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National Institute for Occupational Safety and Health

SC&A'S FOCUSED EVALUATION OF LLNL ORAUT-TKBS-0035-5, REVISION 03, FOR TBD-5 ISSUE RESOLUTION

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ABBREVIATIONS AND ACRONYMS

Advisory Board, ABRWH	Advisory Board on Radiation and Worker Health
Am	americium
C	carbon
Cm	curium
DOE	U.S. Department of Energy
DU	depleted uranium
ER	evaluation report
FAP	fission activation product
HTO	tritiated water
I	iodine
LANL	Los Alamos National Laboratory
LLNL	Lawrence Livermore National Laboratory
MDA	minimum detectable activity
MT	metal tritides
N	nitrogen
Na	sodium
NIOSH	National Institute for Occupational Safety and Health
Np	neptunium
NTS	Nevada Test Site
O	oxygen
OBT	organically bound tritium form
ORAU	Oak Ridge Associated Universities
ORAUT	Oak Ridge Associated Universities Team
OTIB	ORAUT technical information bulletin
P	phosphorus
PPE	personal protective equipment
PPG	Pacific Proving Grounds
Pu	plutonium
S	sulfur
SEC	Special Exposure Cohort

Sr	strontium
SRDB	Site Research Database
TBD	technical basis document
Th	thorium
TRU	transuranic
U	uranium
WB	whole body
WBC	whole-body count

1.0 INTRODUCTION

ORAUT-TKBS-0035-5, Revision 03, *Lawrence Livermore National Laboratory – Occupational Internal Dose*, dated August 12, 2016 (NIOSH 2016b) is a major revision of Lawrence Livermore National Laboratory's (LLNL's) technical basis document (TBD)-5 for internal dose assignments. It clearly addresses some of the concerns contained in SC&A's review of ORAUT-TKBS-0035-5, Revision 00 (NIOSH 2005a), associated with internal dose assignments; e.g., items pertinent to SC&A's Findings 1 through 8 (SC&A 2007). It also implements changes as a result of the current three LLNL Special Exposure Cohort (SEC) classes effective up through December 31, 1989. While SC&A participated in data capture activities in support of the National Institute for Occupational Safety and Health's (NIOSH's) development of its evaluation report (ER) for SEC-00221, the Advisory Board on Radiation and Worker Health (Advisory Board) has not tasked any reviews beyond the original 2007 site profile review for LLNL.

NIOSH's LLNL SEC-00221 ER (NIOSH 2016a) is focused on the lack of specific uranium-233 (U-233) bioassay data for workers in Building 251 up through December 31, 1989; however, for timeliness reasons, NIOSH "*will continue to review and evaluate internal and external exposures other than U-233 from 1974–1989, and all internal and external exposures from 1990–1995*" (NIOSH 2016a) for the main LLNL site and Site 300. On May 2, 2016, the Advisory Board agreed and recommended the SEC for this period, and it was approved and designated by the Secretary of Health and Human Services on June 17, 2016. Given that essentially all of the key remaining exposure potentials are reserved for further evaluation by NIOSH and to avoid unnecessary duplication of effort, SC&A directed its efforts at a focused review of the revised TBD-5, Revision 03, specific to potential internal exposures during the post-SEC period of 1990–1995 and how they are currently addressed in the TBD. Clearly, this review can be extended by the Work Group as additional research is accomplished by NIOSH and the current evaluation is supplemented.

2.0 SC&A'S EVALUATION OF TBD-5, REVISION 03, RELATIVE TO SC&A'S 2007 FINDINGS

Some of the areas of concern addressed in SC&A's original review of ORAUT-TKBS-0035-5, Revision 00, associated with internal dose assignments, have been addressed in TBD-5, Revision 03. These are related to Findings 1–8 (SC&A 2007), involving aspects of dose reconstruction for internal exposures, as follows.

Finding 1: Dose estimation for LLNL personnel assigned to weapons testing has not been adequately considered. *Exposure conditions related to LLNL personnel participation in weapons and safety testing, and subcritical or reactor experiments have not been considered in the LLNL site profile. This involves numerous LLNL-sponsored nuclear weapons tests, including atmospheric, underwater, and underground testing in the U.S. and at PPG* [Pacific Proving Grounds]. *Hundreds of personnel were involved in weapons testing and the Plowshare program. The significance and potential dose contribution due to LLNL personnel participation in testing has not been considered in the site profile, which is of particular concern for those test sites without existing TBDs (e.g., Amchitka, Hattiesburg, PPG, etc.). NTS* [Nevada Test Site] *eventually became the repository for the PPG and NTS dosimetry results; however, dose records*

or evidence that they have been requested is not available for all claimants. There is no apparent explanation provided for the benefit of the dose reconstructor on when and how doses from testing should be considered. Scientists and support personnel were responsible for re-entries to collect diagnostic equipment, cloud sampling after atmospheric tests, and processing of core and air filter samples. They worked side by side with workers in Alaska, at the NTS during atmospheric testing, and at PPG during underwater and atmospheric testing. NIOSH has identified problems with dose reconstruction for both NTS and PPG. The TBD does not provide background information and guidance on how to assess potential missed dose for exposure during weapons tests, subcriticality test shots and experiments, and the Plowshare program at testing sites across the United States. There is no information on how dose reconstruction issues previously identified at the testing sites (e.g., from past respective site profile and Special Exposure Cohort (SEC) reviews) will be addressed.

SC&A review: TBD-5, Revision 03, contains more specific background information and direction regarding weapons residues and LLNL post-testing assessments (e.g., pages 23, 24, and 90) but does not provide the dose reconstructor guidance about internal exposure of LLNL workers participating in LLNL-directed testing and research at NTS, Amchitka, PPG, Hattiesburg, and other offsite locations. Unlike weapons residue analyses, which took place at LLNL, these bioassays would have been conducted at these offsite locations. A similar question was raised during the Los Alamos National Laboratory (LANL) SEC proceedings, and NIOSH's position was that such dose records would be associated with those sites and would be reflected in an individual's dose reconstruction (including inclusion in the SEC if the 250-day rule were satisfied [ABRWH 2011]). However, it would be helpful to dose reconstructors to reflect this consideration in the TBD and to verify (e.g., through sampling or Advisory Board dose reconstruction reviews) that these records can be located at these various sites (or successor cognizant organizations, in the case of Hattiesburg).

Finding 2: Inadequate consideration has been given in the site profile to potential exposure received at Site 300. *The site profile is incomplete in its description of activities occurring at Site 300 and the potential radiological exposure conditions associated with these activities. Minimal dose reconstruction guidance is provided for internal and environmental occupational dose. The assumption of semi-annual bioassay monitoring is in conflict with information provided by former Site 300 employees and, in some cases, results available in dosimetry files and electronic dosimetry databases. The LLNL Site Profile indicates that the sources of radiation exposure at Site 300 include accelerators, DU* [depleted uranium], *activation products from accelerators, tritium, and radiography sources when in use. Batzel (*[1976]*) indicated that the guidelines allowed for experiments with natural uranium, DU, natural thorium, tritium, and beryllium. Sewell (1959) specifically authorized the thorium hydrodynamics program at Site 300 in 1959. No method for assessment of environmental dose from alpha emitters and tritium is available prior to 1961 and 1972, respectively. Potential extremity exposures may have occurred during hand contact with thorium and thorium alloy. An evaluation of non-hydroshot activities at Site 300 is minimally covered in the TBD, although workers were potentially exposed as a part of these activities. Further evaluation of the implemented monitoring for this area and its adequacy for the radionuclides involved in tests is necessary. Varying levels of personal protective equipment (PPE) were worn and bioassay was not routine, according to employees interviewed. In some cases, no protective clothing and/or respirators were used, and it is important to establish when this occurred. The work activities at the site appear to have had the*

potential for internal exposure, with some doubt that any bioassays were taken for monitoring such exposure.

SC&A review: TBD-5, Revision 03, contains more specific treatment of Site 300 in terms of the historic bioassay monitoring program but acknowledges that radionuclide-specific application to individual workers would depend on judgments of exposure potential and the nuclide involved (i.e., those other than airborne uranium would require a different schedule at the discretion of the health physicist). For natural thorium, TBD-5 (page 22) states that "*indications are that WB* [whole body] *counting was used to monitor for thorium intakes*" and that a "*worker's job description should be evaluated*" to determine if his recorded gross alpha bioassay analysis on top of regular uranium analyses actually was indicative of potential thorium exposure. Additional recent (2016) onsite research was conducted by NIOSH (in conjunction with SC&A) at Site 300 to better establish these and other questions, including better source term characterization and operational history; the results of this additional review should be reflected in the TBD.

Finding 3: Completeness, accuracy, and availability of data used in dose reconstruction, and as a basis for the internal coworker approach, not adequately addressed in the TBD. *Information available for dose reconstruction, especially for those involved in testing and special projects, is limited, inadequate, and sometimes not available. There are major issues with verifying the accuracy and usefulness of the data in MAPPER used for the coworker internal dose assessment method. Regarding the MAPPER database, LLNL staff members have indicated that some bioassays cannot be confidently associated with a specific person, and there are ambiguities in some analytes reported. These LLNL staff members indicated that large negative results are included for later periods, letters in the sample type column do not always indicate whether the sample was urine or fecal, and overall, that sample volume and mass must be interpreted carefully. There is very little discussion in the TBD about the quality of the earlier data (1950s–1960s). With the inconsistencies inherent in MAPPER, the use of these data for the internal dose coworker model is suspect and needs to be evaluated. During SC&A's review of classified documents, additional bioassay results were discovered that lead to questions regarding the adequacy of information currently being provided in the claimant files. These additional bioassay results found in classified records, not available for the dose reconstructor's use, could have an important effect on dose reconstruction of the individual claimant's dose. In light of these shortcomings, the verification process for determining the completeness and consistency of the internal dosimetry information provided in hard copy to dose reconstructors by the site needs to be addressed.*

SC&A Review: With the 2010 addition of Attachment B to the TBD in Revision 01 (NIOSH 2010) and carried forward with updates in Revision 03 in 2016 (NIOSH 2016b), a number of stated issues with MAPPER (incompleteness, accuracy, etc.) and its application in a coworker model have been largely addressed. For example, negative results were deleted for coworker data analyses (page 78). Early gross beta bioassay data (1957–1973) are not used (page 79). However, it is not clear from existing documentation whether an independent validation and verification of this database has been accomplished. There also remains the question of whether some bioassay data may continue to reside in classified files unavailable for dose reconstruction.

Finding 4: The Occupational Internal Dose TBD (*[NIOSH 2005a]*) has given inadequate consideration for the impact to worker dose from secondary radionuclides. *Numerous*

radionuclides were handled at LLNL, ranging from microcurie to curie quantities. These have included radium, Th-228, Th-232, Am-241, U-233, Cm-244, C-252, Pu-238, C-14, Na-22, P-32, S-35, I-125, I-131, Sr-90, N-13, and O-15, along with other fission products and activation products. Much of the bioassay data in the database are identified as "gross alpha" and "gross beta" results, and NIOSH has not identified which, if any, of these secondary radionuclides may be associated with these data. NIOSH has commented that the next revision of the Internal Dose TBD will contain guidance on the interpretation of gross alpha, gross beta, and fission product bioassay results. NIOSH needs to determine if there are potential exposures to these radionuclides that cannot be reconstructed accurately, due to inadequacies with the available radionuclide-specific information. These may be similar to the inadequacies cited in NIOSH's SEC evaluation report for Los Alamos National Laboratory (LANL) (NIOSH 2007[d]*). Radium is not identified (DOE 2004) as being used in any buildings or projects to a great extent; however, there is evidence that it may have been present at the site in some abundance. Exposure to a number of these radionuclides was not given adequate or, in some cases, any consideration in the internal dosimetry TBD, although some are listed as facility-specific radionuclides handled in particular technical areas.*

SC&A review: Major secondary radionuclides are now addressed in TBD-5, Revision 03, by the coworker model: americium-241 (Am-241), curium-244 (Cm-244), carbon-14 (C-14), phosphorous-32 (P-32), and strontium-90 (Sr-90), in addition to various fission products and activation products (page 91). Of course, U-233 figured as the basis for the most recent SEC class. In addition, a new section was added on "Other Limited-Exposure Radionuclides." In it, NIOSH acknowledges the presence of a wide scope of small-use radionuclides as part of the historic LLNL research program. It notes that the "*internal dose potentials due to these radionuclides (i.e., other than plutonium, uranium, tritium, metal tritides and organically-bound tritium) were remote because of the requirements for handling unsealed sources in fume hoods, hot cells, gloveboxes, and dryboxes*" (page 17). It is also noted that the whole-body counting program routinely scanned for a broad range of secondary radionuclides in additional to the primary sources, as evidenced by Table 5-9. Radium was present at LLNL and figured in at least one incident in 1975 (Table 5-12, page 33), but little about this operational source term is provided in the revised TBD-5. Additional guidance is provided for potential exposure to thorium-232 (Th-232), Cm-244, and neptunium-237 (Np-237) in this same section with reference to Table 5-11, which lists these radionuclides by work location. As acknowledged by NIOSH in its most recent SEC evaluation report (NIOSH 2016a), further onsite research remains to fully characterize these and other source terms of internal exposure significance.

Finding 5: There is limited guidance on the interpretation of bioassay data for intakes of tritium, metal tritides, or organically bound tritium. *While OTIB-0066 (*[NIOSH 2007a]*) was issued while the SC&A review was underway, and provided generic guidance on the calculation of dose from intakes of special tritium compounds, it only partially addresses some of the issues discussed below. According to the Site Description TBD (ORAUT-TKBS-0035-2)* [NIOSH 2005b], *Building 331 (Hydrogen/Tritium Research Facility) had the bulk of the tritium inventory in elemental form or metal hydrides. Metal hydrides of tritium are special chemical forms for tritium, and are also called metal tritides (MT). These MTs are somewhat insoluble forms of tritium compounds (Inkret et al. 1999, Cheng et al. 1997) that do not exhibit similar biokinetic behavior to the more common forms of tritium, such as tritiated water (HTO) or elemental tritium. Tritium from MTs does not enter the systemic compartment as quickly as HTO after*

inhalation and, therefore, the interpretation of tritium urine bioassay data cannot be treated with standard tritium excretion models (McConville and Woods 1995). Due to being relatively insoluble, inhaled MTs deliver the highest component of dose contribution to the lungs. Tritium from these particles also can convert to organically bound tritium forms (OBTs) from contact with lung tissue and further complicate the metabolic process (DOE 2004). OBTs were not discussed in the TBD. It has been determined that OBTs cause a significantly larger dose than tritium, more routinely found in the form of tritiated water (HTO) (DOE 2004). Not addressing MTs or OBTs could lead to underestimating doses. Bounding techniques proposed in OTIB-0066 ([NIOSH 2007a]*) cannot be effectively developed and applied without some basic understanding of the compounds handled and the extent to which individuals were exposed.*

SC&A review: These issues are now satisfactorily addressed in the revised TBD-5 (pages 15–17).

Finding 6: The Internal Dose TBD does not identify the possible chemical forms of the airborne radionuclides to which workers are exposed. *The TBD is lacking information that allows for the identification of the possible chemical forms of the airborne radionuclides to which workers could have been exposed. This is needed in order to give guidance on the solubility (absorption) class to use (F, M, or S) for inhalation and intake dose assessment. The Occupational Internal Dose TBD (*[NIOSH 2005a]*) states, "Other variables such as particle sizes and clearance classes can be readily reconstructed from historical records." No specific references are made to these historical records. There is no discussion on the potential for exposures to very insoluble and slowly absorbed high-fired plutonium. NIOSH has recently issued OTIB-0049 (*[NIOSH] *2007b), which provides some assistance to the dose reconstructor with respect to high-fired plutonium; however, the LLNL TBDs do not identify the potential for this existing at the site.*

SC&A Review: TBD-5, Revision 03, provides for the use of ORAUT-OTIB-0049 to assess Type SS plutonium and contains specific guidance in the form of Table B-11 to guide the dose reconstructor regarding percentile distribution intake rates on Types M and S solubility class plutonium for plutonium urine bioassay coworker data. It remains unclear as to the availability of solubility information on specific plutonium compounds in use at LLNL for dose reconstruction purposes.

Finding 7: The Internal Dose TBD has not adequately identified and reviewed applicable bioassay frequencies and detection levels. *In many cases, the information given for bioassay frequencies and detection levels is not useful, because of inaccuracy or lack of information. In the table showing bioassay frequencies, several in-vitro bioassays lack identification of radionuclides analyzed, and the frequency of whole-body counts (WBCs) for a period is missing. In addition, the table showing bioassay detection levels include values for in-vitro bioassay that disagree with historical site documents. Doses may not be calculated accurately without this information, and may not be claimant favorable. While the collective origins of these apparent discrepancies are not clear, more complete and validated information should be made available to dose reconstructors.*

SC&A review: Presentation of bioassay methods, frequencies, and detection levels (minimum detectable activities [MDAs]) was completely revamped in the revised TBD in Table 58 (in

vitro) and Table 59 (in vivo), which replaced the former Table 5-5 (bioassay detection levels) in Revision 00 to the internal dose TBD (NIOSH 2005a). The resulting two tables are more comprehensive and provide more detail, as well as new or revised MDAs. While SC&A did not validate each entry, the gaps and deficiencies noted in the original TBD (NIOSH 2005a) and SC&A's original eight issues specific to this finding (pages 48–49 of SC&A's 2007 review) are addressed satisfactorily.

Finding 8: No approaches are provided for determining the internal doses to workers that were unmonitored or inadequately monitored for plutonium, tritium, or other radionuclides. *The* Internal Dosimetry Coworker Data for Lawrence Livermore National Laboratory, *OTIB-0065 (ORAUT 2007*[c]*), provides an approach for determining internal dose only for uranium intakes by unmonitored or inadequately monitored workers, but does not address plutonium, tritium, or other radionuclides. This includes workers that were exposed to radionuclides prior to any bioassay monitoring (appears to be <1960) and those not monitored or inadequately monitored after applicable bioassay became available. If additional guidance is available from other sources, it is not referenced in the TBD.*

SC&A Review: Coworker data are now provided in Attachment B to TBD-5, Revision 03, for uranium, plutonium, americium, curium, and mixed fission products.

3.0 SC&A'S FOCUSED REVIEW OF THE REVISED LLNL TBD-5

3.1 RECORDED DATA ISSUES

SC&A performed a focused review of TBD-5, Revision 03, relative to potential exposures during the period 1990–1995.[1] From a review of both the revised TBD-5 and recent ER, some issues that complicate the use of the recorded bioassay data at LLNL are:

- Recorded gross alpha results could contain emissions from any of the many alpha-emitting radionuclides (e.g., plutonium, thorium, americium, curium, actinium, and neptunium) present at LLNL (Miller 1979, PDF page 43).
- Gross alpha analysis results (labeled "ALPHA") may have been recorded as just ALPHA.
- Plutonium analysis results may have been recorded as Pu-239, Pu, or ALPHA.
- Analyses for americium and curium of the eluate from the plutonium separation may have been recorded as Am-241 or ALPHA and contained both americium and curium.
- Generally, there were no specific radionuclide analyses for thorium, actinium, or neptunium.

[1] As noted in the introduction, NIOSH, in its ER for the most recent SEC petition, SEC-00221 (NIOSH 2016a), has reserved the period 1990–1995 for further research for all internal and external exposures, and for the previous petition period up through December 31, 1989, for all internal sources other than U-233. SC&A's review, here, is focused on any source term, dosimetry, operational, or records-adequacy issues that has not been addressed

- If americium analysis was performed, it could include curium in the sample counting results (NIOSH 2016b, page 75).

- Recorded gross beta results (labeled "BETA") could contain emissions from any of the many beta-emitting radionuclides present at LLNL from weapon-residue-related fixed activation products (FAPs) (through 1992) and/or aged reactor FAPs. Fresh reactor FAPs would have been present only during reactor operations, 1957–1980.

- The radionuclides in the weapon-residue samples varied considerably between the different NTS underground shots (NIOSH 2016b, page 24).

3.2 AREAS OF CONCERN

In reviewing TBD-5, Revision 03, SC&A identified the following areas of concern:

1. **Beta Source Term** – TBD-5, page 23, puts forward an assumption that gross beta bioassay results were only for weapons residue for the period up through 1992. However, bioassay results could have also been from workers exposed to aged FAPs and research radionuclides during this period. Has it been demonstrated that dose assignments from weapon-residue radionuclides are claimant favorable compared to aged FAPs and research radionuclides for all target organs of concern during this period?

2. **Weapons Residue** – TBD-5, page 24, discusses the beta-emitting radionuclides that could have been present in the weapons residue from NTS. It appears from this discussion that the radionuclide mixture in the residue varied widely, depending on the type of underground shot and the elapsed time after detonation. On page 25 of TBD-5, after NIOSH considered different tissue-seeking radionuclides, NIOSH recommends that ruthenium-103 (Ru-103) be used as the major radionuclide to determine potential chronic intake from urinalysis results, and that the cerium-141 (Ce-141) and Sr-89 intakes be assigned as 50% of the derived Ru-103 intake. Has it been demonstrated that this methodology (using only Ru-103, Ce-141, and Sr-89 in these ratios) is claimant favorable compared to the use of the other potential radionuclides in weapons residue, such as those listed on page 24 of TBD-5?

3. **Other Alpha Emitters** – Table 5-11, page 33, of TBD-5 lists recommended alpha-emitting radionuclides to be assigned if the worker's location indicates exposure to other than the primary radionuclides that were monitored. The dose reconstructor is to use the worker's bioassay results for a specific radionuclide (actinium-227 [Ac-227], Np-237, Cm-244, or Th-232) if available. However, if they are not available, the dose reconstructor is to consider using the following intakes:

    - Ac-227 = coworker Pu-239 intake
    - Np-237 = coworker Pu-239 intake
    - Cm-244 = coworker Am-241 intake
    - Th-232 = coworker uranium intake.

    SC&A has the following areas of concern, or areas that need clarification:

- If the worker has Pu-239, Am-241, or uranium bioassays in the files, shouldn't the unmonitored radionuclide (Ac-227, Np-237, Cm-244, or Th-232) be based on the worker's corresponding bioassay data instead of the coworker data?

- The recommendation to make the Ac-227, Np-237, Cm-244, or Th-232 intake equal to the associated indicating radionuclide (Pu-239, Am-241, or uranium) assumes that they were present in equal amounts. What is the basis for this assumption?

4. **Analyte Labels** – Attachment B (coworker data) of TBD-5, as well as communication with LLNL personnel (Mansfield 2006), discuss issues with the labels used to identify the results in the MAPPER electronic database system. For example, the term "gross alpha" could be used to mean results from analyzing gross alphas, Pu-239, Pu-238, Am-241, etc. Is this unique to the MAPPER electronic database, or does this issue also apply to the data in the NIOSH OCAS Claims Tracking System files that will be used for dose reconstruction purposes?

5. **Am-241 Coworker Data** – Section B.1, page 68, of TBD-5 states that "*The MAPPER database had small numbers of analyses for other radionuclides, such as ^{241}Am, curium, and TRU* [transuranic] *materials, but the numbers were too small for statistical analysis as an individual coworker dataset*." However, Table B-15, page 87, contains coworker bioassay data and Tables B-16 and B-17 contain recommended coworker Am-241 intake rates for the period 1957–1996. What was the source of these data if the MAPPER database contained too few Am-241 data entries to use for coworker data?

6. **GA-#** – Section 5.3.1, page 21, states that that "*Claims that have "GA-1", "GA-2", and "GA-3" bioassay results are to be assessed on a case by case basis. The dose reconstructor should contact the site lead for any claim that has "GA-1", "GA-2", and "GA-3" bioassay results for further guidance*." What additional information might be available to assist the dose reconstructor in dose reconstruction for these cases (e.g., what additional information should the dose reconstructor request and what department at LLNL should be contacted to ensure consistency in dose reconstruction cases)?

4.0 CONCLUSION

SC&A finds that the revised LLNL internal dose TBD, ORAUT-TKBS-0035-5, Revision 03 (NIOSH 2016b), is a substantial improvement over earlier versions and, as indicated above, resolves a number of SC&A's earlier findings about Revision 00 (NIOSH 2005a) of that TBD. However, aside from U-233 (the basis for the 2016 recommendation for an SEC for 1974–1989), other major radiological source terms with internal dose implications at LLNL (notably, high enriched uranium, thorium, neptunium, and metal tritides) remain to be fully evaluated and are reserved in the most recent ER (NIOSH 2016a) for further research. Therefore, SC&A awaits the results of that research before reaching any final conclusions about the adequacy of the current internal dose TBD-5, Revision 03.

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