SESSION XIII

INTERNATIONAL SYMPOSIUM ON THE MANAGEMENT OF WASTES FROM THE NUCLEAR FUEL CYCLE

Wednesday, August 4, 1976
CHAIRMAN: R. W. Ramsey

OVERVIEW OF SYMPOSIUM AND EXPLANATION OF TECHNICAL ALTERNATIVES DOCUMENT (TAD)

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TECHNICAL OVERVIEW OF AIR CLEANING ASPECTS OF SYMPOSIUM AND TAD

R. A. Brown

SUMMARIES OF AVAILABLE TECHNOLOGY ON GASEOUS EFFLUENT CONTROL OF KRYPTON, IODINE, TRITIUM, ^{14}CARBON, RUTHENIUM, NOx, HCl, AND PARTICULATES

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OVERVIEW OF SYMPOSIUM AND EXPLANATION OF TECHNICAL ALTERNATIVES DOCUMENT (TAD)

R. W. Ramsey, ERDA

I am serving as session chairman in place of Alex Perge. He was unable to make this, although he made some preparation to do so. I am going to try to give you his impressions and some other impressions that I have gathered from discussions on the topics that are on the agenda.

First, I would like to give you some overview of the Denver Symposium and then an explanation of the Technical Alternatives Document and its relationship to the Symposium and the environmental impact statement.

Then, I would like to turn the program over to Russel Brown, who will cover the air cleaning aspects of the TAD document as an introduction to panel discussions we plan to have later. We will follow this with excerpts from each of the items that were covered in the TAD by those who contributed them.

Following that, we will set up this room in a colloquial arrangement and we will make the several TAD authors available to you for questioning and to talk to you about specific aspects of their part of the TAD documents. In that way, we hope to give everyone a chance to get their questions answered or discussed. Some of you may find it helpful to stand and listen to the questions of others. I think this informal atmosphere of exchange will be very useful to close this meeting. When we do break for the seminar discussion, you may consider that the adjournment of the 14th Air Cleaning Conference.

The International Symposium on the Management of Wastes from the Nuclear Fuel Cycle was held in Denver on July 11-16, 1976, and many of you here attended that meeting. That meeting and the technical alternatives document, called TAD for short, is referred to as ERDA 76-43, Volume I through IV. That document and the meeting are related to the requirement for an environmental impact statement discussing the generic programs for management of radioactive wastes. The idea of a meeting and TAD came into being shortly after the withdrawal of the initial environmental impact statment in April, 1975. It was one of the first actions of the new administrator of ERDA, who withdrew the initial waste management environmental impact statement and promised the preparation of a replacement.

The TAD document was addressed to critical comments that had been made about the initial environmental impact statement. First of all, alternatives were not discussed in that initial EIS and we did not address ultimate disposition adequately. In particular, not enough information was made available to assess the feasibility of the various plans that were proposed in that environmental impact statement and not enough time was given for review and discussion or for experts to interact and give advice on a selection of methods to be used in the management of waste. Therefore, one of the purposes of the Symposium was to provide a timely opportunity for open discussion of the technology. It was made an international meeting so that this discussion of technology could be put on a worldwide basis. Therefore, it covered all the things being developed throughout the world to address the waste management problem.

The Symposium was planned to follow the issuance of a major document [TAD, ERDA 76-43], but to convene well in advance of the issuance of a new generic environmental impact statement. It was to be held at a central location for convenient attendance and to be an open meeting with attendance made available to anyone who wanteo to come, with the program arranged to discuss all of the topics that relate to the technology of waste management and to provide people with an opportunity to ask questions.

The results of the conference are difficult to assess. Everyone has different observations about it. Many are critical. However, the important facts to me seem to be as follows: It was attended by about 800 people. Many of the attendees were not members of the so-called "nuclear club". •The sessions were well attended by a broad group, including just interested people. There were many representatives of utilities, AE's, state and local health officials, university consultants, and, considering the Olympics and the democratic national convention that were being held in parallel, it got reasonable press coverage.

Some people were disappointed by the conference, however. The reasons I have heard are, first of all, it did not enunciate a solution that some people though should be enunciated at such a meeting. Instead, it illuminated alternatives. It did not become a confrontation of opposing and advocate forces: although there was some flavor of that type in specific discussions. It was not as widely publicized as had been hoped, and some of the coverage, as is normal, left distorted impressions; but these were not too serious. Probably the most universal criticism has been that it did not discuss the sociological issues of the question of waste management from the nuclear fuel cycle, which everybody seems to think ought to be discussed by experts. Maybe it is because we don't feel expert in the sociological issues that we don't discuss them. It was recognized that this was to be a purely technical meeting; as it was. But another meeting has now been arranged. It is to be held by the Congress of Environmental Quality on October 27th through the 29th, in Chicago. This should afford an excellent opportunity for discussion of the sociological aspects and be a supplement to the technical program discussed at Denver.

In summary, my observations are that the Denver meeting was ambitious in scope. It was timely to the need for public discussion. It was reasonably accessible to anyone interested. Unfortunately, it will probably be judged by others as another highly technical gathering of the fuel cycle fraternities, rather than a public meeting to eliminate the issues.

Now, to turn to the TAD document. I mentioned that it, the Denver meeting, and the environmental impact statement are closely related. Both TAD and the Denver meeting are key elements of the program to document the generic environmental impact associated with waste from the postfission nuclear cycle. You will hear that title used many times during the discussion of these documents. The environmental impact statement is now planned for issuance in draft form about April, 1977. It is a formidable task to write it because of its volume and coverage, as well as the schedule. As many of the officers will tell you, the TAD was just the first milestone of the environmental impact statement plan. TAD's objective was to provide a comprehensive and authoritative technical document for public consumption in sufficient time to allow it to be digested and discussed. The second milestone is the several meetings and conferences that are following the issuance of TAD. The Denver meeting I mentioned, is, of course, a cardinal milestone, as is the CEQ conference, the 14th Air Cleaning Conference, the ANS-AIChE meeting after it, and the ANS meeting in Washington, this fall. All the various meetings with topics on waste management that will be discussed by experts, are regarded as a part of this illumination. I understand that the University of Arizona is planning a waste management meeting in Tucson this fall. It is evident that a number of the institutions that have sponsored discussion of this topic are getting in at this time in an effort to give us as much illumination as possible.

The next milestone is the issuance of the draft environmental impact statement itself. A review will also follow it at various meetings. I am not an expert on this part of the plan, however, so I can't give you an exact schedule for what happens after the EIS is issued.

In summary, then, the TAD has satisfied an important objective of making all the technology publicly available well in advance and of documenting the technology for referenee in the environmental impact statement. It is hoped that it will give ample time and opportunity for all the experts to digest it and give their advice.

Now, one personal comment is necessary, I think. I believe that there has been an inordinate diversion of technical expertise and creativity from real technical work for the purpose of documentation. This is extremely costly, and we must realize the costs. I don't know whether we can afford it, frankly. I am impressed that we see the same names on papers on this subject and on our progress reports and on the attendance lists of meetings such as this. The authors and contributors to the TAD and to the environmental impact statement are a very select group of people dealing with this subject of radioactive waste. These people are productive and good managers of their time, but I can't help but consider the cost of their continuous documentation efforts to the degree that they take them away from creative productivity in their labs and test facilities. I hope that we, in the field, recognize this problem and that we can think of a way to avoid this cost. Our country really needs to get on with solving this problem instead of getting on with documenting it. I am concerned that we are spending too much effort writing it down instead of doing it right. If you agree, why not let your congressman know your feelings about this, just as environmentalists let theirs know about their feelings?

With these comments, I will now ask Russell Brown to take over and give you an overview of the air cleaning aspects of the document.

TECHNICAL OVERVIEW OF AIR CLEANING ASPECTS OF SYMPOSIUM AND TAD

R. A. Brown, Allied Chemical, INEL

I'm going to call upon each of the task leaders of TAD to present a short summary but first I will give you a quick overview of what TAD is supposed to accomplish in the context of the gaseous waste problem and my impression of the Denver meeting. It bears out Bob Ramsey's description of the Denver meeting in that the world is distinctly not interested in the characteristics or status of technology for waste management. There was public participation and public attendance, and there were questions asked, but technology is not what stimulates people's curiosity. Those who were specifically involved and, I believe, properly involved in advocacy positions in opposition to nuclear power are more interested in finding the problems than hearing about potential solutions. I think when we went to that meeting, we had to bear that in mind. That is a very subjective view.

TAD was subject to several constraints in its preparation. Those of you who had a chance to thumb through the report, (and I don't imagine too many people have done so other than those who were involved in putting it together and for this reason had the opportunity) can see that it's very limited, very constrained, in what it does.

I thought I'd touch upon the same things I did in the introductory remarks to give you the flavor of it. Then we can open it up to the individual task leaders and they can give you a quick summary of what they said. By the way, I gave a paper on one subsection of this report at the Denver meeting and it took me thirty minutes to cover 22 different technologies. I can't help but feel that I didn't even do a service to the listeners of that meeting. I think anyone who presents a paper that comes from, essentially, something like an encyclopedia, must have reservations about presenting papers. Those who attended the meeting had an opportunity to hear it all at one place. But for the speakers, I think it left many of us nervous about doing it.

The TAD document is really just an encyclopedia of technology. It lists and describes. It's very limited in what it evaluates. In fact, its mission was not to evaluate technologies except in a very narrow sense. Technologies were characterized in terms of their availability. The dividing line between those that were judged available and those requiring development was often, of necessity, fuzzy.

Environmental impact was not discussed. That will be discussed in the next stage, the preparation of the generic environmental statement. I view preparation of that statement as a monumental job and, I fear, especially since I'm going to be involved in one aspect of it, that we haven't been given enough time to do the job that should be done to write a generic statement.

Economic analyses were not presented. They will be presented in, and used in, the generic environmental statement. Decisions and plans were not presented. The choice of technologies that were, and were not, included in the document is open to some debate and I hope, in our small discussions today, that if you have questions about what was or was not included, you will raise them.

I also view the next stage as a learning process to find out where we slipped up or where we made erroneous statements. We need to hear about them. We even discussed a few nonradioactive ways. They will not be included in the generic statement, but we included them just because we felt they were of interest and potentially significant.

As you can see from the list I've just given you, the document contains more proscriptions than prescriptions. The TAD can probably be criticzed for all of the things that it couldn't or didn't do. Those who desired ultimate storage of waste, are going to be disappointed. Those who wanted economic or environmental analysis, will find the report unsatisfactory. Those who expect some clear proof or demonstration that all of these management problems are truly solved, once and for all, will be unhappy. The material in TAD could not meet the expectations of all who read it, but the complete report does provide a valuable resource for those who want to know about the technologies of waste management. It provides a simple description of the known methods for handling waste. Those descriptions, combined with rather extensive references, give all readers a chance to know and learn more about recovery and stroage methods. The TAD provides, above all else, a starting point for analysis, evaluation, and discussion, and an expansion of our knowledge of these technologies. I believe that a review by the participants in this conference and by readers in both the national and international technical community will provide the necessary feedback for future additions to and refinements of the document.

With that presented, I would like to call upon those people who will lead the seminar groups. The first will be Dieter Knecht who will give us a very brief review of krypton.

SUMMARIES OF AVAILABLE TECHNOLOGY ON GASEOUS EFFLUENT CONTROL OF KRYPTON, IODINE, TRITIUM, ^{14}CARBON, RUTHENIUM, NOx, HCl, AND PARTICULATES

D. A. Knecht, Allied Chemical, INEL - Of the several krypton 85 methods of collection and storage, the first to be considered is cryogenic distillation. There is available technology which requires a demonstration at a commercial fuel reprocessing plant. Liquid fluorocarbon absorption or, as it was called in the talk today, the FASTER Process, is one that requires additional development. I believe the time scale that was given this morning was around two or three years for commercialization. I think we're getting to the point where they could apply it to a commercial-scale plant. Maybe I'm wrong there. We didn't include other techniques such as solid bed adsorption. We didn't consider them developed enough at this time for fuel reprocessing plant scale. If there are other opinions on this, we would be very happy to hear about them at the later discussion.

In storage technologies, we considered pressurized cylinder storage as an available technology. It is one that requires demonstration. It needs design, but we felt everything was available to do this. Zeolite encapsulation is one of the techniques of immobilization of krypton we felt had promise, but required, again, additional development before it could be incorporated into commercial fuel reprocessing scale. There are other methods of entrapping krypton being investigated on a laboratory scale. But, again, we didn't include them at this point because they need a lot more work before decisions can be made on their applicability to a reprocssing plant.

R. A. Brown, Allied Chemical, INEL - We didn't mention that the cutoff date for technologies was supposed to be September 25th. Although we deviated by a few months here or there, that is roughly the time scale we operated on.

T. R. Thomas, Allied Chemical, INEL - The section on the separation and retention of iodine was co-authored by me and Dr. Yarbro of Oak Ridge National Laboratory. We treated scrubbing techniques and other technologies. In that category, we had caustic scrubbing, mercuric nitrate scrubbing, and the 22 boric nitric acid scrub which is called the iodox process. Of these three processes, caustic is essentially outdated and outmoded. There doesn't appear to be any further development or application of that technology. A mercuric nitrate scrubbing system is being installed at the Barnwell Nuclear Fuel Plant. They anticipate a decontamination factor above 100. However, they have no current plans for the waste other than storage. The mercuric nitrate scrub system will also be tested at the research center in Belgium. There, they anticipate getting more design criteria in a pilot plant study. In the adsorbent technologies, there is only one serious contender and that is the use of silver coated adsorbents. By this, I mean adsorbents such as silica gel, alumina, amorphorus silicic acid, and zeolites which have been exchanged with silver salts. Current plans are to use silver exchange zeolite as a backup bed at the Barnwell Nuclear Fuel Plants, i.e., as a polishing bed for the mercuric nitrate scrub system. They plan to do this also at the nuclear research center in Belgium on a pilot plant scale; again, as a polishing bed.

At the WAD plant in Karlsruhe, Germany, plans are to use silver impregnated amorphorus silicic acid subtrate as a bulk item removal system. They hope to obtain design criteria for a full scale processing plant handling up to 1,500 tons per year. The direct application of other metal-loaded adsorbents does not look too promising in that the loading capacities are extremely low. However, research is being done to use lead exchange zeolites as a storage adsorbent for iodine which has been desorbed from silver loaded adsorbents after they have been used as the primary adsorbent for recovery of processing offgas. The silver adsorbent will be regenerated and the lead exchange zeolite will be used as a final storage medium. I have also treated the subject of the abatement of oxides of nitrogen in the TAD report. There were three categories of technologies: (1) aqueous scrubbing, (2) catalytic reduction of NO_x with reducing agents such as hydrogen, carbon monoxide, propane, methane, and so forth, and finally (3) the very specific catalytic reduction of NO_x by ammonia over hydrogen mordenite. It is anticiapted that all of the nitric acid waste generated by a 1,500 ton per year plant will be converted to NO_x. This would be about 500 ton per year of NO_2.

L. L. Burger, Battelle-Pacific Northwest Laboratory - I'd like to acknowledge the contributions from Exxon Mound Laboratory and by Fieldson from Brookhaven and, of course, a lot of valuable help from Russ Brown.

Tritium is unique with respect to our problems here because quite a bit is produced outside the fuel itself. However, as far as the TAD document is concerned, we're interested in closing the fuel cycle, as you have heard. So we're only really concerned with the reprocessing part of tritium. As you know, tritium, for the most part, doesn't end up at the reprocessing plant. This morning, someone commented that the available technology is to release it and that's essentially what we have. It can be released to the atmosphere as vapor or, in certain cases, it can be released through streams or other bodies of water. This is not completely true since there are technologies which are at various stages of development. One of these is headend treatments following oxidation which isolates the tritium in a concentrated form instead of the very dilute form which comes out of the tailend of the Purex reprocessing plant. Another approach is isotopic enrichment and this is the second place for producing an unusual isotope. It's essentially the only way we can handle this radioactive material if we want to concentrate it from aquaeous or gas streams. There are a number of isotopic enrichment materials that were considered. I won't mention all of them now, but the three that were discussed to some extent in the document were (1) catalytic exchange, (2) electrolysis, including reversable electrolysis which takes advantage of the possibility of cascading electrolytic cells, and (3) a future device, if I can call it that, the application of lasers or, another description, selective molecular excitation to separate the tritium. Isotopic enrichment can be used simply to concentrate any type of stream or it can be applied to recycle within the processing plant.

Another approach that was considered in the TAD document was recycle of the aqueous streams. As far as storage is concerned, we have no available technology, but perhaps that's not quite true. There are techniques that are fairly well along in development, including the application of concrete and impregnated concrete for large volume storage.

The possible use of organic polymers to tie up tritium rather completely, or the use of metal hydrides, looks favorable and is also one of the choice recovery methods considered.

As far as engineering is concerned, tank storage was the only item discussed in the document. There is another way in which one can dispose of tritium; namely, deep well storage, which is not included in this part, but it does show up in other parts of the document.

C. M. Slansky, Allied Chemical - Carbon 14 has proven to be a sleeper. As you know, there was very little interest in this effluent until a few years ago. We're lucky, I think, with carbon 14 insofar as it comes out of several of the gas treatment systems that are fairly well developed. For instance, in the krypton cryogenic system in Idaho. It would come out quite rapidly in the caustic scrubbing and pump system. In the fluorocarbon system, it comes out, as you heard today, quite readily. Of course, the obvious disposal method is the release of carbon dioxide. The technique of atmospheric dispersion is pretty well developed.

I think it might be interesting to dwell a few minutes on final disposition and form. The trend, as you know, has been toward solid forms. I think we can dispose of the gaseous waste in storage cylinders for at least 5,000 years. Let's consider the solid phase. The most obvious is calcium carbonate. I think it's fairly stable and, in cannisters, could be stored quite readily. Quantitatively, it's a little difficult to give a definite number but the calcium carbonate from the carbon 14 content of one year's effluent from a 50 giga-watt process plant would be something like 1.6 kilograms. Let's mix this up with ten to a hundred per cent natural carbon dioxide. This would run you something between 16 and 160 kilograms of calcium carbonate per year. I think this is a reasonable quantity of a beta emitter which could be stored in a salt mine or geological depository.

J. D. Christian, Allied Chemical, INEL - When fission product wastes are evaporated or when they're solidified at a high temperature, a fraction volatilizes. During evaporation, up to 2 per cent can volatilize, but this can be easily suppressed by reducing agents such as nitrous acid. The quantity retained during volatilization processes depends on the specific process and the operating conditions. It can vary anywhere between two thousandths of a per cent up to 100 per cent in certain situations, depending on the process. Then, release to the atmosphere would be anywhere between a tenth of a curie and a hundred thousand curies of ruthenium-106 per day from a five ton per day plant which is processing one year of fuel. This assumes these gases will be condensed in the offgas system for other purposes and that the scrubber will remove 90 per cent and the condenser between 90 and 99 per cent of the ruthenium. Of the various high temperature waste treatment processes that are being developed for commercial wastes, fluid bed calcination and spray calcination offer the best potential for retaining the ruthenium as a solidified product. When fluid bed calcination is used with the bed being heated directly by the burning of kerosene, the volatility of ruthenium from the bed would be approximately 2000ths of a percent.

In a spray calcinator, using the in-pot melting process, approximately 2 per cent of the ruthenium normally volatilizes. If the salt is denitrated before the calcination, as is done in the German process, the volatility is reduced to 100th of a per cent. In other processes, ruthenium volatility would be substantially higher, one to ten per cent at best. Offgas cleaning devices that are effective to varying degrees for removing ruthenium from the gas stream are the (1) Venturi scrubber, which can provide a decontamination factor of approximately 10, (2) condensers that provide decontamination factors of 10-100, and (3) adsorbing materials. The silica gels and ferric oxide are the two commonly used ones and either will provide a DF of 10^3.

C. A. Burchsted, Oak Ridge National Laboratory - Essentially, what I talked about at Denver was dividing the subject of particulate removal from offgases into three categories; pretreatment, prefiltration, and absolute filtration. Absolute filtration, as has been widely discussed in this session during the last couple days, is the final barrier. The main object is to protect the final filter to enable it to remain effective under the most adverse circumstances. Therefore, both pretreatment and prefiltration are intended not only for particulate collection, but also to protect the last element in the chain. Pretreatment equipment includes cyclones, spray towers, electrostatic precipitators, bag filters, and similar devices. Most of the technology is well understood and well established and most of it represents the adaptation of more or less standard items to this particular application. Much of the development work of pretreatment devices is being undertaken by EPA. Prefiltration has also been talked about at this meeting, and includes the enhancement of particle collection by various means. This is the type of development that is ongoing in prefiltration. Otherwise, the prefilters in use today are generally conventional filters that are commonly used in everything from furnaces and air conditioners up to building ventilation. Two special areas of prefiltration I mentioned were deep bed glass filters seen in Hanford, and the deep bed sand filters that Don Orth talked about yesterday. Usually, offgases are characterized by high temperature, high humidity, low flow rates, and very corrosive environments. Pretreatment collection devices are often combined in an effort to get temperature and humidity down to levels that the absolute filters can handle. This is essentially what it is all about. Developments that we will see in the future include packing and reducing the volume of filters, enhancement of prefilters, and material improvements. Unfortunately, many types of pretreatment devices tend to convert a gaseous waste problem into a liquid waste problem. Therefore, the more we can look to dry methods of collection, the better off we will be. We generally reduce gaseous wastes to solid wastes eventually. This was covered in many of the talks during the last few days; the first day in particular. These included papers on incineration, compacters, etc.

DISCUSSION

RAMSEY: We are just a little bit ahead of our schedule, so I would like to take this opportunity for some questions and some comments from the floor. If you have questions that you want to ask me or Russ about the TAD document, you may do so. These would be welcome at this time. I will give you a chance to say anything that you think hasn't been said here that might be helpful to the overview of the status of air cleaning. If you have any questions that you always wanted to ask somebody from ERDA, I will try to answer them.

DEMPSEY: Being part of ERDA and sponsoring some of the possibilities that were mentioned, we are very keen on getting opinions whether they are ready for practical application. One that comes to mind is the excellent work we did and described here for solidification of krypton. The question that comes to mind is: was it worth the effort?

ANON: This is directed either to Burchsted or to someone from ERDA. In storage of plutonium, we have been faced with the problem of coming up with a suitable decontamination factor for plutonium particulates generated by a fire. It seems there is no standard decontamination factor applied across the country. Some of the ERDA offices have applied one factor and some another. I am wondering why we can't have a standard for HEPA filter decontamination or for multiple HEPA's in series.

BURCHSTED: We have been trying to come up with some reasonable numbers for this. There are two aspects to it. First, what is reasonable decontamination? And, second, what is a reasonable number you want to permit for safety analysis purposes under the postulated accident? Under normal operating conditions, considering the excellent filters we have today and the confidence we have in the quality assurance program, we can figure $3x10^3$ as the decontamination factor for each stage. This would be the multiplier. But when it comes to the accident condition, the first filter in the chain could very well be destroyed. So, the most I would allow would be a decontamination factor of $5x10^2$ as it appears that even when there are some gaping holes in the filter, it is an effective device for collecting materials as long as it is protected. This is what I was talking about a few minutes ago. We are trying to arrive at some numbers that would be reasonable for safety analysis reports.

In the Nuclear Air Cleaning Handbook, I discussed the subject but I didn't give any numbers for one, two, and three stages. I had started out to do it, but couldn't get enough agreement among the experts to stick my neck out and put the numbers in the book. But we will have an addendum to the book one of these days in which we will put that in.

RAMSEY: I think this is especially the type of question that will be very useful for us to discuss in the Symposium that takes place right after this meeting.