total fissions, the chemical composition and the geometric form of the event volume must be determined and the criticality calculational model must be applied.

3.9 Radioactive Source Term in the Event Volume Determined (Section C6)

To calculate the radioactive material released in the event of a criticality, the location of that initial criticality event is significant. If the criticality were to occur well below the surface, then the material above the actual event location would also need to be included in the source term determination. In some cases, the primary criticality event may result in higher temperatures or other effects that may trigger a secondary event, so both would need to be determined. Several models would be applied to this level to determine the actual concentration of radionuclides released in the case of an event.

3.10 Toxic Chemical Exposure Determined (Section C8)

To calculate the amount of toxic or hazardous material released in the event of a criticality, the location of that initial criticality event is significant. If the criticality were to occur well below the surface, then the material above the actual event location region would also need to be included in the release. In some cases, the primary criticality event may result in higher temperatures or other effects that may trigger a secondary event, so both would need to be determined. Several models would be applied to this level to determine the actual concentration of toxic or hazardous chemicals released in the case of an event.

3.11 Bounding Analysis Performed (Section C9)

There are several different ways of estimating the probability of a chemical or a geometric form change occurring to cause a criticality event. In the diagram, two are shown. The simplest approach for determining the chemical form change (bounding case) is to determine the range of the chemical form data. For example, determine the range for the total mass and volume of waste in the tank; the initial chemical form of the fissile material and neutron absorbers in the solids, liquids, and gas phases in the tank; the oxidation-reduction potential, the temperature, thermodynamic, and kinetic data; and the absorption properties for the sludge solids. Utilizing these ranges and applying the calculational model, the potential chemical form change can be determined. This simplified approach can also be used for determining the geometrical form change (bounding case). For example, determine the range for the total fuel mass and volume of the tank; the density and particle sizes; the fluid dynamics data, viscosity, tortuosity, porosity, and temperature; the mechanical energy data, enthalpy, diffusion coefficient, and thermal conductivities. Utilizing these data ranges and applying the calculational model, the potential geometric form change can be determined.

Tanks known to have very low fissile material content may be evaluated in this manner and determined to be safe from a criticality event. However, the risk calculations based on this model may be unrealistically high, and values determined by this method may exceed acceptable limits. In that case, an alternative is to use a more explicit, rigorous model in which all components are carefully evaluated.

3.12 Explicit Analysis Performed (Section C10)

The most complex, but one of the most accurate methods for determining geometric and chemical forms in the tank, is to follow the rigorous approach. This approach involves the calculation of chemical and geometric form changes resulting in criticality and the resulting consequences and pressure changes based on extensive knowledge of the tank contents. Large pieces of information are needed to calculate this model. These are the explicit, exact data values for the data mentioned above. Boundary values and conditions may still be appropriate for certain data values, such as tank volume, temperature, and initial chemical form, but certain data values will need to be known with relatively little uncertainty.

4.0 Conclusions

This SLD indicates there are several paths that can be taken to determine if the risk of a criticality is at an acceptable level. The first option to performing exhaustive characterization would be to actively perform intervention to make criticality improbable or impossible. Another is to assume the probability of an initiating event occurring is equal to one. Both of these are conservative. In fact, all "simple" routes in the tree are conservative. This is as it should be. That is, we should not claim that there are lowered dangers in the radioactive waste tanks unless we have good evidence to support our estimates.

If any one of the criticality characterization results demonstrates the risk is acceptable, such as the chemical form precludes criticality or the geometric form precludes criticality, the tank poses no immediate safety concern. In addition, if the tank contains sufficient fissile material in the chemical form to allow criticality to occur, but the probability of the material forming into a geometrically favorable form for criticality is close to zero, the risk would also be very small. Also, if the probability of forming a geometrically favorable shape is high, and the pressure generated by a criticality event could damage the tank, but the amount of toxic or radioactive material in the tanks was within risk limits, then leaving the tank alone may be an acceptable practice. If, however, both chemical and geometric forms are found to be of concern, the risk of leaving the tank in its current condition could be unacceptable.

5.0 Recommendations

The reader of this document is cautioned that this report may be incomplete. The purpose of the report is to create a starting document such that additional information can be added. While this report was generated with care, and with highly trained experts on the chemistry and physics associated with criticality in Hanford tanks, the team does not claim that this SLD contains every possible scenario. However, as the SLD evolves, it will become more accurate and more useful for the ultimate resolution of the criticality safety issue.

In this report, there were three reasons for characterizing the waste in the Hanford tanks. These were to determine the probability of a criticality event; to determine the consequences of a criticality, such as the pressure generated by a sustaining event; and to evaluate how much toxic, hazardous, and radioactive material would be released in an event. All three of these require substantial knowledge of the chemical and geometric forms of the material in the waste. However, estimates can be made based on ranges or gross measurements. Such measurements will have an uncertainty associated with them. Evaluating the uncertainty through the SLD will determine whether these estimates are adequate to determine if the risk is greater or less than acceptable limits.

The next level of sophistication in characterization of the material could be in measuring data from a limited number of regions within the tank. These measurements will also contain certain inherent uncertainties, which taken together with the measured values will help evaluate whether the risk of a criticality in the Hanford waste tanks is acceptable. The advantage of determining the wastes' speciation is the clearer picture it provides in determining whether one of the several scenarios are likely.

Many of the Hanford tanks have already been categorized into several different areas. Placing tanks in categories can be useful, especially in relation to their determined risk. It would be useful, for example, to calculate the criticality risk of each tank based on existing data. The risk could then be used to generate a grading system for each tank. Those with the highest (or lowest) grade could have the greatest risk as determined by the SLD. Existing watch lists could be a starting point for developing these ratings.

New and existing data should be carefully examined with the SLD in mind. Much of the data generated to date may or may not achieve the goals identified in this SLD. Each tank should be evaluated separately to examine how well the existing data meets the needs defined in the SLD.

Appendix A

Contributors

Appendix A

Contributors

A.L. Doherty is the leader of the criticality subtask of the Hanford Tank Characterization and Safety Issue Resolution Project. Ann obtained her bachelor's degree in nuclear engineering in 1982 and master's degree from the University of Washington in 1988. Ann is a technical group manager of the Materials and Engineering Analysis Group at Pacific Northwest National Laboratory (PNNL). She has been with PNNL for 14 years and is also a Senior Criticality Safety Specialist.

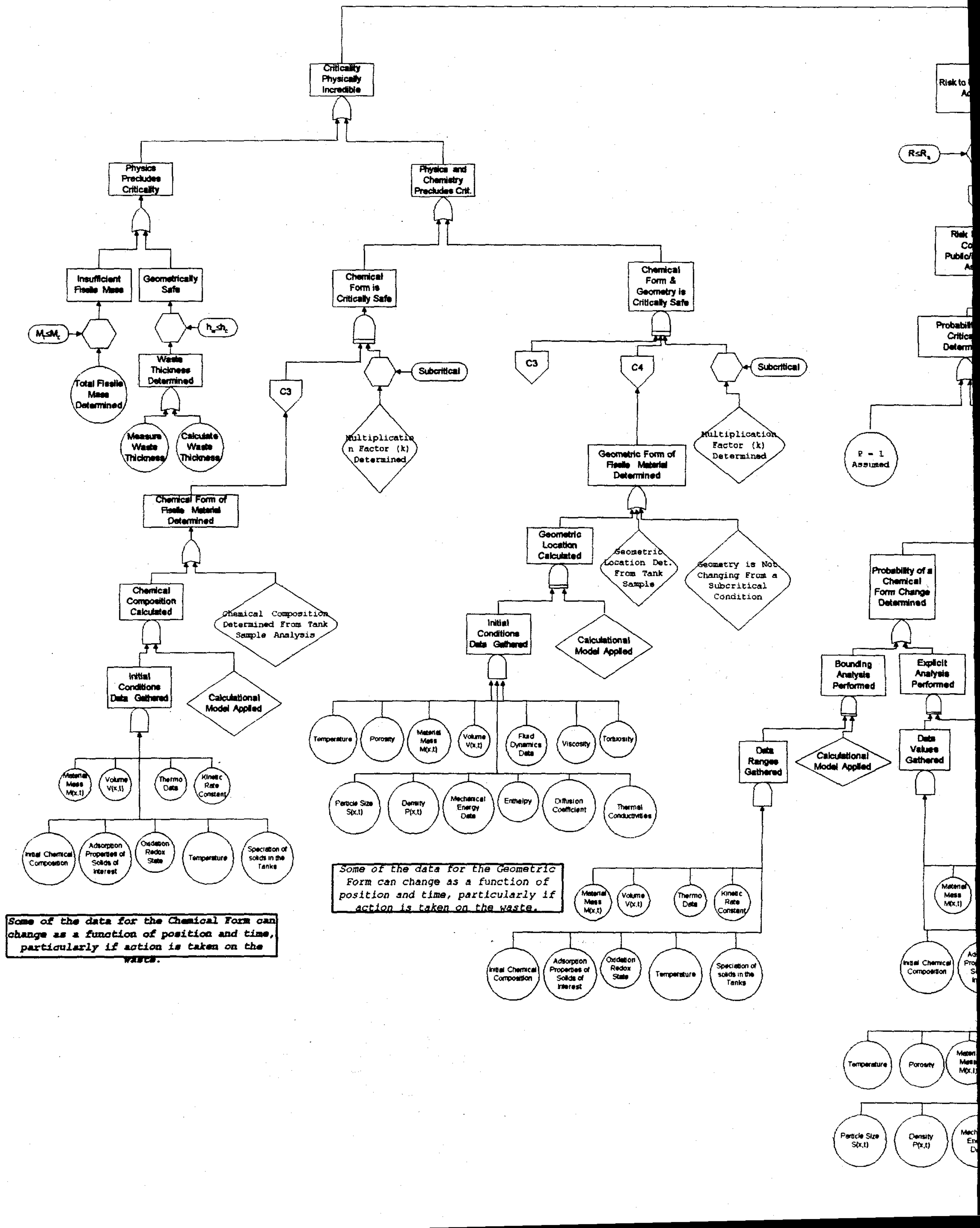
P.G. Doctor, Ph.D., statistics, Iowa State University, 1976. Pam is a scientist at Battelle with 20 years industrial experience. Pam is the author of several journal articles, progress reports, symposium proceeding articles, or invited presentations. Pam has performed human health and ecological risk assessments as part of the DOE "Hanford Site Environmental Remediation Program." She has also been named to the National Academy of Sciences / Natural Research Council's Board on Assessment of NIST programs, been a member of several DOE review committees, and a member of the National Research Council Water Science and Technology Board's committee on Techniques for Assessing Ground Water Vulnerability.

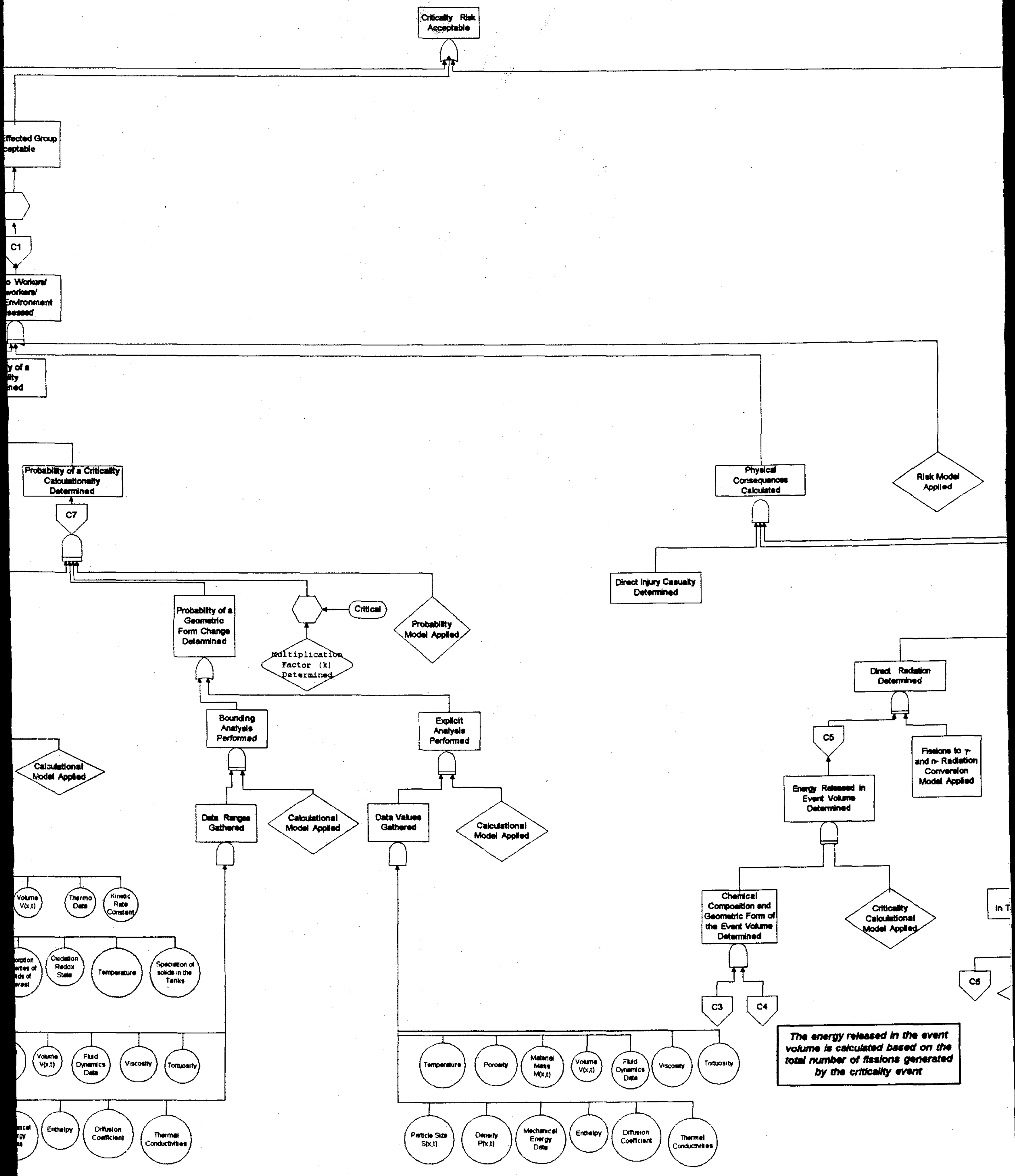
A.R. Felmy is a Staff Scientist at PNNL. He joined Battelle in 1980 designing and building chemical equilibrium models. Andy then went on educational leave of absence and obtained his Ph.D. in chemistry from the Theore

Appendix B

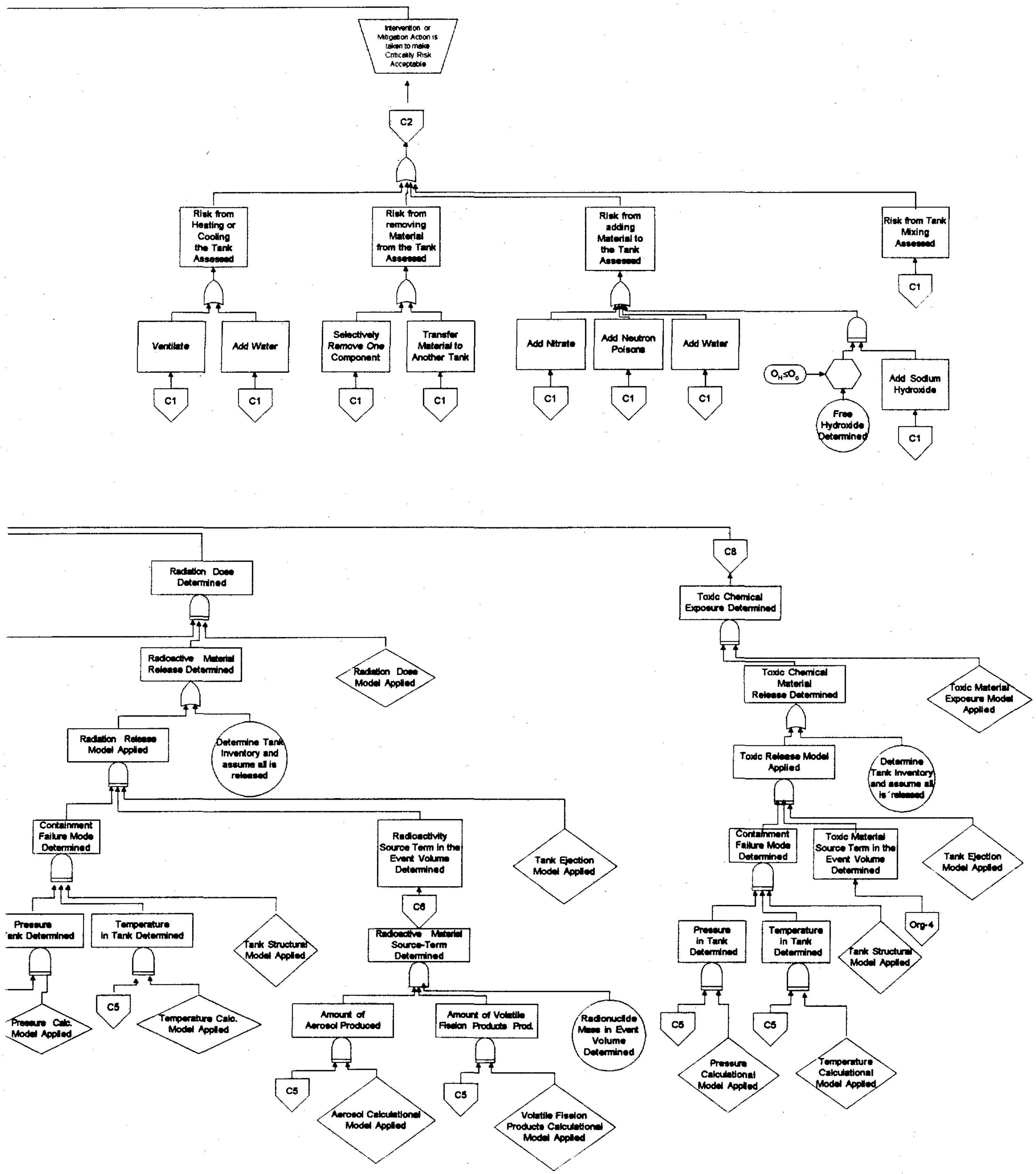
The Criticality Structured Logic Diagram

Structured Logic Diagram for the Criticality Safety Issue





Criticality Risk Acceptable
ffected Group
ceptable
C1
o Workers/
workers/
Environment
sessed
y of a
lity
ned
Probability of a Criticality Calculationally Determined
C7
Probability of a Geometric Form Change Determined
Critical
Multiplication Factor (k) Determined
Probability Model Applied
Bounding Analysis Performed
Explicit Analysis Performed
Calculational Model Applied
Data Ranges Gathered
Calculational Model Applied
Data Values Gathered
Calculational Model Applied
Volume V(x,t)
Thermo Data
Kinetic Rate Constant
Oxidation Redox State
Temperature
Speciation of solids in the Tanks
Volume V(x,t)
Fluid Dynamics Data
Viscosity
Tortuosity
Enthalpy
Diffusion Coefficient
Thermal Conductivities
Temperature
Porosity
Material Mass M(x,t)
Volume V(x,t)
Fluid Dynamics Data
Viscosity
Tortuosity
Particle Size S(x,t)
Density P(x,t)
Mechanical Energy Data
Enthalpy
Diffusion Coefficient
Thermal Conductivities
Physical Consequences Calculated
Risk Model Applied
Direct Injury Casualty Determined
Direct Radiation Determined
C5
Fissions to γ- and n- Radiation Conversion Model Applied
Energy Released in Event Volume Determined
Chemical Composition and Geometric Form of the Event Volume Determined
Criticality Calculational Model Applied
C3
C4
C5
In T
The energy released in the event volume is calculated based on the total number of fissions generated by the criticality event


Intervention or Mitigation Action is taken to make Criticality Risk Acceptable
C2
Risk from Heating or Cooling the Tank Assessed
Risk from removing Material from the Tank Assessed
Risk from adding Material to the Tank Assessed
Risk from Tank Mixing Assessed
C1
Ventilate
Add Water
Selectively Remove One Component
Transfer Material to Another Tank
Add Nitrate
Add Neutron Poisons
Add Water
C1
C1
C1
C1
C1
C1
C1
$O_H \leq O_o$
Free Hydroxide Determined
Add Sodium Hydroxide
C1
Radiation Dose Determined
Radioactive Material Release Determined
Radiation Dose Model Applied
Radiation Release Model Applied
Determine Tank Inventory and assume all is released
Containment Failure Mode Determined
Radioactivity Source Term in the Event Volume Determined
Tank Ejection Model Applied
C6
Pressure ank Determined
Temperature in Tank Determined
Tank Structural Model Applied
Radioactive Material Source-Term Determined
C5
Pressure Calc. Model Applied
Temperature Calc. Model Applied
Amount of Aerosol Produced
Amount of Volatile Fission Products Prod.
Radionuclide Mass in Event Volume Determined
C5
Aerosol Calculational Model Applied
C5
Volatile Fission Products Calculational Model Applied
C8
Toxic Chemical Exposure Determined
Toxic Chemical Material Release Determined
Toxic Material Exposure Model Applied
Toxic Release Model Applied
Determine Tank Inventory and assume all is released
Containment Failure Mode Determined
Toxic Material Source Term in the Event Volume Determined
Tank Ejection Model Applied
Org-4
Pressure in Tank Determined
Temperature in Tank Determined
Tank Structural Model Applied
C5
C5
Pressure Calculational Model Applied
Temperature Calculational Model Applied

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