



19

UNITED STATES
NUCLEAR REGULATORY COMMISSION
ADVISORY COMMITTEE ON REACTOR SAFEGUARDS
WASHINGTON, D. C. 20555

March 17, 1981

Honorable Joseph M. Hendrie
Chairman
U. S. Nuclear Regulatory Commission
Washington, DC 20555

SUBJECT: COMMENTS ON FISSION PRODUCT BEHAVIOR DURING LWR ACCIDENTS

Dear Dr. Hendrie:

During its 251st meeting, March 1-14, 1981, the Advisory Committee on Reactor Safeguards met with members of the NRC Staff and its contractors to continue our review of the draft report, NUREG-0772 on the "Technical Bases for Estimating Fission Product Behavior During LWR Accidents," dated March 6, 1981. This was also the subject of a meeting of the ACRS Subcommittee on Reactor Radiological Effects on March 10 and 11, 1981. Earlier Committee comments on this effort were provided to Chairman Ahearne on February 11, 1981.

On the basis of these latest meetings, which included a review of the initial draft report being prepared under guidance of the NRC Staff, we offer the following comments:

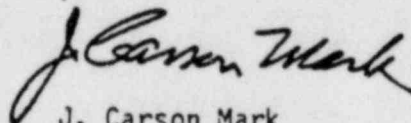
1. The NRC Staff and its contractors have prepared a comprehensive document in a short period of time. The report provides a good up-to-date summary of knowledge on potential fission product releases under a range of postulated accidents.
2. We believe the report does not contain data or information that would justify changing current regulatory criteria at this time. Although regulatory changes may ultimately prove to be warranted, they should be made only after the report has been completed and has been carefully evaluated.
3. While pointing out what is known, the report also identifies what is not known. As such, it represents a useful resource for planning future research on this subject. Such planning should include preparation of a list of research needs and a designation of the priority with which each should be addressed.
4. Specific research areas brought out by the report as requiring attention include the development of a better understanding of:

March 17, 1981

8. In reviewing plans for the development of the report, the Committee understood that the effort was to include a review and evaluation of past accident experience. We have noted, however, that the draft report does not include this information. Because of the benefits it might provide, we recommend that consideration be given to conducting such a review.

The Committee reiterates its view that development of the technical report has been a worthwhile effort. We believe, however, that issuance of the draft report, NUREG-0771 on the "Regulatory Impact of Nuclear Reactor Accident Source Term Assumptions," dated March 1981 should be delayed until the data developed in the technical report can be thoroughly reviewed and evaluated.

Sincerely,



J. Carson Mark
Chairman

- a. The impact on the behavior of the fission products of various thermal-hydraulic and material transport processes accompanying severe accident sequences.
 - b. The impact of intense radiation on the physical and chemical behavior of fission products within a post-accident environment as well as the effects of radioactive decay on the transport of radioactive fission products.
 - c. The effect on fission product behavior of the presence of hydrogen gas, boric acid and other chemicals. Also to be considered is the potential effect of a hydrogen deflagration on fission product behavior.
 - d. Other key factors governing the behavior of the significant fission products so that their movements and releases can be adequately predicted. This effort should not be confined to iodine and cesium.
5. Inasmuch as the use of computer code models (TRAP-MELT, CORRAL, MARCH, etc.) plays a major role in assessing the risks associated with various accidents, it is important that work be continued on improving such codes. This should include developing a better understanding of the soundness of the basic assumptions used in their preparation and in the identification of the range of uncertainties in the projections they produce. Independent review and evaluation of these computer models would also be warranted.
 6. Considerable attention has been directed to possible changes in our concepts of the chemistry of the source term fission products. Comparable attention, however, does not appear to have been directed to the influence of the chemical properties of the fission products on the performance of systems for their removal, on their behavior within the environment, or on their associated health impacts.
 7. The draft report contains a number of assumptions on the behavior of various fission products. In some cases, for example, steady states were assumed when dynamic situations will more probably exist. In many cases, extrapolations were made from the behavior of chemicals in macroconcentrations to that at trace levels; in other cases, data from bench scale experiments have been extrapolated to estimates of conditions in full scale plants. The Committee recommends that the final report include a summary of such assumptions and the associated uncertainties they introduce.

Comments on Technical Bases for Estimating Fission Product Behavior During LWR Accidents

General Comment: This is an extremely important document. It is imperative that it be correct, complete and unbiased.

Section 2.1

Different isotopes of an element are virtually indistinguishable chemically. (But there are subtle differences, and some isotope separation processes are based upon these differences.)

"...Fission results in the release of ... and usually two fission products." This sentence should read "... and at least two fission products."

"The problem with fission products is that they are radioactive (they are unstable)." This statement should read "The problem with fission products is that some of them are radioactive (those that are unstable)."

"The length of time .. before decaying ... is characterized by its half life." Should read "The average length of time a radioactive nuclide will remain before decaying to another nuclide is characterized by its half-life."

Correct

~~Correct~~ the sentence, "Through the emission of radiation, the nucleus decays ... etc."

Correct next sentence also. Is no decay energy absorbed by the coolant?

Correct the sentence, "Some are the direct products of fission, etc."

What is the meaning of "spectrum-averaged cross-sections?"

Section 2.2

Last two sentences of p. 2.5 state that the radioactivity cleanup system is the principal source of radioactive materials released to the environment during normal operation. This should be corrected. It's also wrong in the sense that activation products are the major radioactive species in the coolant in a normally operating reactor.

Section 2.3

Second paragraph, p. 2.6. "They would at worst be expected to occur only and within a few miles of the plant." This statement presupposes the validity of many conclusions which are being challenged by the scientific community. The statement indicates bias, not objectivity.

"Even at low levels of exposure," What is meant by low levels of exposure, 500 mR? The remaining sentences in this paragraph are ambiguous, at best.

Third paragraph, p. 2.6. Sentence one implies that other "isotopes" (more properly, the term is "nuclides") should be added to the listing in Table 2.1 of greatest hazard species.

Correct sentences 2 and 3 of this paragraph.

Fourth paragraph, p. 2.6. "Iodine-131 are the potential major contributors ... in a severe accident." Statement presupposes judgments under challenge and again reflects bias.

"They are also predicted" By whom? Shouldn't the fact that this is under study not be mentioned? Again, bias is evident.

P. 2.7. "In accidents where the engineered safety features radioactivity release. This was the case for the Three Mile Island Accident." These two sentences are incorrect.

The next sentence is poorly composed.

"A number of other fission products ... to impact human health in an accident." Assessed by whom? Bias is evident.

P. 2.8. Suggest last sentence to be rewritten: "Because of their long half-lives, these species, if their release in a severe accident can occur, can have a major influence on the predicted long term exposure to the population over a 10-50 year time span."

Chapter 3

First sentence reflects bias! "... that could result in the release ..."

P. 3.4. Letdown and purification system. This system is also used to remove activation products, which are usually the predominant radioactive species.

Emergency Core Cooling System. First sentence. Two questions: 1) release radioactivity where? 2) if to the primary circuit, doesn't the cladding integrity have to be lost first?

Reactor Containment Building. Remove bias in second sentence by stating "... to retain fission products if released to the building."

P. 3.14. I suggest: "In 1977, Brookhaven National Laboratory undertook a program basis envelope. In this study, realistic estimates, based upon the state of the technology at that time, were made ..."

Next paragraph, I suggest "Based upon the state of the technology at that time, the consequences of core meltdown sequences were calculated to be much larger than those of the Condition I to IV events."

P. 3.15, first full sentence on page. Correct the sentence as follows: "As a result, would rupture and release some fraction of the inventory ... in the interconnected voids in the fuel rods, most notably the gap between the fuel and the cladding." fuel

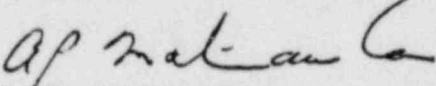
Section 3.3.2

Last paragraph, p. 3.15, last sentence. I suggest: "During the time period of fission product release in this accident, the pathway through the reactor coolant system to the containment is believed to be dry."

P. 3.16, Core Meltdown Sequences. Why is a distinction made between releases of radioactivity and releases of ~~aerosols~~ aerosols? Are not the aerosols radioactive?

Note also that the core meltdown sequences are hypothetical sequences. They may or may not be physically realizable.

P. 3.17, last sentence, paragraph 5. I suggest "... this sequence would be very high, and the consequences potentially severe."


A. P. Malinauskas
Chemical Development Section
Chemical Technology Division

Chapter 1

Section 1.1

First paragraph. Define "severe accidents." Individuals involved with nuclear safety have been concerned with the consequences of all accidents.

Third paragraph. Why does this report focus on accidents involving severe damage to the core and core meltdown (mainly core meltdown)? The core meltdown accident is an extremely low probability event ... emergency preparedness plans should obviously be formulated for less severe, but more likely, accidents ... it is these accidents for which an accurate knowledge of the chemical form of iodine is crucial to emergency preparedness planning. (See letter of Stratton et al. to Ahearne.) Also, Regulatory Guides 1.3 and 1.4 do not address core meltdown. (If so, they're not conservative enough!)

Third paragraph. Nowhere in this report are nucleation phenomena presented, yet it is claimed that "The generation ... of aerosols were also investigated in detail ..." Rather, it is assumed in this report that CsI forms an aerosol (but iodine would not), and this mythical aerosol is treated (and mythical conclusions drawn). This whole area needs to be re-examined carefully, as it makes a significant impact on the study.

Section 1.1.4

P. 6, Item 6. On the contrary, there is a wealth of information on reactor accidents. The authors of this document simply did not search the literature. Make up some other reason.

P. 7, third paragraph. But you did make a number of critical (and frequently unstated) assumptions which dictated what some of the results should be. E.g., see comments above regarding aerosol formation.

P. 9, last paragraph. Correct the sentence, "In water, the soluble fission products are ionized." Correct next two sentences also.

Section 1.3.1

My comments on Chapter 4 should be included in this summary. In particular, the demonstration that elemental iodine is not the dominant chemical form in the fuel should be explicitly stated.

Section 1.3.3, 1.3.4

See my comments on the correspondence Chapters.

~~9~~

P. 17, Contaminant sprays. The conclusions are dictated by assumptions concerning the physicochemical aspects of the system. These must be enumerated.

Pressure suppression pools. Conclusions are incorrect for both limited and severe core damage sequences.

Pressure suppression by ice. Same errors made as for pools.

Section 1.4.3

Too damn much emphasis on prototypical experiments and none on understanding underlying mechanisms. On what information and data bases is the code development effort to proceed? You're advocating more of the same kind of research as was done in the past. Where did this lead? Your own study (this report) indicates this to be the wrong approach.

Chapter 4

Section 4.1.1

^{134}Cs and ^{136}Cs are born by neutron capture involving ^{133}Cs and ^{135}Cs , not by (n, β) involving the corresponding short-lived β isotopes. Correct the next to last sentence in this Section also.

Section 4.1.2

This section espouses a considerably less than perfect understanding of the theory of solids and of migration in solids, as well as an understanding of the statistical nature of the laws of chemistry and physics. The fission product species in the UO_2 matrix is no more isolated than an individual uranium atom comprising the matrix. Suffice it to state that even after relatively short periods of burnup, there are on the order of 10^{16} atoms of a given fission product in each cubic centimeter of fuel. That's a pretty good statistical sample. Note also that the laws of radioactivity decay are not "obeyed" by a single, isolated nuclide. (How would you define a "half-life" of a single nuclide?) But that is not to say that the laws are invalid on a molecular basis.

Section 4.1.3

Sentence two is incorrect. "Iodine, having no chemical affinity for UO_2 ," This sentence is pure conjecture, and should be so identified.

Vaporization reactions (4.4)-(4.6). Are there no solid phases of CsI , I_2 , and Cs ?

Correct typos in this section.

P. 4.5, second paragraph. "In fresh LWR fuel the oxygen excess is such that (to end of paragraph)" References should be cited, else this must be regarded as conjecture.

Third paragraph. References should be cited.

Fourth paragraph. Cite references. Also note that, in Table 4.2, melting points and boiling points of gaseous species [note parentheses around CsI , Cs , and I_2] are listed. I suggest you remove the phase designations.

Fifth paragraph. In spite of the limitations listed by the author, he must realize that chemical thermodynamics is much more powerful than he implies. For example, in the closed system defined by the fuel and cladding, chemical species may not exist if this would result in a violation of the three laws of thermodynamics. Hence, reactions need not occur simply because this results in a decrease in free energy. However, in an isolated system, it is also valid that a reaction cannot occur if this results in an increase in free energy.

P. 4.7, Fifth results of study. I don't understand how a "higher uranate" (2 Cs atoms for 15 U atoms) ties up more cesium than a lower uranate (2 Cs atoms per 2 U atoms).

P. 4.8, first paragraph. Wouldn't higher temperatures favor equilibrium conditions because of the more favorable kinetics? Can you cite references for these broad statements?

Section 4 1.4

There are also observations of the thermomigration of Cs and iodine from in-pile experiments. Particularly significant are the observations of Wiedenbaum, Davies et al. Why were these excluded? They were cited by Campbell, Malinauskas, and Stratton.

Last line of p. 4.8. Cite reference for this observation.

P. 4.9, third paragraph. Doesn't the fact that the iodine, which was admitted in elemental form, deposited at 300-320°C zone ... well above its boiling point ... tell you something?

P. 4.10. "Evidence from thermomigration experiments is therefore also somewhat uncertain." I disagree. The evidence is rather conclusive that, even if you start with elemental iodine, you form an iodide if oxidizing conditions are not present.

P. 4.10, Fission product release-from-fuel experiments. "However, we would expect more rapid evolution of noble gases at lower temperatures because surface deposits would be removed in this range, which could not be distinguished experimentally from diffusional release." This doesn't make sense. Why is the daughter nuclide on a surface deposit when the precursor is not? If it's imbedded afterward, doesn't this mean that iodine was not in the gas phase, and therefore could not be imbedded? Why wasn't the xenon embedded in Parker's experiments?

Also note that Lorenz et al. experiments showed the gap inventories of Cs, I, Xe, and Kr to be equal. This means that the rates of release from the UO_2 matrix are equal, in further conflict with Parker's data. Further note that two other groups who worked with Parker on WASH-1400 did not agree with his interpretations. This should be cited.

Further, all three WASH-1400 groups agreed that iodine release was not equal to noble gas release ... they postulated the formation of zirconium iodide. Why wasn't this cited?

P. 4.11. In discussing Lorenz et al. results, you correctly point out reasons why I (or I_2) could form upon release of iodide from the fuel. What of the converse, i.e., how could a less volatile iodine species (an iodide other than HI or CH_3I) have formed in the test assuming I or I_2 was released? Doesn't this tell you something?

Why is no mention made in Section 4.1 of the iodine spiking phenomenon observed in PWRs? This phenomenon, also cited by Campbell, Malinauskas, and Stratton, likewise yields strong evidence that elemental iodine is not the dominant chemical form in LWR fuel.

Section 4.2.1

P. 4.13, second paragraph. One can arrive at no other conclusion but that iodine is not in elemental form in a LWR fuel rod from the statements made in this paragraph. Why doesn't this appear in Chapter 1 and Section 4.5?

p. 4.13, third paragraph. At this point in time, embedded gas is a concoction on the part of the author, and should be omitted.

P. 4.13, fifth paragraph. Grain boundary release is also unproved. Could this not be due to oxidation of the fuel by steam? "For a high burnup fuel rod, ~20% of the total initial fuel rod inventory ... would be released." Over what period of time? One minute? One day?

Something's missing between pp. 4.13 and 4.14. (It's on p. 4.16!)

P. 4.14. Last sentence of Tests of R. A. Lorenz should read "Occasionally ^{106}Ru and ^{125}Sb were detected."

P. 4.15. Complete last sentence of first paragraph.

P. 4.17, Other out-of-reactor tests. Mention should be made of the JAERI tests as well (see Campbell, Malinauskas, and Stratton paper).

P. 4.18. The release equation presented in Section 4.2.3 cannot describe burst release which is discussed earlier in this Chapter.

The model is much too artificial to be useful. Delete this section. What kind of release is being described? Certainly not burst release. Diffusional release? How does this model compare with the model of Lorenz, Collins, and Malinauskas, which accurately describes cesium and iodine release over six orders of magnitude?

I see no basis whatsoever for showing iodine release to be identical to Kr and Xe. (Nor do I know what specific mechanism is under consideration.) Again, I suggest this Section be removed.

P. 4.23, last paragraph. How much uncertainty is "considerable uncertainty"? One order of magnitude? Five orders of magnitude?

Section 4.4.1

P. 4.27. The author states "Therefore, reduced oxygen pressure (at high temperature) would certainly enhance release of Cs," He is obviously unaware of measurements made by Campbell.

Section 4.5

P. 4.30. In addition to being screwed up, paragraph one fails completely to address the fundamental issue ... whether elemental iodine is present in the fuel, or is a less volatile form involved. I have commented extensively on this.

Paragraph two is taken out of context. It too fails to address the fundamental issue.

Paragraph four. I saw nowhere any basis for the statement "The uncertainty of predicted release rates is estimated as plus or minus one order of magnitude."

I didn't think that the last paragraph was all that good to merit its repetition.

No mention was made of reactor accident experiences. Was nothing learned from these accidents? If the authors had bothered to really search out this information, they would find a wealth of information.

Appendix B

I have already commented on the utility (or lack thereof) of this approach. I only point out further that, in Figs. B.3 and B.4, I don't understand why Zr (F.P.) should be more volatile than Zr (clad), particularly since the former must contact the latter before being released. I expect the two to be identical and closer to U (fuel). Also, the results for Ru are inconsistent with the chemistry of Ru given in Chapter 5.

Chapter 5

Section 5.1, first paragraph. Listing of the iodine group as more volatile than the cesium group denotes bias. This should be reworded. First two sentences should be reworded also.

Correct second and third sentences of second paragraph.

What does sentence one of third paragraph mean?

Correct sentence four of third paragraph.

Section 5.2

Correct second sentence.

P. 5.2, first paragraph, last sentence. "... may react with oxygen if it is present."

P. 5.2, paragraph 4. Note that the arguments presented here are disputed in Chapter 4. The contradictions should be resolved. Next to last sentence ... what's magical about 600°C? Cite references.

Table 5.1. Mention should be made of the species $\text{TeO}(\text{OH})_2$ which could be important at high steam pressures (see publications of Glemser and of Malinauskas).

P. 5.3, last paragraph. Why not perform the sensitivity studies?

P. 5.4. Arguments for bounds on $1, K_{2,3}$ ratio are poor. If all the water in the primary system were converted to steam, the system pressure would be about 5000 bar! Clearly this is unreasonable. Why didn't the authors select the vapor pressure of CsI at the selected temperatures as a basis, for those temperatures where condensed phase CsI is present? After all, thermodynamic equilibrium is assumed in these arguments. I disagree with the last sentence on p. 5.4. Reasonable lower limit is perhaps about 10^{-5} .

P. 5.5, last paragraph. The statements here presume a particular mode of Te release and a particular accident scenario. This should be so qualified or the paragraph omitted.

P. 5.7. Note that TeO_2 boils at almost exactly the same temperature as CsI. Shouldn't the release behavior be nearly identical?

P. 5.9. Note that Ru chemistry paragraph contradicts the data presented in Figs. B.3 and B.4.

Section 5.2.2.7

Note that monatomic iodine may also react with metal surfaces (to further promote CsI decomposition?), but a necessarily more stable iodide must result. Also, if the temperature is sufficiently high, shouldn't the H₂O-metal reaction be considered?

Chapter 6

This chapter is best described as a state-of-the-art report of the TRAP-MELT code, and the results of sample runs. I see nowhere any provision for the formation of aerosol or the calculation of primary particle size distribution.

Documentation, particularly with regard to input data (other than the code reference for the thermalhydraulic data) is poor. What is the data base? Has any kind of experimental test of the code been made?

In view of the difficulties which were experienced with the simpler, well-tested, thermodynamics codes during this exercise, I seriously question the wisdom of citing any results of TRAP-MELT runs.

A more detailed description of the code, and a listing of all the underlying assumptions are in order. Also, references should be cited for all of the mass transfer equations employed.

All of these comments should be reflected in Chapter 1.

Chapter 7

Again, no discussion is made of aerosol formation. Contrary to what is stated, NAUA assumes an initial distribution of particle sizes; it does not calculate aerosol generation.

This chapter correctly points out that most of the validation studies have been conducted in dry atmospheres. To what extent is steam gravity and the condensing of steam onto vessel surfaces expected to enhance plateout (see p. 7.10)? Surely work has been done in this area!

Apparently no elemental iodine is allowed to deposit on the core melt aerosol in the containment building, whereas all the CsI so deposits.

The absurdity of the conclusions of this chapter, viz., that greater amounts of less volatile species will escape from the reactor than more volatile materials, merits a serious examination of all of the input, the code innards, and especially the assumptions employed.

Although some comparisons of the code results and experiment have been made with respect to aerosol behavior, there are no experimental tests of the overall processes, and particularly of the thermohydraulic conditions assumed (this is especially true of the conditions within the primary system). In other words, we're not sure that the "dry" accidents can actually occur at the time that CsI is being released!

Again, some of these points should be mentioned in Chapter 1.

Chapter 8

This chapter is simply premature as written. It relies heavily on highly suspect results of the preceding chapter. Chapter 8 should be written in a more objective manner.

A. P. Malinauskas
March 16, 1981

21

Comments on Iodine Report
March 16, 1981
D. O. Campbell

The abstract and Chapter 1 do not give the perspective required for interpreting the information. The slant of the whole report is toward the "risk dominant accident" or the very severe accident, with no indication of the relative probability of different accidents or of the net risks. For example, in the abstract question (2) about "postulated" accidents is answered by reference first to the risk dominant accident. The two sentences in answer (2) would be reversed in order.

Section 1.1, page 1, first sentence - expresses concern only with the "possible magnitude of the public consequences of severe accidents." This may be the past approach, but it would appear that this report should be presented in the context of overall risks, including the probability factor. However, this is not specifically included in this report.

Section 1.1.1, last sentence (also sentence 2, paragraph 2, Section 1.1) should be put in perspective, perhaps with a graph of probability versus consequences. Nowhere in this report is there anything about probability, and the report focuses so much on severe accidents that the casual reader may think this is the typical accident. The more likely accidents may not be very interesting because nothing much happens, but that is not a good reason to ignore them and emphasize severe accidents to the exclusion of any balance.

Section 1.1.3, page 5 does not present a perspective at all; it considers only the most severe case. See comments above.

Page 7, top - I disagree with conclusion. I think a great deal can be learned from prior accidents. It may be qualitative only, but it is extremely significant because things really happened that way; they aren't figments of man's imagination. For example, iodine from Windscale could be followed for hundreds of miles, whereas that from accidents with water around was very localized. Atmospheric dilution would more than compensate for the greater release at Windscale, so the iodine apparently really behaved differently.

A few comments of somewhat general nature are made on the Chapters.

Chapter 2 - There should be a table like Table 2.1 that gives the amounts of the fission products by element, in grams, as well as Curies. (This should be a true end-of-core value; some numbers that appear later seem to assume discharge fuel composition for all the fuel, although part is of much lower burnup.)

Chapter 3 - There seems to be unnecessary confusion about accident names. There is WASH-1400, then the "Conditions" I, II, etc. of Table 3.1, and the "Groups" in the Regulatory Report. Could everyone standardize on one set?

In Chapters 3, 6, and 7, especially, I am concerned that, throughout the accident analyses, the water that came out of the primary system seems to get lost. It apparently is assumed to be not present to interact with fission products. There has to be about 10^5 gallons of water somewhere in containment (even if containment fails, it will be there for quite a while), and there will be a lot of condensation from the very humid atmosphere, with water running down and covering a very large surface area. Is this taken into account? There seems to be an assumption that if ice melts or sprays don't work, there is no contact of released material with water; this just can't be so.

Vaporization of control rod material seems to be ignored; this could be very large for a PWR meltdown. Also, tin vaporization from cladding may not be properly included, and zirconium behaves differently from cladding than ~~the~~ from fission product. Why?

There have been lots of comments about the computer programs. The concern is that the individual cannot evaluate the suitability of the program for the problem being analyzed in many cases. It might help if a fairly detailed list could be presented giving the assumptions built into the programs, both explicit and implicit assumptions. This should include some discussion of just what these assumptions mean with respect to material behavior, if possible. It appears that some assumptions are very poor representations of nature.

There is talk in several places about a "TMI-like" accident, but why not put in the TMI accident. One might think someone doesn't really want to test the models against a real data point. NRC did an analysis based on prior methods, and as I recall the error band of the consequences was generally above the actual accident. Since this is a good experimental value, and not a model, it should be treated in this report. (Section 6.3.2.1.1, for example).

There are lots of examples of fuzzy wording that should be cleaned up editorially. However, statements like "factor of 2 on Fahrenheit scale" on p. 6-12, 6 lines up from the bottom, should be clarified.