

**MULTI-STATE SYSTEM IN A FAULT TREE ANALYSIS
OF A NUCLEAR BASED THERMOCHEMICAL
HYDROGEN PLANT**

by

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ABSTRACT

Nuclear-based hydrogen generation is a promising way to supply hydrogen for this large market in the future. This thesis focuses on one of the most promising methods, a thermochemical Cu-Cl cycle, which is currently under development by UOIT, Atomic Energy of Canada Limited (AECL) and the Argonne National Laboratory (ANL).

The safety issues of the Cu-Cl cycle are addressed in this thesis. An investigation of major accident scenarios shows that potential tragedies can be avoided with effective risk analysis and safety management programs. As a powerful and systematic tool, fault tree analysis (FTA) is adapted to the particular needs of the Cu-Cl system. This thesis develops a new method that combines FTA with a reliability analysis tool, multi-state system (MSS), to improve the accuracy of FTA and also improve system reliability.

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NOMENCLATURE

ϕ	System structure function
X	System state vector, $x = (x_1, x_2, \dots, x_k)$
x_i	Current state of component i
d	System demand
ω_i	Number of states for component i
b_i	State space vector for component i
U	Unsatisfied demand, a random variable
ρV_h	Density of $MMCV_h$
R_S	Binary series-parallel system reliability
P_n	Reliability of subsystem n
J	Index indicating the state, $J= 0, 1, \dots, M$
M	Highest state that each component or system may be in
S_i	Subsystem (i is the number of the subsystem)
m_{ij}	State of component N_{ij}
D	Random demand
x_{ij}	Performance of subsystem i in state J
$P_S(J)$	Probability of subsystem i in state J
ϕ_S	Random variable indicating the state of the subsystem
N	Number of working components in a subsystem
N_{ij}	j component in subsystem i
P_i	Performance of subsystem i

λ_c	Component failure rate
λ_b	Base failure rate
f_q	Factor that accounts for the component quality level
f_e	Factor that accounts for the influence of the environment
C	A constant (an empirical relationship between temperature and rate coefficient)
k	Boltzmann's constant
e_a	Activation energy for the process
T_a	Absolute temperature

Chapter 1 Introduction

Hydrogen is expected to be an important alternative energy carrier in the future. The current market for hydrogen is growing rapidly, due to needs in oil refining, ammonia industry, methanol industry, etc. The usage of hydrogen provides a clean fuel to reduce the emissions of carbon dioxide. In Canada, 25% of greenhouse gas emissions can be reduced by usage of hydrogen (NRC, 2004). To meet the growing requirements of hydrogen in the market, an environmental friendly and economical way is needed to generate large-scale capacities of hydrogen. Nuclear-based hydrogen generation is promising since it does not require any fossil fuels, results in lower greenhouse gas emissions and other pollutants, provides large-scale production capability and it is sustainable.

Every project has risks and uncertainties. Hydrogen is a colorless, odorless and highly flammable gas. It has a wide flammability range from 4% to 74% in air. In addition, a relatively small amount of energy is required for ignition by inadvertent mixing of hydrogen with air, such as sparks from electrical equipment, static electricity sparks, open flames or any extremely hot objects. Monitoring the leakage of hydrogen is an important aspect of hydrogen generation processes. Also, there are some other safety issues, such as overheating and overpressure, which need to be considered in industry. Therefore, an effective risk analysis is necessary for the hydrogen generation process.

1.1 Overview of Risk Analysis

Risk analysis involves the development of an overall estimation of risk by gathering and integrating information about scenarios, frequencies, and consequences. It is one major component of the whole risk management process of a particular enterprise. In the process of risk analysis, both qualitative and quantitative techniques can be used, as shown in Figure 1.1 (Krishna *et al.*, 2003). Risk analysis starts from processing the data and information about a specific system, then, a hazard analysis, which is a process used to assess risks. There are two main categories of hazard analysis, namely the probability and consequence. They are also called qualitative and quantitative risk analysis, respectively. Modarres (2006) stated that in qualitative risk analysis, the potential loss is qualitatively estimated by linguistic scales such as low, medium, and high. In this type of analysis, a matrix is formed that characterizes risk in the form of the frequency or likelihood of the loss, versus potential magnitudes of the loss in qualitative scales. The matrix is then used to make policy and risk management decisions, which is the next step in the process of risk analysis. A quantitative risk analysis attempts to estimate the risk in the form of the probability or frequency of a loss. It evaluates such probabilities to make decisions and communicate the results. The next step of the process is risk evaluation. The results obtained from the risk analysis will be evaluated based on the importance or criticality of the risks, which will affect the system operation, and quality of products, etc. Then, if the results are acceptable, the process of risk analysis will stop immediately. Otherwise, the system needs to be improved and all steps repeated again.

Why perform a risk analysis? The risk of incidents (such as explosion, fire, and chemical release) is increasing with the rapid progress of industrialization. It became recognized that there was a worldwide trend for losses due to accidents, to rise more rapidly than gross national product (Lees, 1996). The results of major industrial accidents can be

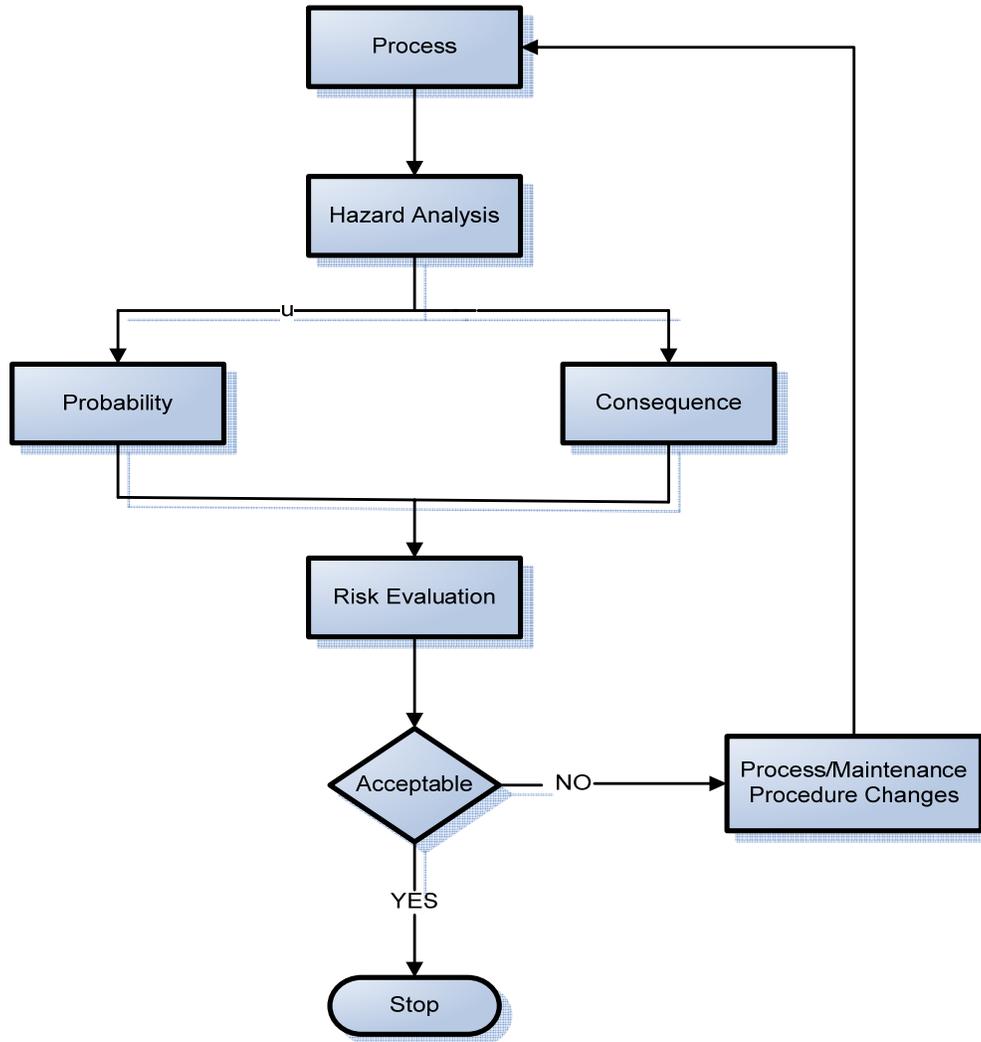


Figure 1.1 The process of risk analysis (Krishna et al., 2003)

devastating, such as a massive explosion in Pasadena, Texas on Oct. 23, 1989, which resulted in 23 fatalities, 314 injuries, and capital loss of over \$715 million (Lees, 1996); Bhopal, India accident, which killed more than 2,000 civilians and injured 20,000 more (Crowl & Louvar, 2002); the Flixborough, England accident, which cost the lives of 28 people, the whole plant and many injuries (Crowl & Louvar, 2002). These are extreme cases of major accidents in the process industry, but minor incidents are very common in the process industry, occurring day to day, resulting in many occupational injuries, illnesses, and costing industry billions of dollars every year. Almost all of the major accidents could have been avoided through an effective risk analysis and safety management program.

1.2 Objective of Thesis

As a risk analysis tool, FMEA can also help build fault trees. However, it is particularly useful to identify single failure modes that lead to an incident directly, while it is not useful to identify combinations of equipment failure, as it is not as flexible as FTA for a design system. Therefore, this thesis is focusing on the improvement of FTA for the Cu-Cl hydrogen generation cycle. FMEA will not be discussed here. The objective of this thesis is introducing multi-state system (MSS) into FTA, in order to improve the evaluation results in FTA. On the other hand, by using MSS, the system reliability is improved simultaneously.

1.3 Organization of Thesis

In chapter 2 of this thesis, a literature review is presented. The technology of FTA is further introduced, and the previous research in MSS is reviewed. In chapter 3, the popular techniques of nuclear-based hydrogen generation are discussed, and detailed explanations about the Cu-Cl cycle are presented. In chapter 4, some main fault trees for the Cu-Cl cycle will be constructed. Then, the reliability model will be built based on the FTs to make the MSS. Chapter 5 gives an example to show how MSS works for FTA and improves system reliability. Finally, the conclusions of the current work will be summarized and the future work will be recommended in chapter 6.

Chapter 2 Literature Review

2.1 Overview

A briefly review of risk analysis is presented at the beginning. Risk analysis methods and some important terminologies are introduced. To achieve a higher reliability is one of the most important tasks that engineers desire when designing a system. Because of the limitation of information and data of a system, an efficient and useful tool, FTA, is used widely. In the Cu-Cl hydrogen generation system, there are chemical reactors, heat exchangers, pumps, pipes, etc, and other necessary equipment, such as mechanical and electrical components to be installed in the plant. These different sources of failures will be part of the future reliability analysis of this hydrogen generation system.

As mentioned in the previous chapter, reliability and failure are two complementary concepts. A failure analysis should be performed first, in order to improve the reliability of the hydrogen generation system. Then, more information about FT and FTA will be presented in the failure analysis. To improve the system reliability, MSS will be introduced later in this chapter. Some discussion will be given about the recent research advances in MSS.

2.2 Risk Analysis Methods

A principal theme of risk analysis is to ensure that we know the hazards before the

system starts operating. The deterministic methods focus on consequence assessment (such as worst-case scenario analysis), while the probabilistic approaches consider both consequence and frequency. The techniques that have been used for risk analysis which are Hazard and Operability Study (HAZOP), Relative Ranking, Checklist Analysis, Preliminary Hazard Analysis, “What-if” Analysis, Failure Modes and Effects Analysis (FMEA), Fault Tree Analysis (FTA), Event Tree Analysis (ETA), Human Reliability Analysis (HRA), and Cause-Consequence Analysis (CCA) (CCPS, 1992; Lees, 1996). Some of the techniques require special expertise such as FTA, ETA, CCA, and HRA. Brief overviews of the most popular methods, FMEA and FTA, will be presented with their strengths and limitations.

2.2.1 Failure Mode and Effect Analysis

Failure mode and effect analysis (FMEA) is a systematic procedure. Each equipment failure mode is examined to identify its effects on a system and classify them according to severity and criticality. As an inductive method, FMEA is oriented toward equipment rather than process parameters. All of the failure modes for each item of equipment are tabulated with their related actions, which are listed safeguards, and effects. An FMEA is especially useful to identify single failure mode, which can lead to an incident directly, but it is not powerful enough to identify combinations of equipment failures and human errors as risk contributors.

There are three independent FMEA documents, which are the Service FMEA, Design FMEA, and Manufacturing FMEA (Bloch and Geitner 1990). The service FMEA evaluates service tools and manuals to ensure that no improper operation, which leads to malfunctions, occurs. The design FMEA emphasizes the failures that could occur with a product and the effects on the end users. The manufacturing FMEA evaluates and lists the variables that could affect the quality of a certain process

Table 2.1 Basic failure modes of mechanical components

Application	Part/Element level	Assemble level
Force/Stress/ Impact	Deformation Fracture Yielding Insulation rupture	Binding Seizing Misalignment Displacement Loosening
Reactive environment	Corrosion Rusting Staining Cold embrittlement Corrosion fatigue Swelling Softening	Fretting Fit corrosion

Thermal	Creep Cold embrittlement Insulation breakthrough Overheating	Thermal growth/contraction Thermal misalignment
Time related factors	Fatigue Erosion Wear Degradation	Cycle life attainment Relative wear Aging Degradation Fouling/contamination Plugging

FMEA has various advantages. The FMEA procedure can be implemented systematically by using a standardized FMEA analytical form (Rao, 1992). FMEA presents a consistent document for investigating all potential faults of a system, evaluating the risk associated with the faults, and preventing the occurrence of high risk (Bloch and Geitner, 1990). FMEA enables the designers and people who are involved in operations, maintenance, and repair, to gain a better knowledge of the system (Bloch and Geitner, 1990).

The disadvantages of FMEA are listed as follows. Uncertainty exists in FMEA, which is used to diagnose the failure causes. Since one failure mode may be caused by more than one possible cause, and it is hard to decide which cause is more responsible for the failure, it is difficult to use the appropriate measures in FMEA to improve a certain component or

delete a certain failure cause. In addition, it is difficult to achieve a precise FMEA result. Precision of this analytical method greatly depends on the skill and experience of the analysts. Some items, such as occurrence and severity in forms are decided subjectively. Thus, the forms developed by different analysts may not achieve the same conclusion. They may emphasize different components or subsystems, and use different measures to detect the potential failures and improve the reliability of the system. A relatively accurate FMEA requires very detailed analysis, so that all of the components can be traced. It is typically not possible to study all of the components in the system, since it is an extremely time consuming process.

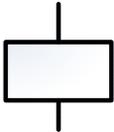
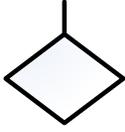
2.2.2 Fault Tree Analysis

Fault tree analysis (FTA) was first developed in 1961 at Bell Telephone Laboratories for missile launch control reliability during the Polaris project. It has been extensively used in reliability studies in the nuclear and aerospace industry.

FTA is a deductive method that proceeds from general to the specific. Using this method, an undesired event, a failure, called a top event, is put forward, and all the possible events and faults, which might lead to the top event, are determined. The failure paths are represented graphically by the use of a fault tree drawing. A fault tree is constructed from events and logical operators. Ideally, this top-down analysis can reach the level where the reliability data of the events are available, and therefore the quantitative analysis can be performed.

Fault tree gates and symbols. Logic gates and some common symbols of fault tree analysis are listed in Table 2.2 (Roland and Moriarty, 1983).

Table 2.2 Logic gates and selected symbols of a fault tree

	OR Gate denotes the situation in which an output event occurs if any one or more of the input events occur.
	AND Gate denotes the situation in which an output event occurs only when all the input events occur.
	RECTANGLE denotes an event that results from the combination of fault events through the logic gate.
	DIAMOND denotes a fault event led by an undeveloped cause
	CIRCLE denotes a basic fault event.

Major advantages of FTA are listed as follows.

- FTA systematically subdivides a big system into smaller manageable subsystems that can be individually assessed and quantified. FTA provides analysts with an insight into the system behavior by indicating the interactions of components and subsystems (Roland & Moriarty, 1983).
- FTA can give qualitative or quantitative outputs, which are the most advantageous for system analysis, compared with other reliability methods (Roland & Moriarty, 1983).
- All events present in fault trees are fault or accident related events (Roland & Moriarty, 1983).
- FT is presented in graphics consisting of symbols and gates. FTA provides an illustrative description of system functions and the relationships between events that lead to the same fault through an OR or AND gate.

Major disadvantages of FTA are listed below.

- There is no effective method to avoid overlooking events, or neglecting operating, or environmental conditions in FTA (Bloch & Geitner, 1990). A relatively accurate FTA requires very detailed analysis so that all components need to be traced. It will be an extremely time consuming process for a complicated system.
- Lack of reliable failure data weakens the power of FTA.

- In fault trees, it is usually assumed that the events through an OR gate are independent. However, it is difficult to determine if a condition of independence truly exists in a complex system (Roland & Moriarty, 1983).

2.3 Basic Terminologies in Risk Analysis

This section lists some important terminologies in risk analysis, which will be discussed later in the thesis.

Probability

Probability defines the quantitative likelihood of the occurrence of an event or events. A probability could be a value between zero and one. The lower limit “zero” stands for no chance of an event occurring, while the upper limit “one” stands for inevitable or unavoidable occurrence of an event (Kendrick, 2004). The situation when both events X and Y take place is denoted as $X \cap Y$ (intersection of the two events). The situation when either X or Y or both may occur is denoted as $X \cup Y$ (union of the two events). The probability of occurrence of events X and Y are denoted as $P\{X\}$ and $P\{Y\}$ respectively. The probability of occurrence of a union or intersection of events X and Y can be calculated as shown below (Roland and Moriarty, 1983).

$$P\{X \cap Y\} = P\{X\}P\{Y\} \quad (2.1)$$

$$P\{X \cup Y\} = P\{X\} + P\{Y\} - P\{X \cap Y\} \quad (2.2)$$

Equations (2.1) and (2.2) deal with the combinations of two related events. If m events X_m are all independent, then the relationships expressed in equations (2.1) and (2.2) can be expanded to m events as follows (Lewis 1996),

$$P\{X_1 \cup X_2, \dots, \cup X_m\} = 1 - [1 - P\{X_1\}][1 - P\{X_2\}] \dots [1 - P\{X_m\}] \quad (2.3)$$

$$P\{X_1 \cap X_2, \dots, \cap X_m\} = P\{X_1\}P\{X_2\} \dots P\{X_m\} \quad (2.4)$$

Failure rate

The failure rate is the number of faults of an event or failures of a component per unit measure of life (Roland and Moriarty, 1983).

Network modeling

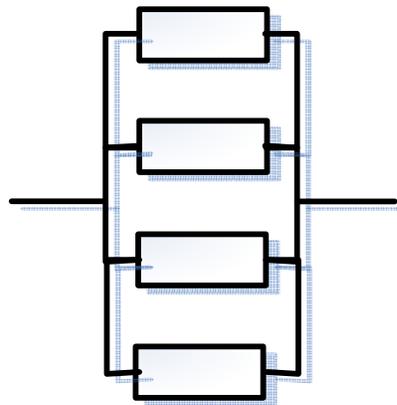
Network modeling is one of the system analysis methods. Since the system units are usually connected to each other in series, parallel, meshed, or a combination of these in a given system, networks are easily represented by block diagrams, in which blocks stand for the system units. Network modeling can be used for reliability and failure analysis of a system. Correspondingly, there are two basic types of systems: series and parallel.

In series systems, the units are connected in series, which is shown in Fig. 2.1(a), and only when all of the units work, the system works. Fig. 2.1 (b) shows a simple parallel

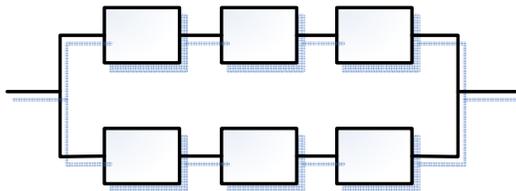
system, where the units are assembled in parallel, and when all of the units fail, the system fails. Combinations of series and parallel systems make more complicated networks (Elsayed, 1996), such as parallel-series, series-parallel, and mixed-parallel systems. Figs. 2.1 (c), (d), and (e) show the block diagrams of these different kinds of systems. Because of the limitations of simple series and parallel systems that were mentioned above, in order to achieve higher reliability, combinations of these two systems are used widely, and in practical fields, the reliability models are more complex than any other figures shown here. However, they are able to be broken down to these types of common systems for analysis.



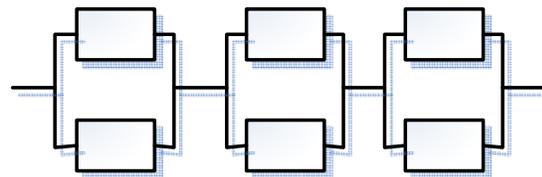
(a) Simple series



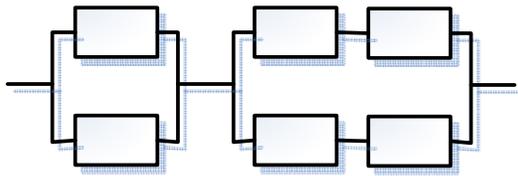
(b) Simple parallel



(c) Parallel-Series



(d) Series-Parallel



(e) Mixed-Parallel

Figure 2.1 Different types of networks (Elsayed, 1996)

Cut Sets

A cut set was defined by Billinton and Allan (1992) as a set of system units that causes failure of the system when it fails. For a series system consisting of units A and B, there are three cut sets, A, B, and AB, while for a parallel system consisting of two units of A and B, there is only one cut set, AB. Also, this terminology is widely used in FTA. Minimal Cut Sets provides one of the methods to simplify FTA processes.

2.4 Method of fault tree analysis

As one of risk analysis methods, FTA has been used widely in reliability evaluation of standby, protection and complete mission oriented systems, particularly for safety system. Billinton and Allan (1992) stated that this technique was first developed and used in qualitatively assessing the failure processes of a complex system. It identifies the consequences of failure on system behavior. Also, it can be used to perform quantitative assessments. As a qualitative evaluation method, it can assist the designer, planner or operator in deciding how a system or subsystem may fail as a result of individual

component behavior, and what remedies may be used to overcome the causes of failure. In quantitative evaluation, the reliability assessment of the complete system can be calculated by inserting the reliability data for the individual components into the tree at the lowest hierarchical level, and then combining it together using the logic of the tree (Billinton and Allan 1992). This thesis focuses on the quantitative FTA.

2.4.1 Fault Classification

Distinguishing different types of faults is an important process in determining the logical structure of a fault tree, as well as parts or components that are more likely to cause a system failure. The following classifications have been widely used in FTA (Lewis, 1996).

- a) Primary, secondary, and command faults. A primary fault happens in an environment or under a loading where the component is qualified. A secondary fault occurs in an environment or under a loading for the component not qualified. For instance, if a transmission shaft fails under a loading which is higher than the design loading capacity, the shaft has a secondary fault. Secondary faults occur randomly, but usually, they have constant failure rates. A command fault leads to a primary or secondary fault when a component operates correctly under a command, but this command is given at a wrong time or situation. For example, if an erroneous command causes a valve to open and release reactants before a process is done, the valve will have a command fault.

b) Passive and active faults. Components of a system operate in either a static or dynamic manner, so components can be classified as passive or active components, accordingly. Components such as bolts, pipes, and reactors are passive components, while valves, switches and motors are active components. A passive component may be considered as a transmitting mechanism between active components. Faults occurring on passive components are considered as passive faults, while those occurring on active components are defined as active faults. Usually, the active components have much higher failure rates, typically two or three orders of magnitude higher than the passive components.

2.4.2 Fault Tree Construction

The applications of the fault tree technique have been increasing gradually since the 1960s. However, it remains more an art rather than a science in fault tree construction processes, since there is no standard taxonomy for fault tree construction. In the practical field, there are some general guidelines that have been extracted to assist engineers to produce high-quality fault tree methods. These rules have been examined and listed in the fault tree handbook as ground rules and procedural rules (NUREG-0492, 1998).

Ground Rule 1: Write the event statements precisely about what is the fault and when it occurs. Do not abbreviate words that might lead to ambiguous meaning.

Ground Rule 2: If a specific fault consists of a component failure, classify the event as a “state-of-component fault”. Otherwise, classify it as a “state-of-system fault”.

Procedural Rule 1 (No Miracles Rule): If the normal functioning of a component propagates a fault sequence, then it is assumed that the component functions normally. In other words, it is treated as a certain event. If the normal functioning of a component prevents the propagation of a fault sequence, AND logic is used to consider its failures.

Procedural Rule 2 (Complete-the-Gate Rule): All inputs to a particular intermediate gate should be completely defined before further analysis of any one of them is undertaken. Each child must be an immediate and direct cause of its parent. Each child must be either sufficient, in the case of an OR gate, and necessary in case of an AND gate.

Procedural Rule 3 (No Gate-to-Gate Rule): All gate inputs should be properly defined, and no gate should directly feed into another gate.

2.4.3 Evaluation of a Fault Tree

Top-down and bottom-up evaluations are the two most straightforward methods for qualitatively evaluating the fault trees (Lewis, 1996). Top-down evaluation starts with the top event and continues downward through each level of the tree. The bottom-up evaluation method, unlike the top-down method, starts from the basic events and

continues upward to the top event. The logical relationship between the input events and the top event can be expressed by using the OR or AND logic operators. For both methods, the reduction results are exactly the same. Logical reduction is usually used to simplify a fault tree, in order to make the evaluations of the fault tree performed more easily and efficiently. Logical reduction is particularly useful for complex fault trees where there are many intermediate levels, and many basic events.

In some examples, a quantitative measure of the occurrence of the top event is required, which is based on a qualitative evaluation. Probability is used to calculate the occurrence of the top event when the probabilities of the events in the lowest level are known. In fault tree quantitative analysis, various reliability measures can be used. Among these measures, the failure rate is used commonly. The failure rate is the number of faults of an event or failures of a component, per unit measure of life (Roland and Moriarty, 1983). The measure of life could be time, cycles, miles and so on. The data of failure rates of the components may be obtained from a related reliability database.

2.4.4 Quantitative Evaluation of a Fault Tree

In order to quantify the probability of the top event of the FT, the probability of every basic event (BE) in the FT must be provided. Then, these BE probabilities are propagated upward to the top event using Boolean algebra for the FT. Alternatively, the minimal cut sets of the FT can be generated and used to quantify the top event.

Data requirements

Quantitative data needs to be used for input to the basic events, to carry out quantifications of an FT. In the simplest form, the input data consists of the probability of basic events in the fault tree. Thus, the main data that is required based on the type of basic events being quantified. Bedford and Cooke (2001) gave the types of data that are generally required: a) Component failure rate data; b) Human error data; c) Common cause failure data; and d) Phenomenological data.

Importance Measures for a FT

One of the most important outputs of an FTA is the set of importance measures (IM) that are calculated for the top event. These top importance measures give the significance for all events in the FT, in terms of their contributions to the top event probability.

Vesely *et al.* (1983) developed an importance measure, which is called the Fussell-Vesely (F-V) Importance. It is also called the Top Contribution Importance sometimes. Both the absolute and relative F-V importance can be determined for each event modeled in the FT, not only for the basic events, but for every higher level event and contributor. This provides a numerical significance of all the FT elements and allows them to be prioritized. Some other importance measures that can be calculated for each event in the FT are: Risk Reduction Worth (RRW), Risk Achievement Worth (RAW), and the Birnbaum's Importance Measure (BM) (Bedford and Cooke, 2001).

Efficient Quantitative FT Evaluation Method

Based on Vesely's theory (1970), Amari *et al.* (2002, 2003, 2004) demonstrated how to use the Vesely Failure Rate (VFR) as an approximation in the FT evaluation process. By comparing the VFR method with some other approximations and simulation results, it is much more efficient and close to the simulation results. In addition, it can handle a large system. Thus, this method is recommended to be used in the FT evaluation of the Cu-Cl hydrogen generation process.

2.4.5 Mini Fault Tree Based Method

Fussell (1973) developed a formal fault tree synthesis methodology for electrical systems. In his method, the system-independent component failure transfer functions were developed with a system schematic diagram and associated system boundary conditions to construct fault trees. The methodology was extended to fault tree construction beyond the area of electrical systems later in his paper. Taylor (1982) modified Fussell's method in order to manage loops in the system.

A series of papers have been published based on component mini-fault tree models (Kelly & Lees 1985; Hunt *et al.*, 1993). A large system is decomposed into individual items of equipment. The mini fault trees are developed by propagation equations, event statements, and decision tables. Due to the higher requirement for storage and

computational complexity of this method, there has been no further advance in this method since 1993. However, it provided a useful tool to split a large FT into small parts, which refers to different equipment, to make the FTA easier.

2.4.6 Dynamic FTA

The Dynamic Fault Tree (DFT) methodology was developed to provide a means for combining FTA with Markov analysis. The difference between traditional FT and DFT is that DFT can capture the dynamic behavior of the system failure mechanisms associated with a sequence dependent event, spares, and priorities of failure events (Dugan *et al.*, 1992; Gulati and Dugan, 1997; Manian *et al.*, 1998). In spite of the advantages of DFT, the disadvantage is a large and cumbersome method of using Markov models in DFT. Also, the generation of a Markov model for many systems can be tedious and error-prone. In addition, neither the mini fault tree nor DFT can take care of the events with different states, which is a popular concept in reliability analysis area.

2.5 Multi-state System

Traditionally, system reliability has been analyzed as a binary perspective, assuming the system and its components can exit in two different states: either completely functioning or failed. However, many systems that provide basic services, such as telecommunications, gas and oil production, transportation and electric power distribution, operate at various levels of performance, which is usually more than two

states (Lisnianski and Levitin, 2003). Therefore, a multi-state system is introduced into the model to analyze these systems accordingly (Barlow and Heidtmann, 1984; El-Newehi *et al.*, 1978; Kuo and Zuo, 2003; Natvig, 1982).

In a multi-state system model, both the system and its components may experience more than two levels of performance. In other words, a system and its components may lie in $M+1$ possible states, which are 0, 1, 2, ..., M . Here 0 indicates the completely failed state, M indicates the perfectly working state, and others are degraded states.

Since there are more states, a multi-state system is able to generate more accurate evaluation results than a binary state system (Lin, 2001; 2002). Generally, MSS with multi-state components (MSC) has to fulfill a known demand, based on the different component performance states (Ramirez-Marquez and Coit, 2003; Ramirez-Marquez *et al.*, 2005).

2.5.1 Multi-State K-out-of-N System

Huang *et al.* (2002) provided a review of the multi-state k-out-of-n models. In a multi-state k-out-of-n system, it is the same as a binary k-out-of-n system, where at least the number of k components in the system must work to make the system work, whereas in a multi-state k-out-of-n system, the components must operate at a required state or above. The commonly used series and parallel system can be treated as special cases of the simple multi-state k-out-of-n system. El-Newehi and Proschan (1978) proposed that the

state of a multi-state series system is equal to the state of the worst component in the system, and the state of a multi-state parallel system is equal to the state of the best component in the system.

2.5.2 Importance Measures in MSS

The IMs introduced in FTA, which are RAW, RRW, FV, and Birnbaum, are also widely used in binary state reliability systems (Vasseur and Llory, 1999). Some of them are extended to multi-state cases, such as RAW, FV, and Birnbaum. In order to define IM in MSS, the IMs in MSS are divided into two types (Ramirez-Marques and Coit, 2005):

- 1) Quantify the impact of a component as a whole on the system reliability (Aven and Ostebo, 1986; Meng, 1993; Wu and Chan, 2003);
- 2) Quantify how a particular component state or set of states affect system reliability (Barlow and Wu, 1978; Griffith, 1980; Zio and Podofillini, 2003; Levitin *et al.*, 2003).

There are three IMs for MSC, which were introduced by Ramirez-Marques and Coit (2005). The first IM is called an unsatisfied demand index (UDI), which identifies the impact of a component or component state in terms of unsatisfied demand for the MSS. The second IM is a multi-state failure frequency index (MFFI) that quantifies what percentage of system failure can be attributed to a specific component or component state. The last IM, a multi-state redundancy importance (MRI), quantifies the increase in

reliability when a redundant component is added to the system. MFFI belongs to Type 1 IM, UDI is Type 2 IMs, and MRI helps to identify where to allocate component redundancy to improve system reliability.

2.6 Example of Using Importance Measures

Levitin *et al.* (2003) gave a model of a multi-state system, which is shown in Fig. 2.2. This model is a combination of series and parallel systems, and it is similar to the one for a sensing system that will be discussed in upcoming sections. Note that elements 2, 3, 5, and 6 are identical, but the pairs of elements 2, 3 and 5, 6 have different influences on the entire system performance, since they are connected in series with different elements, 1 and 4, respectively. Therefore, while elements 2 and 3 have the same importance as well as elements 5 and 6, the importance of elements 2 or 3 differs from the importance of elements 5 or 6. This will be further demonstrated in the following section of results.

Based on this MSS model, Ramirez-Marquez *et al.* (2006) presented IM calculation results that are listed in the following tables.

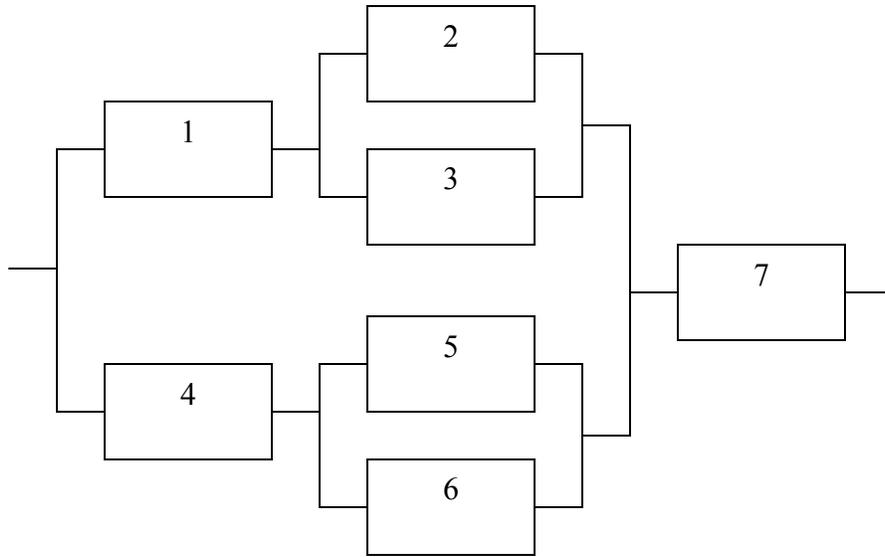


Figure 2.2 Example of Multi-state system (Levitin *et al.*, 2003)

Table 2.3 Component states and associated probabilities (Ramirez-Marquez, *et al.*, 2006)

I	States					State occupancy probabilities				
	m_{i1}	m_{i2}	m_{i3}	m_{i4}	m_{i5}	p_{i1}	p_{i2}	p_{i3}	p_{i4}	p_{i5}
1	0	1	2	3	4	0.1	0.05	0.15	0.35	0.35
2	0	1	2			0.1	0.05	0.85		
3	0	1	2			0.1	0.05	0.85		
4	0	1	2	3		0.2	0.1	0.45	0.25	
5	0	1	2			0.1	0.05	0.85		
6	0	1	2			0.1	0.05	0.85		
7	0	1	2	3	4	0.15	0.15	0.05	0.45	0.2

Table 2.4 MMCV at level 3 (Ramirez-Marquez, *et al.*, 2006)

MMCV	x ₁	MMCV	x ₁												
1	4	2	2	3	2	2	2	14	0	2	2	3	0	2	4
2	4	2	0	3	0	0	4	15	1	2	2	3	1	0	4
3	4	0	2	3	0	0	4	16	1	2	2	3	0	1	4
4	4	0	0	3	2	0	4	17	0	2	2	3	1	1	4
5	4	0	0	3	0	2	4	18	4	2	0	0	2	2	4
6	4	1	1	3	0	0	4	19	4	0	2	0	2	2	4
7	4	1	0	3	1	0	4	20	4	0	0	2	2	2	4
8	4	1	0	3	0	1	4	21	4	1	0	1	2	2	4
9	4	0	1	3	1	0	4	22	4	0	1	1	2	2	4
10	4	0	1	3	0	1	4	23	4	1	1	0	2	2	4
11	4	0	0	3	1	1	4	24	2	2	2	0	2	2	4
12	2	2	2	3	0	0	4	25	1	2	2	1	2	2	4
13	0	2	2	3	2	0	4	26	0	2	2	2	2	2	4

Table 2.5 Type 1 IM and ranking (Ramirez-Marquez, *et al.*, 2006)

Rank	i	MFFI	I	UDI	i	MRI
1	7	0.55902	7	1.31011	7	0.20586
2	4	0.37294	1	1.09668	1	0.06361
3	1	0.33501	2	1.00036	4	0.06264
4	2	0.07243	3	1.00002	3	0.01090
5	3	0.07243	4	0.99392	2	0.01084
6	5	0.03849	6	0.97039	6	0.00331
7	6	0.03849	5	0.97039	5	0.00324

Table 2.3 gives the component states and associated probabilities. In table 2.4, a multi-state minimal cut vector (MMCV) is listed and associated with a network model. To calculate MMCV, Lin (2002) and Yeh (2004) have made developed reduced implicit enumeration methods for finding MMCV. Both methods depend on a priori knowledge of the system minimal cut sets, and can only be applied to systems where the components have consecutive states (i.e. $m_{i,j+1} = m_{ij} + 1$). Then, Ramirez-Marquez et al. (2005) developed an information sharing approach that reduces the number of implicit enumerations necessary to obtain the MMCV. The rationale of this approach is that since all MMCV can be obtained from the set containing all minimal cuts (Lin, 2002; Yeh, 2004), a select number of MMCV called offspring cuts can inherit information from a select number of MMCV called parent cuts, therefore reducing the number of implicit enumerations. Table 2.5 presents the IMs to show which quantifies that component 7 is

the weakest in the MSS network. Ramirez-Marquez et al. (2006) presented mathematical expressions for calculating these IMs, as listed below.

Firstly, as a Type 1 IM, MFFI can be used to approximate the system failure frequency, due to failure of a particular component.

Type 1:

$$MFFI = \sum_h \rho V_h \forall h \text{ such that } x_k < \max \{b_{kj}\} \text{ in } MMCV_h \quad (2.5)$$

Secondly, UDI can be considered in the category of risk averse measures, from a general perspective. The mathematical expression for this measure is shown as follows:

$$UDI_k = \frac{1}{\omega_k} \sum_j UDI_{kj} \quad (2.6)$$

$$UDI_{kj} = E[U | x_k = b_{kj}] \quad (2.7)$$

MRI can be linked to an estimate of increased profit caused by adding redundancy to a component or improving a particular state. The expression is expressed as follows:

$$MRI_k = P(\varphi(X, x_k^+) \geq d) - P(\varphi(X) \geq d) \quad (2.8)$$

Here $P(\varphi(X, x_k^+) \geq d)$ is defined as the probability associated with an event, where the capacity of the system is greater than or equal to demand, d , when a copy of component k has been added to the system design.

Even though the results can represent the importance of each component in a system adequately, the calculation is time-consuming, so it is not recommended for the beginning of a reliability analysis, such as Cu-Cl cycle, since if any variable changes in the system, all calculations should be repeated again. It is preferable to apply it when redundancies of critical components are added.

2.7 Summary

A brief review of the past research work in FTA and MSS were provided in this chapter. Because of the limitations of the two FTA methods, neither will be used in this thesis. Some background knowledge of MSS was briefly reviewed, and an example was given to show how the IMs can be used in MSS reliability analysis.

Chapter 3 Nuclear-Based Thermochemical Hydrogen Plant

Nuclear-based hydrogen generation has an important potential advantage over other methods, as it does not rely on fossil fuels, and thus results in lower greenhouse-gas emissions and other pollutants. It provides a large-scale method of hydrogen production, and is environmentally sustainable.

3.1 Overview of Hydrogen Generation Techniques

Currently, there are three main ways that nuclear energy can be used in hydrogen production (Yildiz et al., 2005):

- a) By using the electricity from a nuclear plant for conventional liquid water electrolysis;
- b) By using both the high-temperature heat and electricity from the nuclear plant for steam electrolysis or a hybrid process;
- c) By using the heat from the nuclear plant for pure thermochemical processes.

Table 3.1 presents an overview of nuclear hydrogen production technologies (NRC & NAE, 2004). The leading methods among all these candidate technologies are high-temperature steam electrolysis and high-temperature thermochemical water-splitting cycles, since a higher temperature gives higher efficiency in the table.

Table 3.1 Overview of nuclear hydrogen production processes (NRC&NAE, 2004)

Feature	Electrochemical		Thermochemical	
	Conventional electrolysis	High-Temperature Electrolysis	Thermochemical Water Splitting	Steam-Methane Reforming
Required temperature (°C)	70-80	>600	530-850	>700
Efficiency of chemical process (%)	75-80	85-90	>45	70-80
Efficiency coupled with current generation reactors (%)	27	30	Not feasible	Not feasible
Efficiency coupled with future high-temperature reactors (%)	<40	40-60	40-60	>70
Advantages	Proven technology	<ol style="list-style-type: none"> 1. Reactors operating at intermediate temperatures 2. Eliminates CO₂ emissions 	<ol style="list-style-type: none"> 1. Potential for high efficiency 2. Eliminates CO₂ emissions 	<ol style="list-style-type: none"> 1. Proven technology 2. Reduces CO₂ emissions
Disadvantages	Low energy efficiency in the near term	Requires development of durable, large scale HTE units	<ol style="list-style-type: none"> 1. Aggressive chemistry 2. Requires very high temperature reactors 	<ol style="list-style-type: none"> 1. CO₂ emissions 2. Dependent on methane prices

In addition, high-temperature steam electrolysis and high-temperature thermochemical water-splitting cycles have margins for improvement in their efficiency and cost. Water electrolysis coupled to a Light Water Reactor (LWR) is the least energy efficient, but it is well commercialized, and it is the only currently available technology for producing hydrogen without greenhouse gas emissions.

Thermochemical processes for hydrogen production involve thermally assisted chemical reactions that release hydrogen from hydrocarbons or water. The most widespread thermochemical process for hydrogen production is steam methane reforming (SMR), which is shown in table 3.1. A disadvantage of this technology is the emission of carbon dioxide, although it is the most economic method today. Yildiz et al. (2005) stated that alternative thermochemical processes are those that do not use hydrocarbon feedstock, but split water into hydrogen and oxygen through a series of thermally driven chemical reactions. The purpose is to produce hydrogen at lower temperatures. Currently, two of the popular thermochemical cycles are sulfur-iodine (SI) and copper-chlorine (Cu-Cl) cycles, which require a temperature higher than 500°C. In order to provide high temperature steam for hydrogen generation reactions, the Super-Critical Water Reactor (SCWR), Canada's Generation IV nuclear reactor, will be widely deployed around 2025 (Spinks *et al.*, 2002). As the principal Canadian developer, Atomic Energy of Canada Limited (AECL) is participating in an International Nuclear Energy Research Initiative (I-NERI) program. AECL is currently exploring development of this nuclear reactor, as a Generation IV system based on its successful CANDU[®] reactor system.

S-I cycle development is being investigated by the U.S., France, and Japan due to its high efficiency (Wu and Kaoru, 2005). It consists of the following three chemical reactions to yield the dissociation of water (Brown et al., 2003):



The net process requires water and high temperature heat input. Then, hydrogen and oxygen are generated. All reagents are recycled, and there are no external effluents. The highest temperature is required at the second step, which is over 800°C, so a very high temperature reactor (VHTR) must be used. Yildiz et al. (2005) described the major challenges of S-I cycles as follows: a) material durability at high temperature and high acidity environment; b) HI inventory recovery in the system; c) separations between reactants and products in solutions.

3.2 Introduction to the Copper-Chlorine (Cu-Cl) Cycle

This thesis focuses on a copper-chlorine (Cu-Cl) cycle, which has been identified by AECL and belongs to the category of thermochemical processes. UOIT and AECL have been collaborating with the Argonne National Laboratory (ANL) in studies of the Cu-Cl cycle. ANL developed enabling technologies for the Cu-Cl thermochemical cycle and created a flowsheet of the hydrogen plant by using Aspen Plus simulations (Lewis *et al.*,

2005). The highest required temperature is ~500°C, and SCWR is able to provide this high temperature steam for the reactions. Compare with the S-I cycle, the corrosion issues of the Cu-Cl cycle are more tractable at 500 °C. The energy efficiency of the process is projected to be about 43%.

In the Cu-Cl cycle, water is split into hydrogen and oxygen through intermediate Cu-Cl compounds in a relatively low temperature thermochemical cycle. From past studies at ANL and AECL, the Cu-Cl cycle consists of four reactions, which are listed in table 3.2. The Cu-Cl cycle uses the following four reactions to achieve the splitting of water into hydrogen and oxygen:



Table 3.2 Reactions in Cu-Cl Cycle

Step	Reaction	Temp. Range(°C)	Feed/Output	
1	$2\text{Cu}(\text{s}) + 2\text{HCl}(\text{g}) = 2\text{CuCl}(\text{l}) + \text{H}_2(\text{g})$	425-450	Feed:	Electrolytic Cu+dry HCl+Q
			Output:	$\text{H}_2 + \text{CuCl}(\text{l})$ salt
2	$4\text{CuCl}(\text{s}) \rightarrow$ $4\text{CuCl}(\text{aq}) = 2\text{CuCl}_2(\text{aq}) + 2\text{Cu}(\text{s})$	30-70	Feed:	Powder/granular CuCl and HCl+V
			Output:	Electrolytic Cu and water slurry containing HCl and CuCl_2
3	$2\text{CuCl}_2(\text{s}) + \text{H}_2\text{O}(\text{g}) = \text{CuO} \cdot \text{CuCl}_2(\text{s})$ $+ 2\text{HCl}(\text{g})$	400	Feed:	Powder/granular $\text{CuCl}_2 + \text{H}_2\text{O}(\text{g}) + \text{Q}$
			Output:	Powder/granular $\text{CuO} \cdot \text{CuCl}_2 + 2\text{HCl}(\text{g})$
4	$\text{CuO} \cdot \text{CuCl}_2(\text{s}) = 2\text{CuCl}(\text{l}) + 0.5\text{O}_2(\text{g})$	530	Feed:	Powder/granular $\text{CuO} \cdot \text{CuCl}_2(\text{s}) + \text{Q}$
			Output:	Molten CuCl salt+ O_2

(Q = heat, V = electricity)

This Cu-Cl cycle is a closed hybrid cycle that consists of thermochemical and electrochemical processes as shown in Table 3.2. Copper and chlorine are entirely recycled within the system. As shown in the table, the net inputs are heat, feed water, and electricity. In the first step, hydrogen gas and CuCl in a molten state are produced from the reaction of solid Cu with HCl. The second step uses CuCl from the first step and the fourth step to produce Cu, which is required for the first step, through an electrochemical process. In the third step, solid CuCl₂ reacts with H₂O gas to produce a solid CuO·CuCl₂ complex for step 4, and HCl gas for the first step. In the last step, the CuO·CuCl₂ complex is decomposed to produce O₂ gas and CuCl for step 1. Therefore, all chemicals are recycled, except hydrogen and oxygen as products.

Lewis et al. (2003) at ANL have conducted some experiments for the generation of hydrogen (step 1), Cu (step 2), HCl (step 3), and oxygen (step 4). However, there are still a number of challenges associated with individual steps and integration of these steps into a complete cycle, in addition to the challenges related to coupling this cycle to SCWR. The characteristics and requirements of these reactions in the Cu-Cl cycle, as presented by Lewis et al. (2003) are introduced in the next chapter.

ANL used Aspen Plus software to prepare a flowsheet of the Cu-Cl cycle, which is shown in Fig. 3.1 below (Lewis et al., 2005). They defined various unit operations and streams for

the processes. Available experimental data for parameters and process design goals for unknown parameters are used. Lewis et al. (2005) obtained the enthalpies and entropies of the various streams from a physical property databases in Aspen Plus. Since some thermodynamic data for $\text{Cu}_2\text{Cl}_2\text{O}$ were not available, they used the free energy and entropies calculated for equimolar mixtures of CuO and CuCl_2 .

Stream data and heat balances from the simulation are shown in Table 3.3. It presents the inlet and outlet temperatures of each block in the flowsheet, and the operations in the components. Simulation results are provided by ANL. The sum of the enthalpies for the reactions is 3.98 MW(t). The electrochemical and shaft work, after converting work to heat at 50%, is 2.18 and 0.17 MW(t), respectively. Their calculation is based on a hydrogen production rate of 101.73 kg hydrogen per hour or 0.028 kg/s. The low heating value for this rate of hydrogen production is 3.42 MW(t). The open cycle efficiency was calculated as 54% (Lewis et al., 2005).

Rosen et al. (2006) gave a conceptual layout of the Cu-Cl cycle shown in Fig. 3.2, which relates to the flowsheet above. The block diagram represents the flowsheet, except there is one more step added into the block diagram, which is step 3 to dry CuCl_2 .

Table 3.3 Stream data and heat balance for the Aspen Plus Simulation (Lewis et al., 2005)

Block	Stream Data						Temperature, °C		Unit Operation
	In	In	In	In	Out	Out	In	Out	
B10	11				13		25	25	Mixer
B11	13				15		25.001	25	Electrolysis reaction
B12	15				21		25	450	Hydrogen reaction
B83	21				87		450	25	Cool Hydrogen
B84	87				88	89	25	25.007	Hydrogen separator
B31	31				33		24.63	100	Heat CuCl ₂
B32	33				35		100	115.95	Heat CuCl ₂
B33	35				36	40	116	116	Dry CuCl ₂
B41	40				42		115.95	104.81	Condensate CuCl ₂
B42	42				44		104.81	25	Cool CuCl ₂
B1	44				2		25	25.58	Compress CuCl ₂
B51	36				54		116	116	Pump CuCl ₂
B61	61				63H		113	90	Equilibrium reactor
B61A	63H				63I		90	26.85	Cool solid CuCl ₂
B61B	63I				63		26.85	425	Heat CuCl ₂
B62	63				64		425	400	Hydrolysis reaction
B53	64				57	58	400	400	Separate CuO•CuCl ₂
B62A	57				57A		400	89.77	Cool mixtures
B2	57B				47		89.77	232.9	Pump
B54	58				70		-	-	Increase pressure
B70	70				72		375	550	Heat reactants
B71	72				74		-	550	O ₂ generation
B72	74				76		550	25	Cool O ₂
B73	76				77	78	25	25	Separate O ₂
B65	78				50		-	-	Pump
B90	2	47	50	89	91		-	116.99	Recycle mix
B91	91				93		116.99	25	Recycle cool
B92	93				94	95	25	25	Recycle split

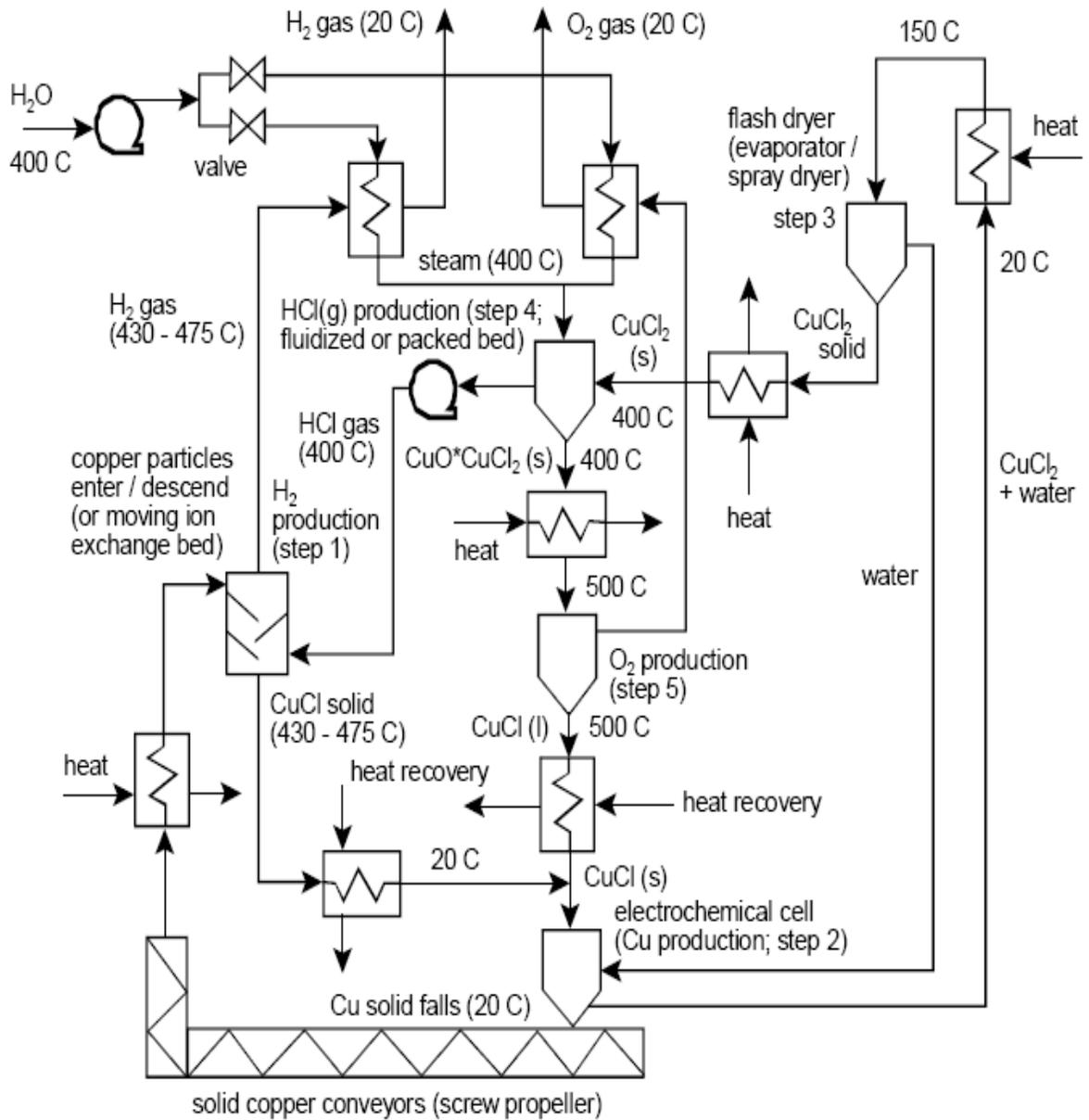


Figure 3.2 Conceptual layout of a Cu-Cl cycle

In Fig. 3.2, the primary components are five interconnected reaction vessels, with intermediate heat exchangers. Rosen et al. (2006) showed that the process of the first step involves three distinct phases and turbulent gas mixing. Copper particles are transported to the mixing chamber, then, they descend along an inclined bed and melt to produce CuCl (l). At the same time, heated HCl gas passes through the chamber to generate hydrogen gas. The challenge is how to improve the reaction efficiency in the chamber of the copper particles mixed with liquid salt and HCl gas passing therein.

The second step of the Cu-Cl cycle is an electrolysis reaction, which requires an operating voltage between 0.4 V and 0.6 V (Lewis et al, 2005). This may be implemented by using an electrochemical cell to produce solid copper particles for the first step (Rosen et al., 2006). For this step, effective heat exchangers are needed to cool the CuCl coming from the first step at about 400 °C. The challenges of this electrolysis reaction are: a) the design of the electrochemical cell and identification of durable membrane and electrode materials; b) the determination of operating parameters that will cause small dendritic copper particles at the cathode (Yildiz et al., 2005).

Step 3 is a hydrolysis reaction that operates at 400 °C. Solid CuCl_2 particles mix with high temperature steam in the reactor to produce solid $\text{CuO}\cdot\text{CuCl}_2$ for Step 4 and HCl gas for Step 1.

From Step 3, solid $\text{CuO}\cdot\text{CuCl}_2$ will be heated up to $530\text{ }^\circ\text{C}$ in Step 4, which is the highest temperature required in the reactions of the Cu-Cl cycle. At that high temperature, $\text{CuO}\cdot\text{CuCl}_2$ is decomposed to produce O_2 and liquid CuCl for Step 2.

Referring to both figures 3.1 and 3.2, heat exchangers have an important role through all reactions in the Cu-Cl cycle. They are needed for heat input and heat recovery to fit the requirement of each reaction. In practice, a series arrangement of heat exchangers may be implemented as shown in Fig. 3.1 to minimize heat losses or cool materials effectively.

3.3 Summary

This chapter has given an overview of the current popular methods of hydrogen generation. Comparing with the advantages and disadvantages of each technology, thermochemical water splitting is a highly promising method. The nuclear-based thermochemical method of the Cu-Cl cycle is described in detail. Additional information about each reaction in the Cu-Cl cycle is introduced in the next chapter to assist the fault tree construction.

Chapter 4 Fault Tree Construction and Multi-State System Application in Fault Tree Analysis

4.1 Overview

In Chapter 3, background information about the Cu-Cl thermochemical cycle was introduced. This chapter constructs fault trees in terms of the knowledge about the Cu-Cl cycle, and the construction process will be introduced briefly.

With failure rate data for the basic events, which are the events at the lowest level of FT, the probability of an undesired top event can be estimated. Since adding the proper number of redundant components is one of the methods to improve system reliability, and place some of the events into MSS, the redundancies will be estimated by a certain demand. Then, the improvement of reliability could be evaluated by repeating to the FTA.

4.2 Fault Tree Construction

In the Cu-Cl cycle, water is split into hydrogen and oxygen through intermediate Cu-Cl compounds, in a low temperature thermochemical cycle. From past studies at ANL and AECL, the Cu-Cl cycle consists of four reactions, which were listed in table 3.2.

The Cu-Cl cycle uses the four sequential reactions to achieve the overall splitting of water into hydrogen and oxygen as follows:



The cycle of the reactions was presented by ANL using an Aspen Plus[®] Simulation, which is shown in Fig. 3.1.

Four reactors corresponding to the four steps in the flow sheet are B12, B11, B62, and B71. Based on part of the mini fault tree theory, the FTs are built separately by four reactions, respectively. Fault tree construction is a top-down analysis process. It starts with the unfavorable operational outcome at the top level (top event). System functional and architectural structures are represented by fault trees. The construction does not consider any human factors, which would act in the scenarios. In addition, there are several undeveloped events, which will be determined in the future.

4.2.1 Fault Tree for Hydrogen Formation

From Table 3.2, step 1 of the Cu-Cl cycle is the reaction of hydrogen generation:



This reaction is exothermic and reversible. As listed in the table, the operating temperature between 425 and 450°C is preferred to avoid the formation of solid CuCl, passivating the copper metal surface (Lewis, 2005). Also, since hydrogen is a flammable gas without odour and color, monitoring leakages of H₂ will be an important issue in future safety. If leakage happens, the hydrogen generation reactor must stop operating, which would influence the operation of the whole system. Other scenarios related to the hydrogen generation process are overheating and overpressure of the reactor, since it is an exothermic reaction and buildup of impurities will influence the release of heat inside.. HCl(g) passes through particles of Cu to produce H₂. The speed of a stream of HCl and the size of Cu particles need to be studied and considered in the fault tree construction.

Based on the above conditions, the fault tree is built and shown in fig. 4.1. The top event of the FT is the failure H₂ formation reaction. There are two main scenarios: failure of the H₂ formation reactor, or excessively incomplete reactions and byproducts, which could cause the top event. In terms of the characteristics of the Step 1 reaction, overheating and overpressure are issues that relate to failure of the reactor. In addition, potential release of H₂ needs to be considered since it is a highly flammable gas with a wide flammability range in air. Then, the previous experience in FT construction helps to determine the lower level events of this FT, which involve sensors or detectors. For another branch of the FT, incomplete reactions and passivation will result in different byproducts through the reaction process. Improper reactants could be caused by poor distribution or buildup of impurities. Passivation will occur at a lower temperature for this step.

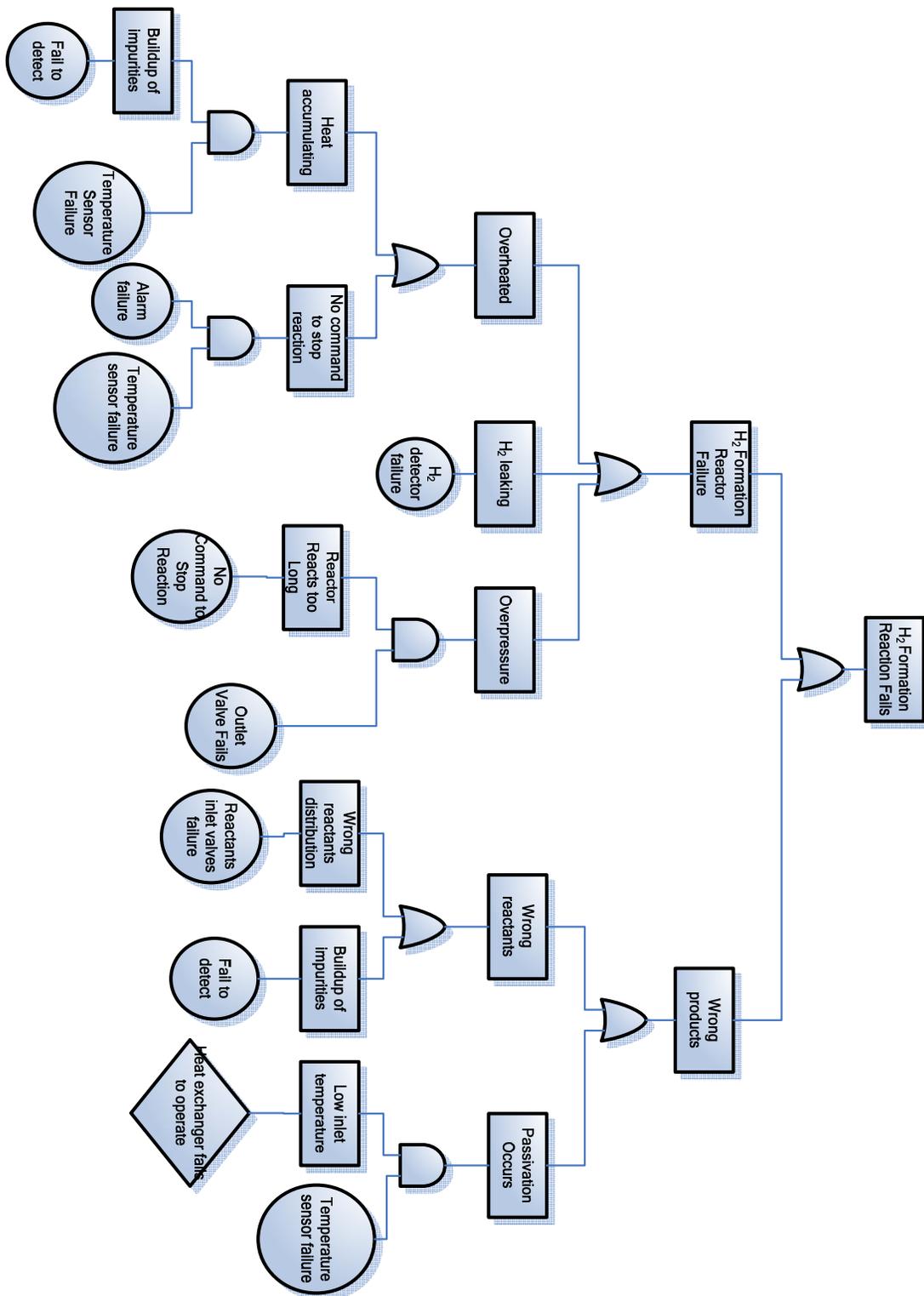
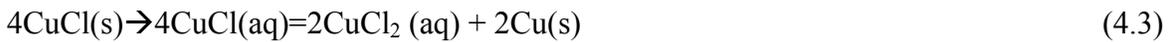


Figure 4.1 Fault tree of Hydrogen Generation (Zhang *et al.*, 2008)

Heat exchanger failure is designated by a diamond because it is an undeveloped event, which will be investigated later.

4.2.2 Fault Tree for Electrolysis Reaction

Step 2 is an electrochemical reaction, which is disproportionation of CuCl exiting from step 1 and 4.



CuCl(s) moves into the electrochemical cell and Cu particles leave on a conveyer going back to step 1 (Rosen *et al.*, 2006). ANL has completed proof of principle experiments for this reaction. In their experiments, the voltage for depositing copper on the cathode varied between 0.4 and 0.6V. There are two challenges to accomplish this electrochemical reaction. The first is development of the electrochemical cell and identification of an appropriate membrane and materials of the electrode. Another aspect is to find the best operating parameters to produce small dendritic copper particles at the cathode and a concentrated CuCl₂ solution at the anode (Lewis, 2005). The first challenge must be considered in the reliability and safety assessment, since it will influence the operating condition. Fig. 4.2 shows the fault tree for the electrochemical process. As mentioned previously, the main issue of this reaction is reliability and durability of the electrodes, as well as the membrane of the reactor. Hence, the FT is constructed as follows.

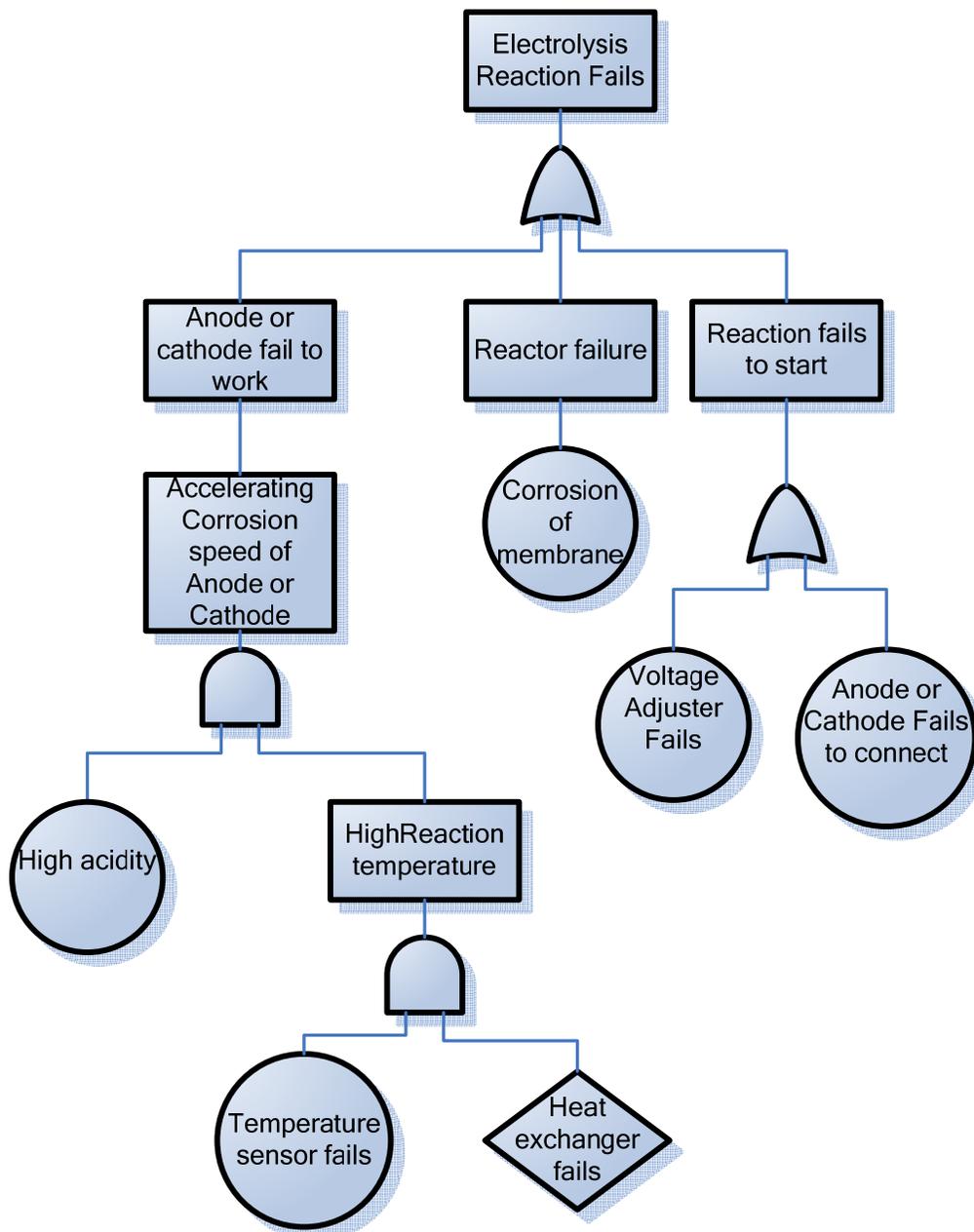
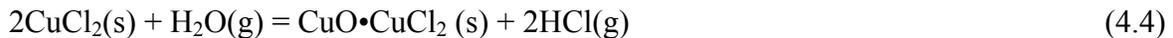


Figure 4.2 Fault tree of electrochemical reaction (Zhang *et al.*, 2008)

The top event is the failure of the electrolysis reaction. Since this reaction operates at about 25 °C, to avoid unexpected scenarios and keep the reaction operating at high efficiency, the temperature sensor and heat exchanger are placed at the bottom as the basic events. In addition, the working voltage should fit the requirement, and the connections for the anode and cathode must work well. This is represented in the right hand branch of the FT.

4.2.3 Fault tree for Hydrolysis Reaction

This step is an endothermic hydrolysis reaction



From past experiments at ANL, a temperature of 300 to 400°C will give reasonable kinetics. The results showed when the temperature was 300°C, the rate of reaction was 2 to 2.5 times slower than 350°C. In addition, the generation rate of a side product, CuCl, strongly relates to the temperature (Lewis, 2005). Another important issue in this reaction is the contact between CuCl₂(s) and H₂O(g), which means what water vapor concentration is needed for the reaction. There are different results reported by Glasner *et al.* (1959) about the influence of temperature and water vapor concentration (Lewis, 2005). The fault tree in fig. 4.3 corresponds to the results provided by ANL. The fault tree is based mainly on temperature since this is the main parameter of the reaction.

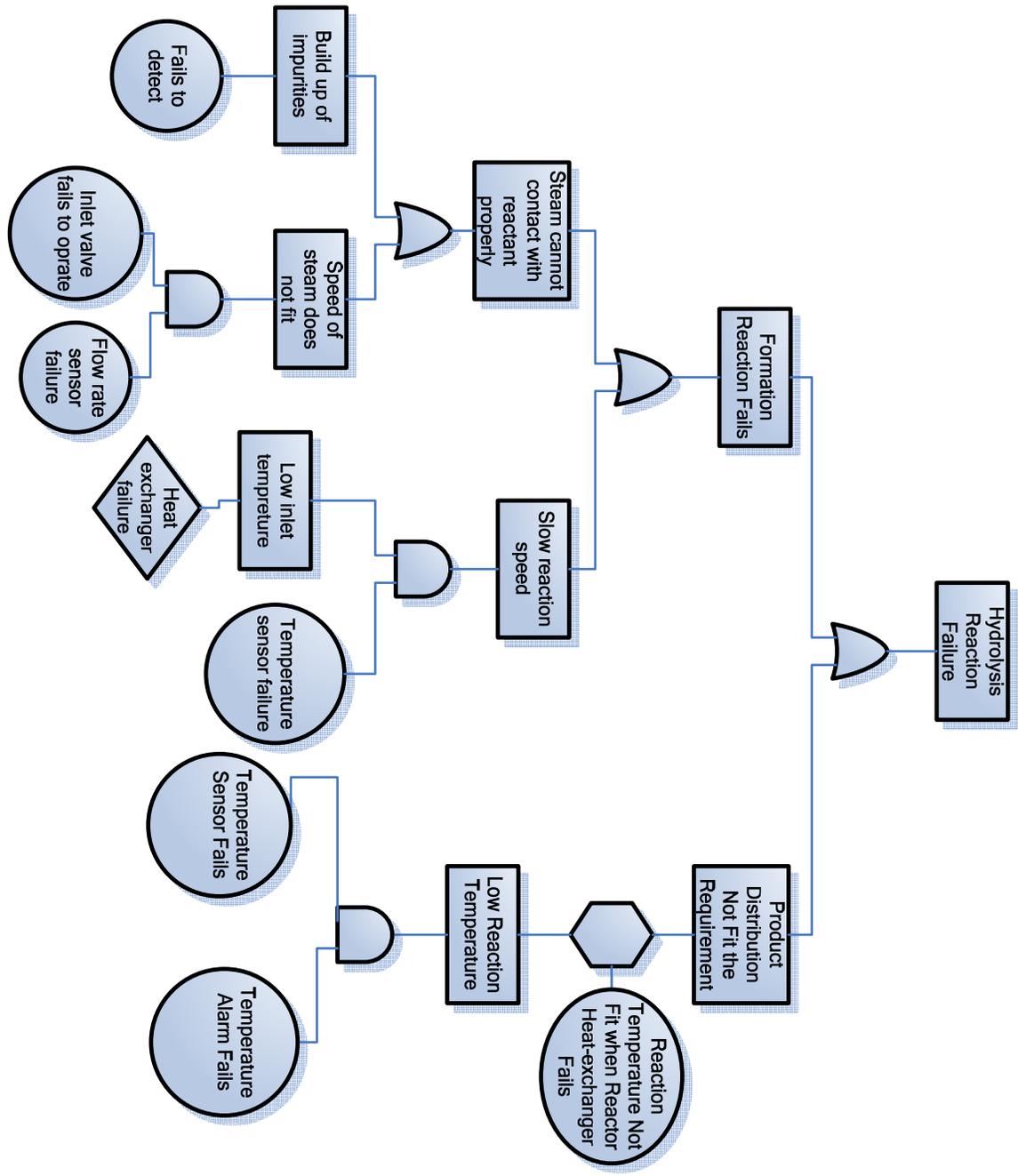
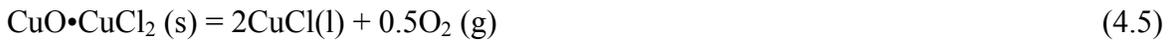


Figure 4.3 Fault tree of Hydrolysis reaction (Zhang *et al.*, 2008)

The left branch on the third level represents the influence of steam on the reaction. A special gate is used on the right side of this fault tree, which is called an inhibit gate, and the event on its right is called a conditional event. When the temperature sensor and alarm fail in a condition of failure of the heat-exchanger at the same time, then the product distribution will be impacted.

4.2.4 Fault Tree for Oxygen Formation

The last step of the Cu-Cl cycle is the oxygen generation reaction. The reactant exits from the third step, which is $\text{Cu}_2\text{Cl}_2\text{O}$ (equimolar mixtures of CuCl_2 and CuO).



It is an endothermic reaction and oxygen is released at a temperature from 450 to 530°C (Lewis, 2005). Therefore, temperature will be one of the important variable in the safety analysis, and it is represented in the fault tree in Fig. 4.4.

The fault tree for oxygen generation is similar to the FT for hydrogen generation. Since the reactant is solid, the impurity remaining in the reactor will be a main issue, which could cause excessively incomplete reactions, byproducts and overheating. Two remaining undeveloped events will be developed later.

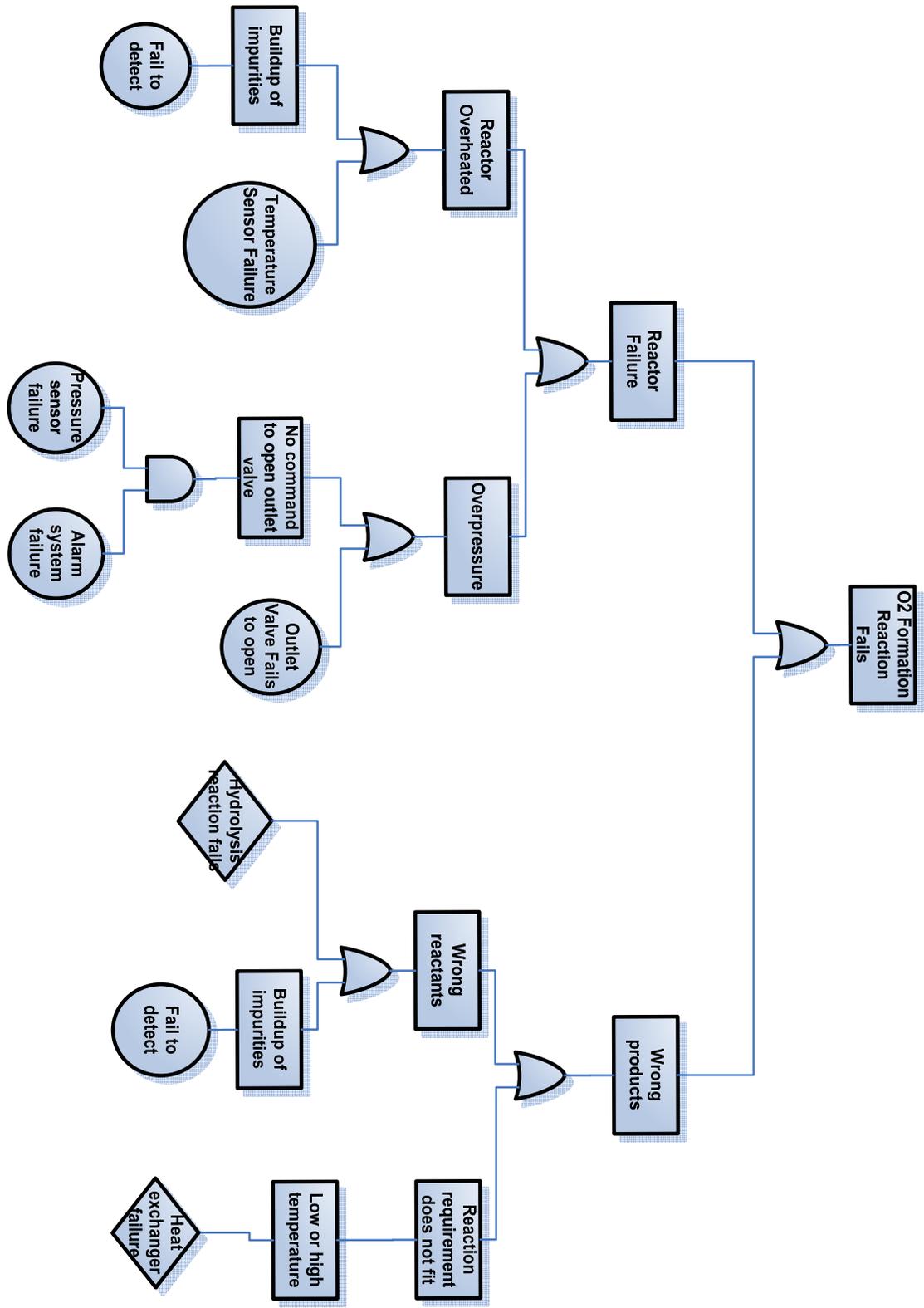


Figure 4.4 Fault tree of Oxygen Generation (Zhang et al., 2008)

The four isolated fault trees are a subset part the entire fault tree of the Cu-Cl system. In Fig. 4.4, heat exchangers have an important role in the cycle. They connect reactors and pipes to reaction and other equipment. They are also mainly responsible for heat input and heat recovery. To finish building the fault trees for the Cu-Cl cycle, functions of heat exchangers and the interrelationship among different components need to be considered in the future research. In addition, there are some undeveloped events in the FTs which are shown by diamonds, and human factors are not considered here.

4.3 Application of Multi-State System

A multi-state system model is a more flexible tool (Barlow and Heidtmann, 1984; Levitin *et al.*, 2003). A power plant which has states 0, 1, 2, 3, and 4 that correspond to generating electricity of 0%, 25%, 50%, 75%, 100% of its full capacity is an example of a multi-state system that has ordered multiple states (Wood, 1985). Most recent studies about multi-state systems assume that all components in a system have the same number of states. However, in a practice world, each component or each different group of components should have different properties, so the number of states should be different (Barlow and Wu, 1978; Zuo and Yam, 1999). One of the available solutions is to define the performance or utility for every component in a multi-state system, which relates to their states. Then, the components in a multi-state system can be compared by their performance (Levitin *et al.*, 2003; Wu and Chan, 2003). The performance is defined as the output ability or the net profit.

4.3.1 Reliability Model

From the FTs in the previous section, one of the important components in of basic events is the sensor. These include temperature sensors and pressure sensors, which has an important role in system control. Also, the performance of the sensors decides the probability of the top events.

The construction of the reliability model of sensors is based on how sensors work. In practise, it may have power supplies, one or several controllers/computers, and alarms in a sensor system. These components will be connected as a series-parallel system as shown in Fig. 4.5.

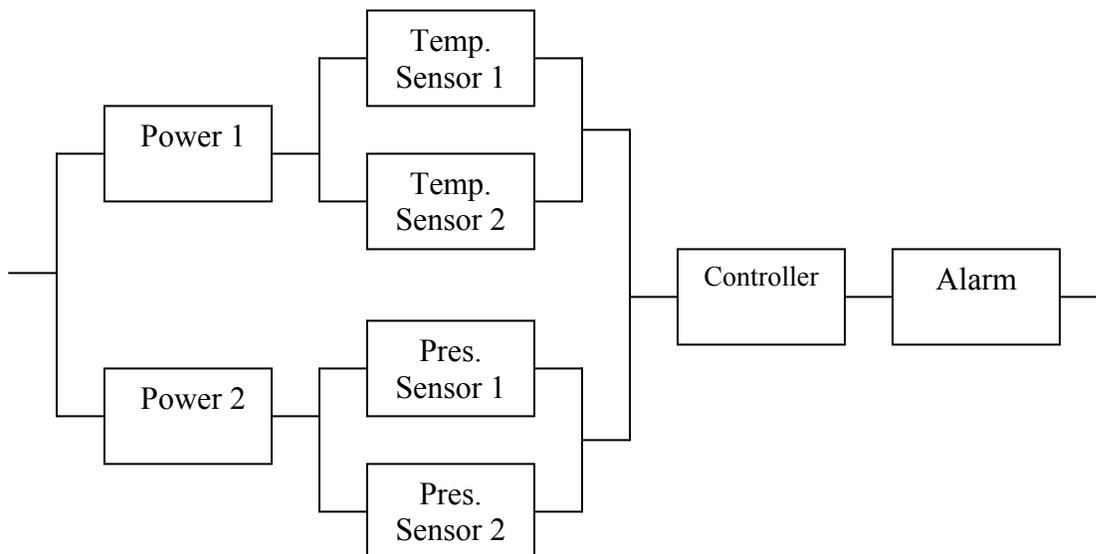


Figure 4.5 Reliability model of sensor system

4.3.2 Multi-State Series-parallel System

For a binary series-parallel system, the reliability is calculated as follows:

$$R_S = \prod_{n=1}^i P_n = \prod_{n=1}^i (1 - \prod_{k=1}^j (1 - P_{nk})) \quad (4.6)$$

$$P_n = 1 - \prod_{k=1}^j (1 - P_{nk}), \quad n=1,2,\dots,j. \quad (4.7)$$

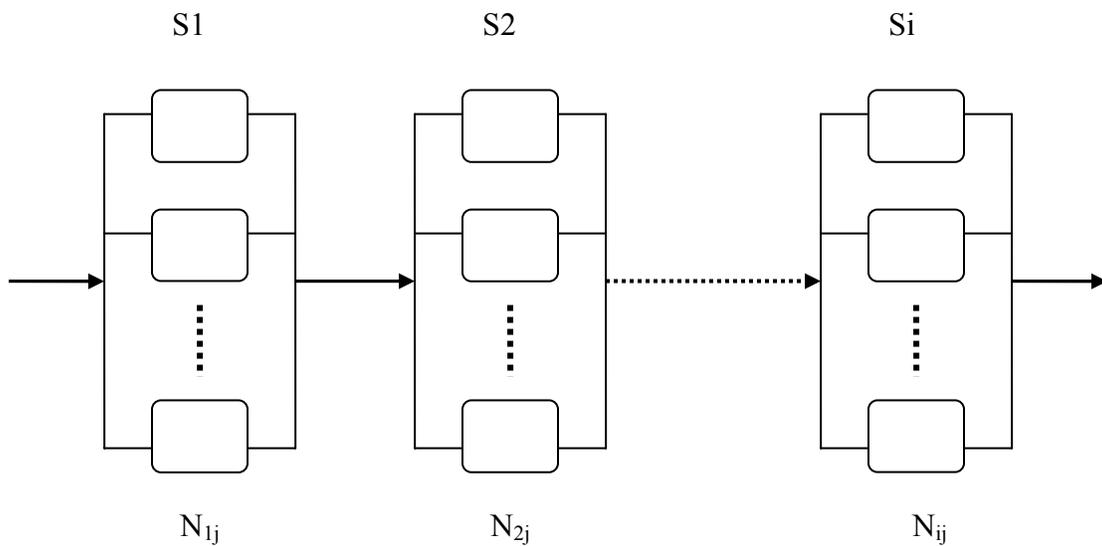


Figure 4.6 Series-parallel system structure

The equations can be expanded to apply to multi-state series-parallel systems. Based on a definition given by El-Newehi and Proschan (1978), where the state of a multi-state series system is equal to the state of the worst component in the system, and the state of a

multi-state parallel system is equal to the state of the best component in the system, each subsystem connected in series can be analyzed individually.

The following Assumptions are adopted.

1. The components in all of the parallel subsystems are independently and identically distributed.
2. Different subsystems have different state parameters and probability distribution.
3. Each state has its own performance. The performance represents the output ability or the net profit.

A common model of a series-parallel system is shown in Fig. 4.7.

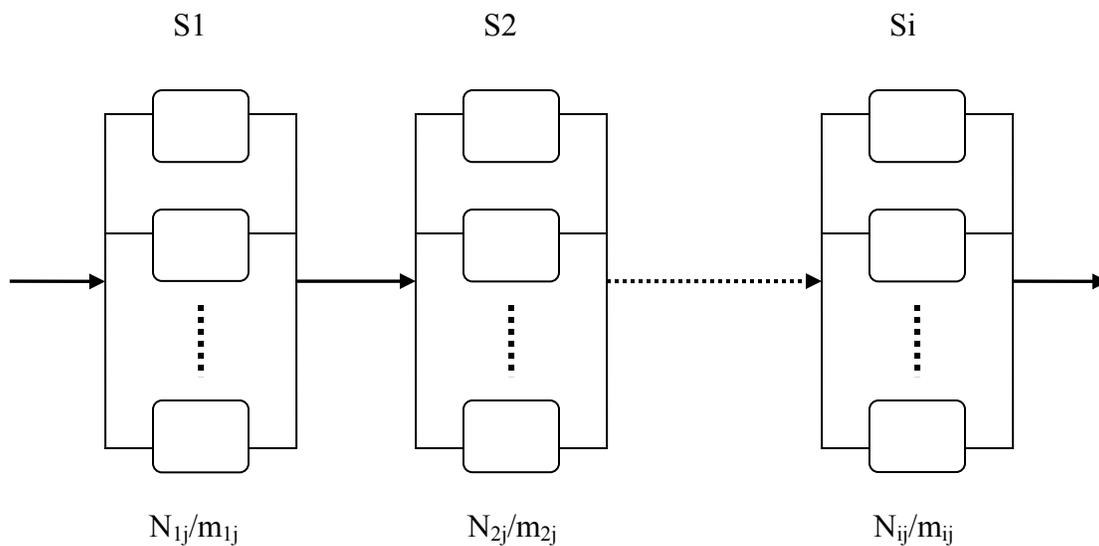


Fig. 4.7 Multi-state series-parallel system

The method of calculating probability distributions of MS series-parallel systems is given below.

$$P_r(\phi_s \geq J) = 1 - \prod_{j=1}^N P_r(m_{ij} < J) = 1 - \prod_{j=1}^N \sum_{m=0}^{J-1} P_{ij}(m) \quad (4.8)$$

$$P_s(J) = P_r(\phi_s = J) = P_r(\phi_s \geq J) - P_r(\phi_s \geq J + 1) \quad (4.9)$$

Since there are more than two states, in order to find the probability when the system is in a specific state, two steps are needed. First, the system state, ϕ_s , when greater or equal to a given state J can be calculated as 1 minus the product of the probabilities when the states of all components in a subsystem are less than the given J (assume all components in a subsystem are the same state). Then, the system probability at state J is obtained.

The performance of a subsystem is the sum of the products of the system probability at a specific state and the component's performance, respectively.

$$P_i = \sum_{J=0}^M P_s(J) x_{iJ} \quad (4.10)$$

$$P_i \geq D \quad (4.11)$$

Because the probability distribution, $P_s(J)$, depends strongly on the number of components in a subsystem, the performance is also decided by the amount of

components. This gives a way to manage MS series-parallel systems to fit the random demands (D).

4.4 Improvement of Fault Tree Evaluation

The number of redundant components can be identified by the MSS analysis. The next step is to re-evaluate the probability of the top event in a FT. To enhance the reliability of a system, the logic AND gate is needed.

4.4.1 Fault Tree Example

Use the temperature sensor as an example. If two temperature sensors need to be installed in a system to monitor the same activity, they can be considered as a group. The FT model is shown in Fig. 4.8.

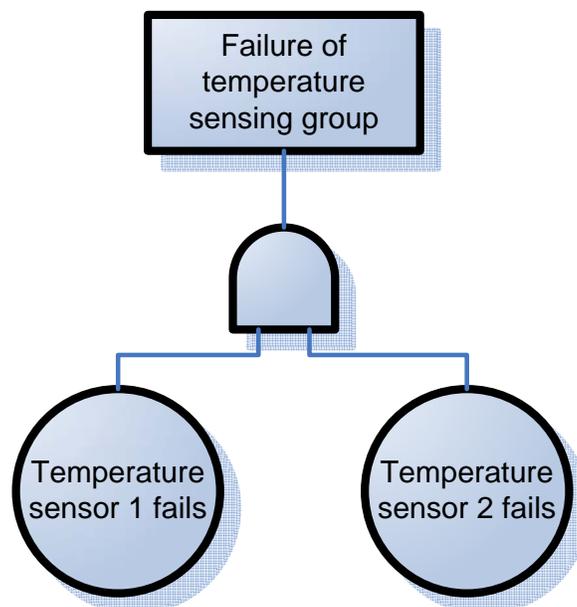


Figure 4.8 Fault tree of temperature sensing group

4.4.2 Failure Rate Estimation

Usually, the failure rates of components used in the product are estimated during a design process. The failure rate of the overall product or equipment is calculated by adding all of the component failure rates. For example, the failure rate of electronic parts is calculated by using MIL-HDBK-217 (Dhillon, 2000). The handbook gives an equation of the following form to estimate failure rates of various electronic components (Dhillon, 2000):

$$\lambda_C = \lambda_b f_q f_e \cdots \quad (4.12)$$

Where λ_b is a base failure rate, which is normally expressed by a model relating the temperature and electrical stress influence on the component under consideration. For many electronic parts, it is obtained by using the following equation:

$$\lambda_b = C \exp\left[-\frac{e_a}{kT_a}\right] \quad (4.13)$$

For MSS, different working states will refer to a different failure rate for the same equipment/part. Thus, the failure rate estimation needs to be performed several times to get various failure rates for every part in MSS.

4.4.3 Fault Tree Analysis

The quantitative FTA for this type of FT is straightforward. The Boolean algebra for an AND gate is used to find the top event probability in Fig. 4.8 when the failure rate of two sensors is constant. Otherwise, when the working state varies, the percentage of each state ranking is needed. The estimation of the overall failure rate equals the sum of the percentage, multiplied by the failure rate, respectively.

4.5 Summary

A new method was developed in this chapter to improve the FT evaluation results by applying an MS series-parallel system. First of all, faults trees were constructed for the four main chemical reaction processes in the Cu-Cl cycle, and the construction processes were introduced briefly. Secondly, the reliability model for basic events, which are the sensors, was given and the model was identified as a series-parallel system. Then, the MSS method for series-parallel system was introduced and used to determine the redundancies based on the reliability model. Finally, the redundancies will improve the system reliability.

Chapter 5 Case Study in the Cu-Cl Cycle

5.1 Overview

The previous chapters introduced FT, FTA, MSS, and nuclear-based hydrogen generation methods, especially focusing on the Cu-Cl cycle. FTA is one of the risk analysis tools, that have been widely used in the past. In addition, MSS is a flexible tool in system reliability analysis, and it gives more accurate evaluation results, compared with binary models. Both of these two techniques were presented in chapter 4, respectively. In this chapter, an example is given to show how MSS and FTA work together to determine the number of redundant components in the system, so as to improve its reliability..

The FT of the oxygen generation is used from Step 4 of Cu-Cl cycle, which is shown in Fig. 5.1 below. The reliability model is the same as Fig. 4.5. Firstly, an MSS analysis will be implemented for the reliability model, without redundancies. It is compared with a random demand to decide whether redundancies need to be added to improve system performance. After redundancies are added, the FTA will be analyzed. The FT analysis software, Relex Studio, will be used to calculate failure frequencies of the system. The basic events of the FT will perform under a multi-state situation. Also, the comparison will be performed in the FTA, for which the system with and without redundancies are added.

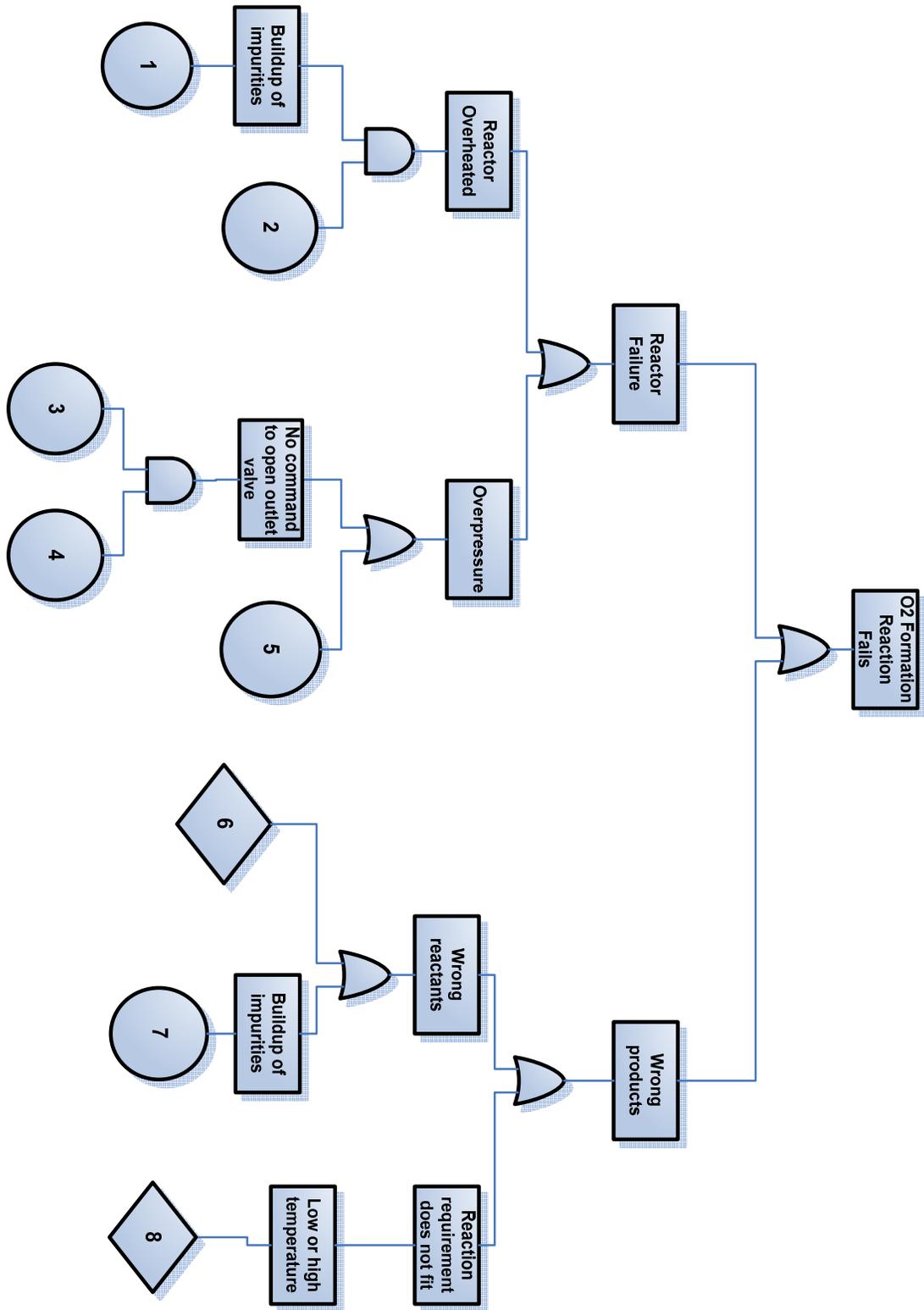


Figure 5.1 Fault tree analysis model

5.2 Multi-State System Analysis

A reliability model is developed first to perform the MSS analysis. Considering the FT (Fig. 5.1), the reliability model (Fig.4.5) from chapter 4 can also be used since the key basic events are temperature sensors, pressure sensors, and alarms.

5.2.1 System Analysis without Redundant Components

Firstly, the performances of the components without redundant components are calculated. The model is shown below in Fig. 5.2.

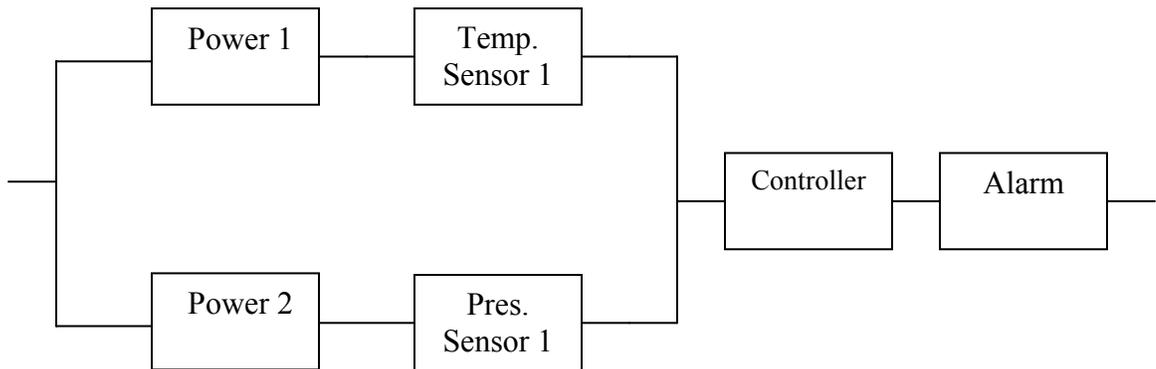


Figure 5.2 Reliability model without redundant component

Fig. 5.3 shows a simplified model with a combination of a parallel system and a series system. The equivalent simplified model becomes a series system shown in Fig. 5.3 below, which has three components. The front component represents the combination of the parallel system in Fig. 5.2. In this model, the three components can be considered as

three subsystems in a series-parallel system. Hence, the system performance equals the worst value among the three components. Then, back to Fig. 5.2, if two branches of the parallel system are dependant, the system performance equals the better results between them. However, these two branches are independent, so the performance should be the worse value, as well for this parallel system.

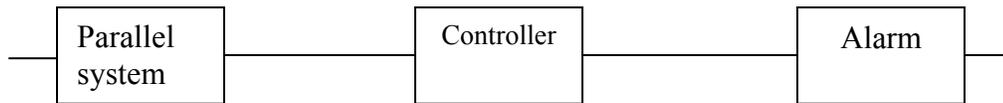


Figure 5.3 Equivalent simplified model of Fig. 5.2

Random defined probabilities refer to each state and performance distribution given in Table. 5.1. P_{ij} represents the probability of component i staying at state J . x_{ij} stands for the performance of component i in state J . Assume a coming Demand is 4.4, currently.

Table 5.1 System probability and performance distribution

State	Power 1&2		Temperature Sensor		Pressure sensor		Controller		Alarm	
	P_{1J}	x_{1J}	P_{2J}	x_{2J}	P_{3J}	x_{3J}	P_{4J}	x_{4J}	P_{5J}	x_{5J}
0	0	0	0	0	0	0	0	0	0	0
1	0.3	3	0.25	1	0.25	1	0.4	2	0.2	2
2	0.7	5	0.45	3	0.45	2	0.6	6	0.8	5
3			0.3	6	0.3	7				

To calculate the performance of each component, Eqns. (4.8) – (4.10) need to be used. Eqn. 4.8 below again calculates the probability when the combination of redundant component states are less than a state J.

$$\begin{aligned}\Pr(\phi_s \geq J) &= 1 - \prod_{j=1}^N P_r(m_{ij} < J) \\ &= 1 - \prod_{j=1}^N \sum_{m=0}^{J-1} P_{ij}(m)\end{aligned}$$

First, the calculation process for Powers 1 & 2 is given below.

$$\Pr(\phi_s \geq 0) = 1 - 0 = 1; \tag{5.1}$$

$$\Pr(\phi_s \geq 1) = 1 - 0 = 1; \tag{5.2}$$

$$\Pr(\phi_s \geq 2) = 1 - 0.3 = 0.7; \tag{5.3}$$

$$\Pr(\phi_s \geq 3) = 1 - 1 = 0. \tag{5.4}$$

Then, Eqn. (4.9), $P_s(J) = P_r(\phi_s = J) = P_r(\phi_s \geq J) - P_r(\phi_s \geq J+1)$, calculates the component performance at a specific state J.

$$P_s(0) = \Pr(\phi_s \geq 0) - \Pr(\phi_s \geq 1) = 1 - 1 = 0; \tag{5.5}$$

$$P_s(1) = \Pr(\phi_s \geq 1) - \Pr(\phi_s \geq 2) = 1 - 0.7 = 0.3; \tag{5.6}$$

$$P_s(2) = \Pr(\phi_s \geq 2) - \Pr(\phi_s \geq 3) = 0.7 - 1 = 0.7. \tag{5.7}$$

After this step, the calculated results are the same as the original values, because there is no redundant component, so the probabilities will not change. As the same rule, the probabilities for other components will not change either.

Then, Eqn. 4.10, $P_i = \sum_{J=0}^M P_S(J) x_{iJ}$, will be used to predict performances. The calculation results are shown in Table 5.2.

Table 5.2 Performance of each component

	Power 1&2	Temperature sensor	Pressure sensor	Controller	Alarm
Performance	4.4	3.4	3.25	4.4	4.4

From the results, the performance of the temperature sensor and pressure sensor is lower than the coming demand, which is 4.4. Therefore, the improvement of the system needs to be accomplished to make it fit the demand. The other three components will not be adjusted since their performance values are just 4.4.

As mentioned in chapter 2, there are mainly two ways to improve the system reliability. One method is trying to improve the component's capability itself; while another is adding redundancies. In this thesis, the second method is adopted, and it will be discussed in the next section.

5.2.2 System Improvement by Adding Redundant Components

Determining the number of components that need to be added is often a process of trial and error. Since the system performance is unknown when adding different numbers of components, the common procedure is to add one more component, and then calculate the results. Repeat this step until the performance is equal or higher than the demand.

For this example, it will start by adding one more temperature sensor and pressure sensor into the system. The structure of the system is shown in Fig. 5.4.

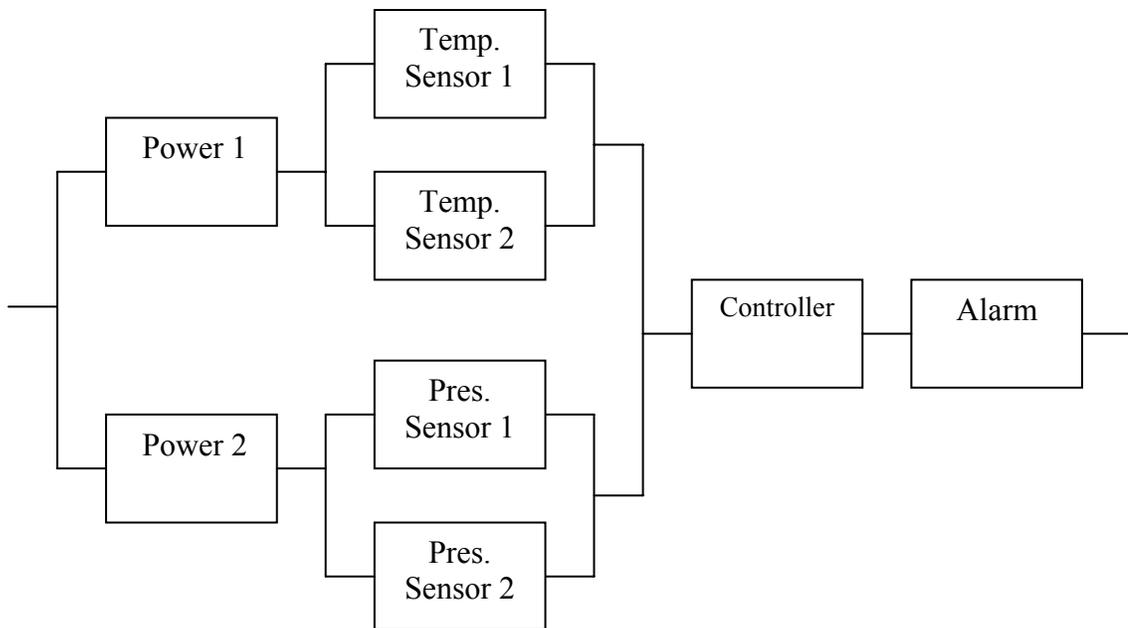


Figure 5.4 Reliability model of redundant system

Since the probability distributions of the temperature sensor and pressure sensor are the same, the probability of the temperature sensor combination will be the same as pressure sensors as well. The calculation is shown below, which uses the same equations (Eqn. 4.8 - 4.10):

$$\Pr(\phi_s \geq 0) = 1 - 0 = 1; \quad (5.8)$$

$$\Pr(\phi_s \geq 1) = 1 - 0 = 1; \quad (5.9)$$

$$\Pr(\phi_s \geq 2) = 1 - \prod_{j=1}^2 0.25 = 0.9375; \quad (5.10)$$

$$\Pr(\phi_s \geq 3) = 1 - \prod_{j=1}^2 0.7 = 0.51; \quad (5.11)$$

$$\Pr(\phi_s \geq 4) = 1 - \prod_{j=1}^2 1 = 0. \quad (5.12)$$

Then,

$$P_s(0) = \Pr(\phi_s \geq 0) - \Pr(\phi_s \geq 1) = 0; \quad (5.13)$$

So, $P_s(1) = 0.0625$; $P_s(2) = 0.4275$; $P_s(3) = 0.51$.

Using Eqn. 4.11, the performance of the temperature sensor system is increased to 4.405.

These same probabilities used for the pressure sensor system will increase the performance up to 4.48.

By adding sensors, both of the temperature and pressure sensor systems satisfy the demand, which is 4.4, so the calculation is completed. If the demand changes, the number of the component needs to be increased or decreased until the performance meets the demand.

5.3 Fault Tree Analysis for a Multi-State Redundant System

In this section, the FTA with MSS will be conducted. The FT is shown in Fig. 5.1. To make it readily represented, all of the basic events are changed to numbers: 1 represents “Fail to detect”; 2 is “Temperature sensor failure”; 3 is “Pressure sensor failure”; 4 is “Alarm system failure”; 5 is “Outlet valve fails to open”; 6 is “Hydrolysis reaction fails”; 7 is “Fail to detect”; and the last number 8 is “Heat exchanger failure”.

Similarly to the previous section, two temperature sensors and two pressure sensors are needed for the system. To perform the calculation, Relex Studio, a popular reliability analysis software tool, will be used.

The failure rates of these numbered events are shown in Table 5.3. The data listed here refer to each state, which are randomly defined. Since numbers 6 and 8 are undeveloped events, they are not considered in the calculation. The equivalent failure rate (EFR) of these components equals the sum of each probability value multiplied by its failure rate, respectively.

Table 5.3 Reliability data for calculation

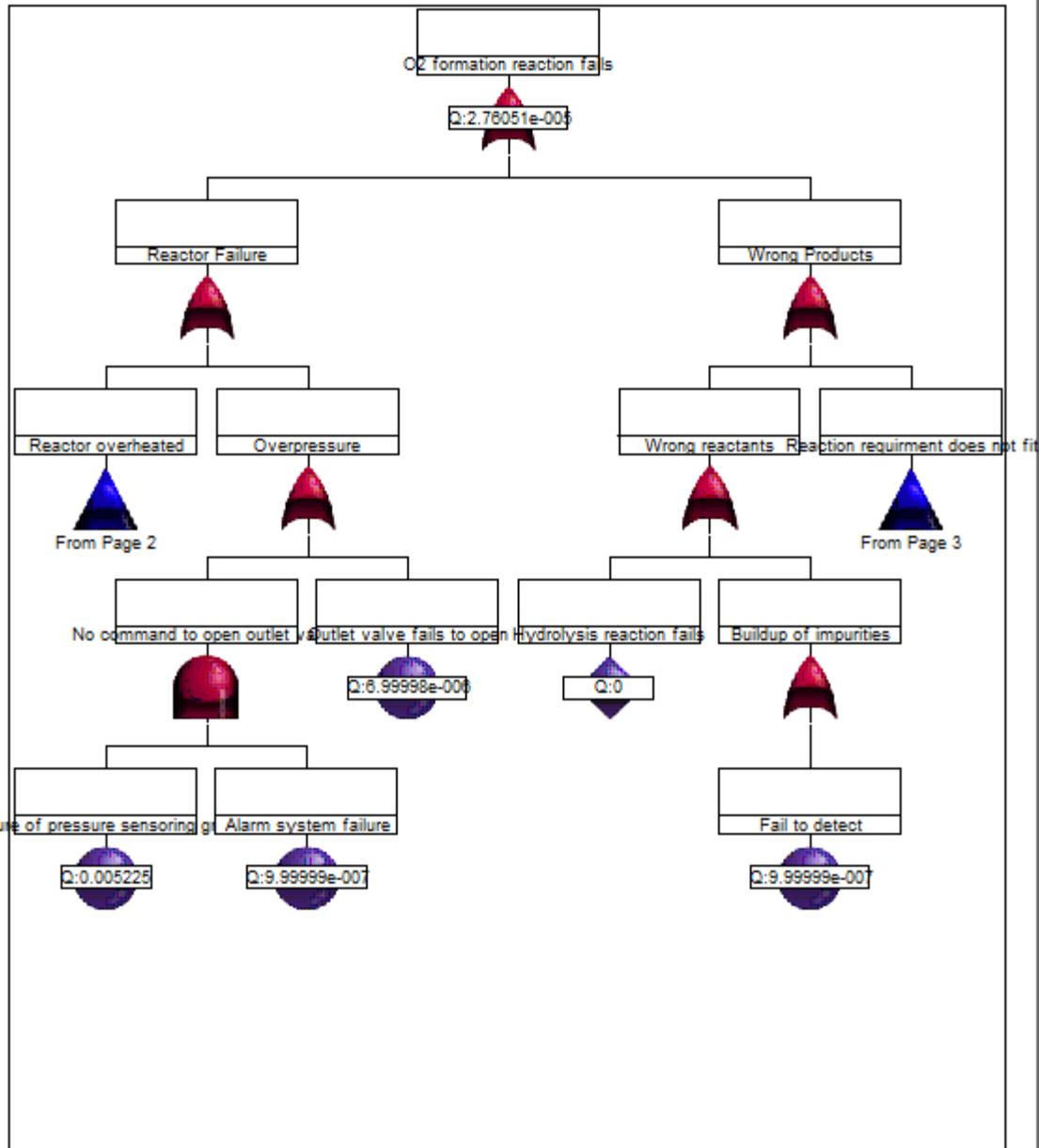
State	1		2		3		4		5		7	
	P ₁	λ ₁	P ₂	λ ₂	P ₃	λ ₃	P ₄	λ ₄	P ₅	λ ₅	P ₇	λ ₇
0	0	-	0	-	0	-	0	-	0	-	0	-
1	1	0.01	0.25	0.006	0.25	0.0035	1	0.001	0.25	0.007	1	0.01
2			0.45	0.008	0.45	0.005			0.45	0.01		
3			0.3	0.015	0.3	0.007			0.3	0.025		
EFR	0.01		0.0096		0.005225		0.001		0.007		0.01	

(P represents the probability of each event in a specific state; λ represents the failure rate)

Figs. 5.4(a), (b), (c) show the FT developed by Relex Studio, and the calculation results are shown in Fig. 5.5. To calculate the failure frequencies, both the failure rate percentage and exposure time percentage are set to 100.

The temperature and pressure sensing groups are built as shown in Fig. 4.8 to replace the original single sensor, and two And Gates are used. The new failure rate of each sensing group equals the square of its EFR.

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Figure 5.5(a) Fault tree model constructed by using Relax Studio

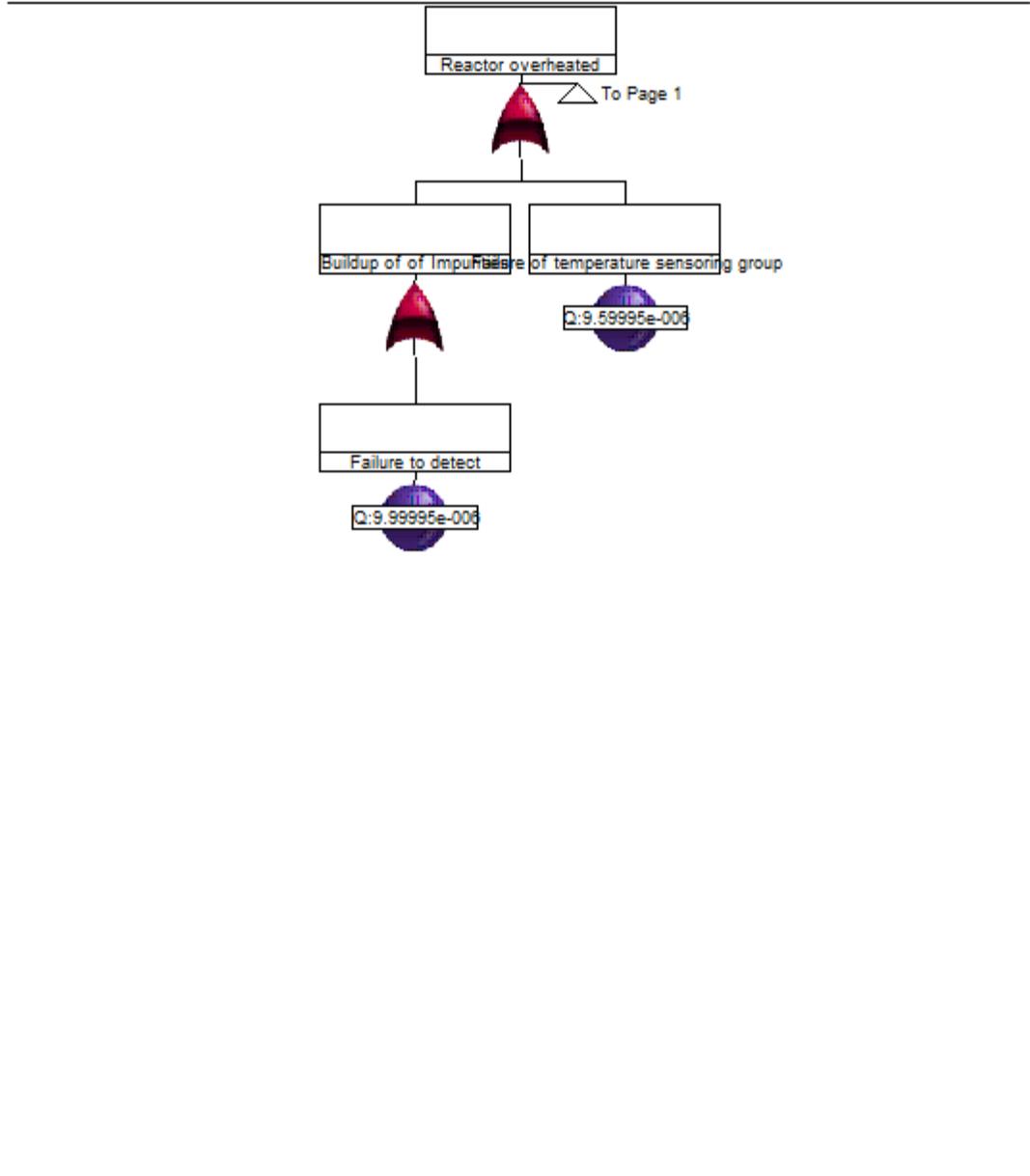


Figure 5.5(b) Fault tree model constructed by using Relax Studio

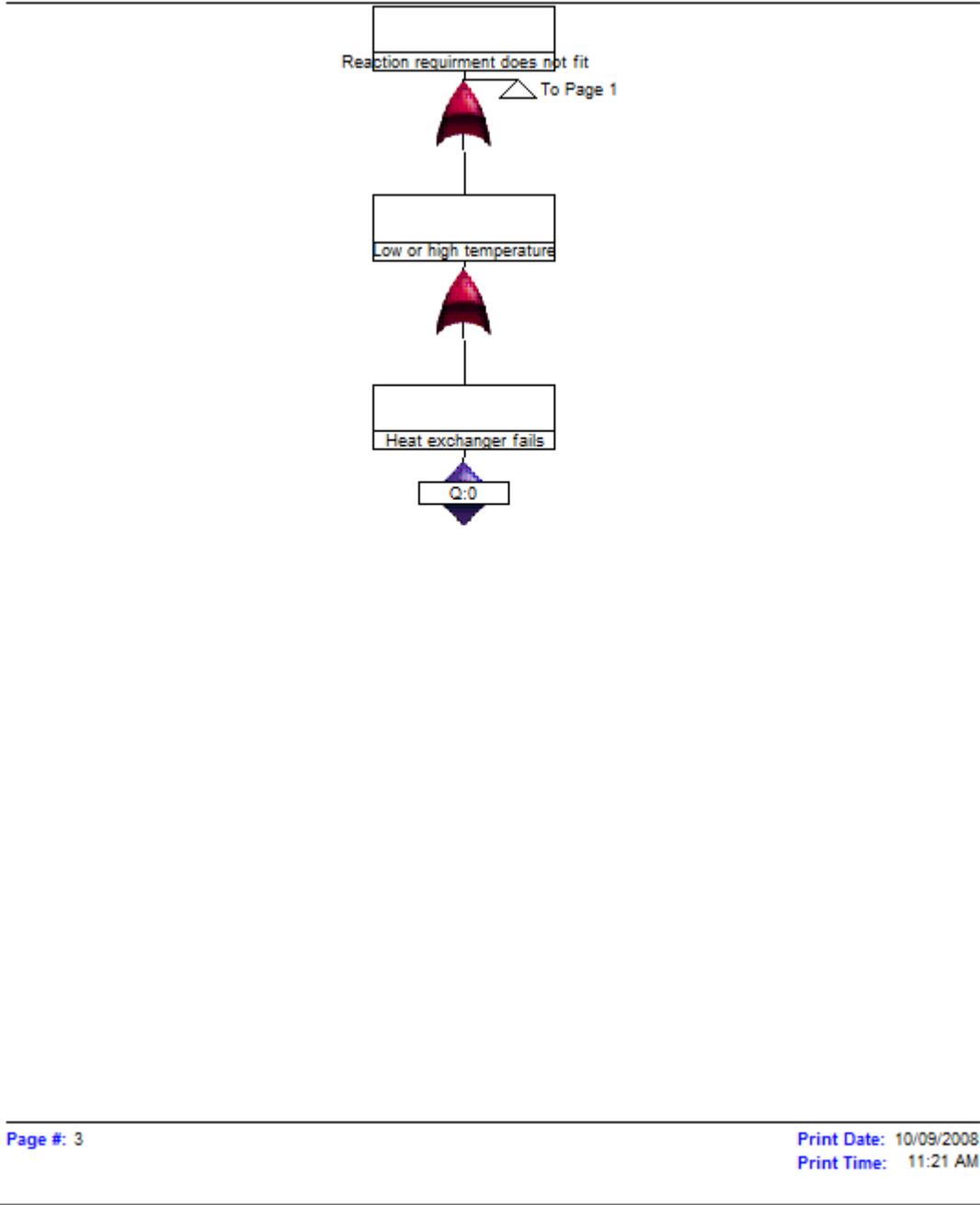


Figure 5.5(c) Fault tree model constructed by using Relax Studio

Substitute the data into FT, then after 1,000 hours, the system unavailability, unreliability, and failure frequency are 0.010017, 0.010017, and 0.017092, respectively, which are shown in the figure below.

Table 5.4 Fault tree analysis results for redundant system

View Calculation Results

Results for Gate: O2 formation reaction fails

Results at time 1000.00:

Unreliability (F): 0.01001709 Number of Failures: 1.7092086e-005
 Unavailability (Q): 0.01001709 Frequency (f): 0.01709201

Time	Unavailability	Unreliability	Failure Frequency
0	0.010000	0.010000	0.017092
100.00	0.010002	0.010002	0.017092
200.00	0.010003	0.010003	0.017092
300.00	0.010005	0.010005	0.017092
400.00	0.010007	0.010007	0.017092
500.00	0.010009	0.010009	0.017092
600.00	0.010010	0.010010	0.017092
700.00	0.010012	0.010012	0.017092
800.00	0.010014	0.010014	0.017092
900.00	0.010015	0.010015	0.017092
1000.00	0.010017	0.010017	0.017092

Close Help

The calculation results for the original FT are given in Fig. 5.6 by following the same steps. Compared with the redundant system, the unavailability, unreliability, and failure frequency are all higher, which means that this system is weaker.

Table 5.5 Fault tree analysis results for simple system

View Calculation Results

Results for Gate: O2 formation reaction fails

Results at time 1000.00:

Unreliability (F): 0.01002661 Number of Failures: 2.6605104e-005

Unavailability (Q): 0.01002661 Frequency (f): 0.02660498

Time	Unavailability	Unreliability	Failure Frequency
0	0.010000	0.010000	0.026605
100.00	0.010003	0.010003	0.026605
200.00	0.010005	0.010005	0.026605
300.00	0.010008	0.010008	0.026605
400.00	0.010011	0.010011	0.026605
500.00	0.010013	0.010013	0.026605
600.00	0.010016	0.010016	0.026605
700.00	0.010019	0.010019	0.026605
800.00	0.010021	0.010021	0.026605
900.00	0.010024	0.010024	0.026605
1000.00	0.010027	0.010027	0.026605

Close Help

5.4 Summary

This chapter has shown how MSS works to help identify the number of redundant components at the beginning. Then, the redundancies were substituted back to the FT,

and the key parameters were calculated by using Relex Studio, such as the unavailability, unreliability, and failure frequency. In order to show the improvement by adding redundancies, the comparison with the original FT is made at the end. In addition, the FTA applied the concept of multi states, which is different from the traditional binary FTA. This approach can give more accurate evaluation results.

Chapter 6 Conclusions and Recommendations for Future Work

6.1 Conclusions

The thermochemical Cu-Cl cycle is one of the promising methods for generating hydrogen. If the hydrogen generation system is built close to a nuclear plant, and hydrogen is a flammable gas, the safety and reliability of the system become more important. The risk analysis of this large system will be complex and time consuming work. To make it easier and more efficient, this thesis presented a new method that combined a fault tree analysis and multi-state series-parallel system. Redundancies can be calculated through the reliability model. The series-parallel model is widely used in reliability analysis, so it can be applied to Cu-Cl cycle. In addition, MSS is used in the reliability analysis to have better estimations than a binary system since more states will represent system conditions better. Some remaining challenges are how to determine the demand for MSS, find the probabilities for different states, and identify different failure rates in various states.

6.2 Future Research

To analyze the thermochemical Cu-Cl cycle, a computer aided method needs to be developed in the future to analyze the complex system. To add redundancies for a large fault tree, the basic events in minimal cut sets should be considered first. In practise, there

are standby parts, so how to deal them with redundancies should be examined. Also, human factors need to be considered further in future research.

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