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SUPERVISION OF THE RADIOACTIVE INVENTORY IN WASTE WATER PLANTS

Abstract: *Different technical variants of cooling plants for radioactive liquid waste (decontamination plants) for nuclear medicine facilities are presented. With the aid of modern measuring hardware and refined data analysis software, one can now guarantee nuclide-specific, gapless supervision and documentation of the activity inventory without any risk that personnel comes in contact with open radioactive or biologically critical material.*

Key words: decontamination plant, liquid waste, automatic supervision, software, I-131

INTRODUCTION

Facilities using or storing radioactive substances in quantities exceeding exempt levels are bound to supervise and document the inventory of radioactive nuclides and their activities without a break. Such supervision and documentation must be in compliance with federal law or regulations [e.g. 1,2]. This statement applies generally to all variations of application of radioactivity like in e.g.:

- nuclear power plants
- universities and research institutions
- production of radioactive samples
- technological use of radioactivity such as well logging
- industrial level or density or humidity monitoring
- nuclear medicine and radiology
- decontamination processes such as gas-washing in conventional power plants
- radioactivity control of exhaust gases, or to similar processes.

The key to all quantitative applications of radioactivity supervision is specialized software that provides

- supervision and control of system hardware facilities
- spectrum measurement and automatic high-precision analysis of spectra
- communication with external control- and documentation-systems

In this paper decontamination plants of nuclear medicine facilities, where radioactive substances are administered to patients for diagnostic or therapeutic purpose, are described as examples of waste water treatment systems. Administered activities are often too high to discharge the patient to the public after treatment; rather the person must remain in the hospital for some supervision time. During the supervision phase the patient's excretions – e.g. waste water from wash basins, showers and toilets - cannot be released to the public sewage system, but the water must rather be collected and stored until activities have decayed below regulated release limits. Such storage requires management of the inventory of radioactive waste water. The activity must be measured and chemical and/or biological treatment of waste water is necessary in regular time intervals. Requested control measurement and documentation must be made when the activity is low enough for release. Similar supervision may also be necessary for exhaust gases from patient rooms where gaseous radioactive substances have been released by breathing.

DECONTAMINATION PLANTS IN NUCLEAR MEDICINE

There are various kinds of decontamination plants in current use which originate from the historical development of radioactive waste water handling. Some countries provide a norm for contamination plants [e.g. Austria, 3]. In almost all systems, the most important measurement is quantification of gamma-rays from which is the most relevant nuclide in decontamination plants for nuclear medicine. Gross counting or energy-dispersive spectrometry are the methods of quantification.

Land-burial as method for disposal of radioactive waste [4] will not be considered in this paper.

There are very few systems that deal with waste water containing beta-active nuclides which emit essentially no gamma-rays at all. In these rare cases, one may use special beta counters or liquid scintillation counters. These cases are not considered in this presentation.

Tun systems

In very old facilities the waste water is collected into an open or covered container (a tun) typically having a volume of up to 10 m^3 (see Figure 1) The activity level of the inventory in the vessel is determined with an external counter when the tank is full. For this measurement the operator of the decontamination plant takes a sample from the tun, brings it to a shielded detector and after the assay pours the sample back. In most cases there is no nuclide specific control measurement but rather an assumption is made as to which nuclide is contained in the waste water. The activity of the assumed or identified nuclides is determined from the respective count-rate and the waiting time is calculated until the waste water activity is below regulatory level and the water can be released to the public sewage system.

Figure 1. *Tun system with tanks in a concrete bed*

In many countries such a priori calculation of decay time and release without final control measurements is no longer permitted. Moreover, open tank systems also require regular stirring of the water in order to avoid sedimentation of solids. Chemical neutralisation of the water before release is often mandatory. Such open handling of the waste water poses a threat to the personnel through potential contact with radioactive material which may also contain biological contamination.

The advantage of tun systems is its very cheap construction.

Disadvantages, however, are significant:

- There is normally no nuclide-specific information available
- The counter system used for measurement needs frequent re-calibration
- Integral activity counting is no longer permissible in modern legislation where nuclide specific measurement shortly before emission to the environment is mandatory
- The operator has to handle open waste water, i.e. biological and radiological contamination is possible
- Tun systems create stench
- There is no information on the status in the tun during the decay time
- The storage time is determined by the most long-lived nuclide.

Closed tank-stack system

In more advanced systems the waste water is collected in closed tanks where the water can be automatically treated (typically by mechanical stirring, airing, pH control and/or temperature control) in order to support biological decomposition during decay time. Several tanks are stacked and the supervising hardware controls filling states and other variables. Automatic selection of the next empty tank when the last one is full proceeds under software control. The tanks are either located in the basement of the hospital (Figure 2) or very big tanks can also be buried in the ground outside of the building.

Figure 2. *In-house tank-stack system*

There are two procedures of feeding waste water from the wards into closed tank-stack systems. The technologically simpler method feeds by gravity (which may need more shielding of the tubing) whereas other systems suck the water with differential pressure (vacuum) into a collector tank from where it is transferred into a storage tank. Vacuum technology needs smaller storage tanks but longer decay times, and its construction is more expensive. In all tank-stack systems it is an issue that the inflow of water should be kept at a minimum. Appropriate precautions must be taken at the patient level.

For radioactivity measurements a water sample is taken by the operator from the filled tank and nuclides are measured via γ-ray assay in an external spectrometer. Typical spectrometers consist of a 3”x3” NaI(Tl) detector inside a lead shield of 5 cm thickness, a 1024-channel multichannel analyser (MCA), a PC with appropriate software for measurement control and spectrum analysis, and a printer. An example of such a spectrometry system is shown in Figure 3.

Figure 3. *External γ-ray spectrometer: Lead castle with NaI(Tl) detector and MCA inside, laptop PC and printer*

The waiting time needed for the most long-lived nuclide in the waste water to decay below the federal activity release limit is calculated. In order to comply with the regulation that a nuclide-specific measurement of activities must be made shortly before release, another sample is taken and measured when the calculated decay time has elapsed. If the measured activity is actually below the release limit then the contents of the tank can be pumped out into the public sewage system.

There are significant simplifications and improvements of waste water treatment in sealed tanks; however, exposure can still happen with respect to biological and radiological contamination from sample taking into a Marinelli beaker until the sample is poured back into the tank.

The advantages of closed tank-stack systems are:

- The construction is relatively cheap
- The water is treated automatically in the tank
- Stench does not develop
- Modern software can analyse complex spectra and provide reliable results; nuclide activities are reliably determined, even at very low level
- The nuclide-specific spectrum analysis is in agreement with legislative demand.
- The disadvantages are:
- The operator has to handle open waste water, i.e. biological and radiological contamination is possible
- There is no information available on the activity status during decay time
- The storage time is determined by the most long-lived nuclide.

There are installations in which the contents of each tank can be pumped in a closed circle through a well-shielded measuring container having a volume of typically 10 to 20 litres in which a detector sits inside a submerged tube. The activity in the flowing water can be determined very precisely provided that one can control the water flow so that the generation of air bubbles in the measured volume is avoided. Such flow-through measuring systems require only one detector for supervision of all tanks. There are, however, two intrinsic problems associated with this type of activity measurement which prevent its use in most instances.

The tubing used for circular pumping from any tank must share some common volume, at least inside and near the measuring container, before separate tubing lines for each tank are selected. Therefore it is unavoidable that some of the radioactive water from the last measurement will flow into the next selected tank and contaminate it. This cross contamination is a problem as can be seen from a realistic example. Having measured a tank that contains only 1000 Bq/l of ^{131}I and then switching to another tank which is ready for release (5 Bq/l), the 50 litres of higher activity water from the shared volume will increase the activity in the other 10 m^3 tank to 10 Bq/l, which means that the water cannot be released, but another week of decay time is rather needed. One can prevent such cross contamination by a special flushing circuit where the activity from the common volume is added to the last tank.

A more annoying property of measurement in a flow-through volume is the generation of an organic "film" on the inner container walls which has the property of accumulating and concentrating ^{131}I from the water very fast (this film may well exist inside all tubing). The physical effect of this film is as follows: when biologically active water is pumped through the measuring volume the film will accumulate iodine to concentrations much higher than in the water. As the film sits on the walls there are two distinct sources of very high activity. The accumulation on the outer walls of the container is almost negligible as this activity is far away from the detector which sits in the submerged tube in the middle of the container. The activity accumulated on the wall of this tube, however, is very close to the detector and it is measured with high efficiency. Thus, when one pumps low-activity water through the volume, then the measured activity will gradually rise to higher and higher incorrect values, thus yielding too high apparent activity. The organic film can be chemically removed but it turns out that it re-accumulates very quickly.

In-tank measurement

The most advanced tank systems consist of stacked tanks as described above, but there is a 2"x2" NaI(Tl) or other detector mounted inside a submerged tube into the middle of each tank. Existing tank-stack systems can be modified to accommodate modern in-tank measurements.

Computer controlled switcher hardware is used that connects the high voltage supply for the photomultiplier and the analogue signal from the voltage divider output from each detector to the spectrometer. In this way one needs only one single spectrometer which is connected to any selected detector in a tank for measurement. When the distance between photomultiplier and multichannel analyser exceeds 10 meters one should use a preamplifier that drives the signal over the cable. Automatic supervision software performs measurements in all tanks in a cyclic sequence, thus providing continuous monitoring of activities (see below). For example, in an installation of 6 tanks cyclic sequences with 4 hours measuring time per spectrum will provide one data point every day for each tank.

In such a fully automatic stand-alone measurement, the ^{131}I activity in each tank is reliably determined down to below 1.0±0.5 Bq/l as can be seen in Figure 4.

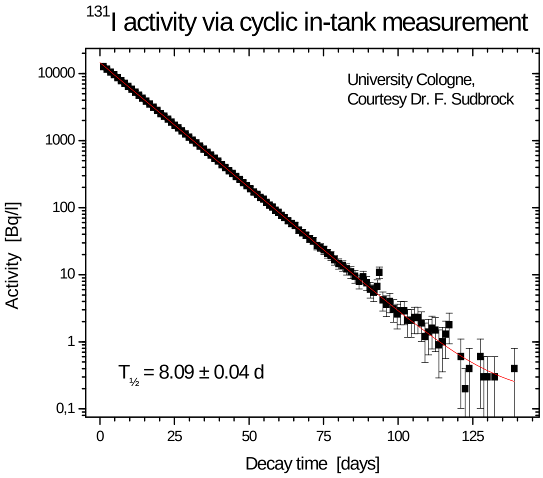

Figure 4. *^{131}I activity in a decay tank measured automatically in a cyclic sequence with 3 hours measuring time per tank. Note a significant ^{131}I contamination around day no. 93*

A decontamination plant with in-tank measurement is intrinsically safe for the responsible supervisor as no sample of open radioactive material needs to be taken at any time, and there is normally no human intervention needed.

Automatic in-tank measuring systems have clear advantages:

- Modern software can analyse complex spectra and provide valid results; even at very low levels activities are reliably determined in agreement with legislative demands
- Continuous control of the tank status during the decay time is guaranteed. There was a case in which that feature revealed a technical fault, where foam from a neighbouring tank had transferred radioactivity into a tank whose activity had almost reached release level. The foam was produced during aeration of the other tank because excessive amounts of detergents had been poured into a sink
- The most important advantage is that operators never have any contact with radioactive waste water
- As the high voltage for the photomultiplier and the signal from the detector are routed through appropriate hardware only one MCA for supervision of up to 64 tanks is needed.
- The disadvantages of in-tank measurement are:
- The additional cost of one detector in each tank
- The storage time is determined by the most long-lived nuclide.

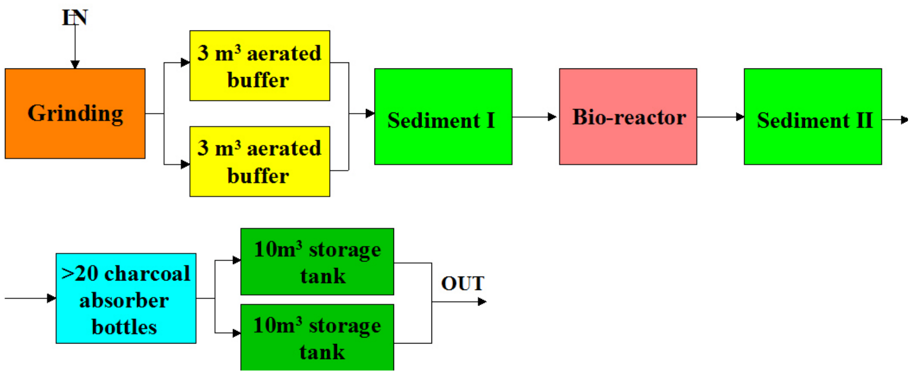

Figure 5. *Schematic of a Bio-Chroma decontamination plant for nuclear medicine and radiology*

Bio-Chroma system

Bio-Chroma systems are the latest development in radioactive waste water purification for nuclear facilities such as nuclear medicine and radiological hospitals. The idea behind this newly patented method is the adsorption of radioactive species onto activated charcoal and resin, where they completely decay in the bound state. In order to enable proper retention the waste water must first be “cleaned” very well from biological and other debris. A small but highly effective traditional sewage treatment plant (aerated buffer) is the first stage for biological decomposition, followed by a mud removal stage, digestion of organic material in a bio-reactor, sedimentation, and finally adsorption of radioactive (and other) species onto a combination of selected ion exchange resin and special activated charcoal. In the end the water is collected in storage tanks for final treatment and for measurement of the content of residual radioactivity. A schematic display of a Bio-Chroma decontamination plant is shown in Figure 5 and the view of a real plant in Figure 6.

Figure 6. *View of a Bio-Chroma plant*

The charcoal and resin absorber (the blue containers in Figure 6) must be exchanged whenever its absorption capacity is exceeded. As Bio-Chroma plants are quite recent there is no experience as to the duration before exchange is necessary. The first installed plant is still running with the original set of absorber bottles after approximately two years now. In case of activity breakthrough one will have to exchange the first set of 150 liters each of ion exchange resin and charcoal.

The advantages of Bio-Chroma plants are:

- Less than 50% of the floor space of a conventional stack plant is needed; costs are slightly lower than in-house tank-stacks
- Operation is flexible in terms of numbers of patients; +30% of excess water over the design level can be handled for up to 4 weeks
- Less components and equipment are required
- Operation is more reliable and fault-free (e.g. no risk of cross-contamination)
- Stench does not develop in Bio-Chroma plants
- Large safety-volumes for potentially leaking storage tanks are not needed
- Small openings in the building are sufficient for construction of the plant
- Existing tanks can be used as buffers or for storage
- Contact with radioactive or biologically active waste water is eliminated. The final control measurement before release is made on an activity-free and biologically clean sample
- Modern software can analyse complex spectra and provide valid results; activities are reliably determined, even at very low levels
- The nuclide-specific spectrum analysis is in agreement with legislative demand
- Since all radioactive atoms are retained in resin and charcoal, the storage time before release of the waste water is NOT determined by the most long-lived nuclide.

The disadvantage of Bio-Chroma plants is the fact that the technology is new and experience in operation and handling has to be accumulated.

SOFTWARE CONTROL

It is a well-known fact [5] that the key ingredient for safe waste water handling is the process-software which controls all operations such as spectrum measurement and analysis, data handling, status display and user alert. For this purpose we have developed a "nuclear medicine" package in the software SODIGAM for high-precision analysis of scintillator spectra.

When radioactivity levels in several locations are to be measured, or when the extraction of radioactive samples for measurement is prohibited or undesired, the installation of a remote detector system and in-situ data taking is an effective solution. In these systems one detector is mounted into each measuring site (for example into some tubing inside a sewage tank or in an exhaust line) and wired to the PC with special HV and signal cables. The MCA can be operated in selecting, sequential, or in a parallel mode, depending on the desired operation (see below). In addition one can use the system for on-line controls with reaction times of much less than 1 second per data point.

The selecting mode

The PC controls one MCA together with a special signal switcher which selects the desired detector according to definition by user or by program. The connected detector then measures a spectrum which is analyzed and nuclide activities are quantified with SODIGAM. This system is the most cost effective setup to measure spectra from several locations but using only one single MCA. The user controls the system and he selects one location at a time for measurement and spectrum analysis. SODIGAM can also carry out automatic batch-mode spectrum analyses with MCA restart thus allowing running of continuous measurements in multi-spectrum scaling mode over any desired time; all results are stored on disk for later use.

The sequential (cyclic) mode

The PC controls one MCA together with a special signal switcher. An option in the SODIGAM software sequentially routes at given time intervals data from one detector after the other to the MCA and measurements are made for a pre-determined time. Each spectrum is immediately analyzed and the results are provided to the user on the monitor and/or in the printout and in a backup control file. By use of extra 20 mA interfaces the actual activity levels can be transferred to a monitoring site (e.g. to the control room). The user can at any time interrupt the sequential mode for measurements in any other mode. On return to the sequential mode, the system will restore the results of the previous measurement and resume automatic cycling; thus the cyclic monitor information for all tanks is maintained.

Sequential measurement is the most frequently applied mode in decontamination plants of nuclear and radiological hospitals. Several tanks are equipped with one detector each and the actual inventory for each tank is continuously monitored and indicated. When the radioactivity in one tank is low enough for release the system can be operated in the selecting mode and a longer, very precise measurement of the releasable activity is made in order to confirm the limit value of e.g. <5 Bq/l with low uncertainty. During the pumping out of the low activity sewage, the outlet can be monitored in the fast on-line mode (see below) and the effluent is stopped if a tank is erroneously pumped out. After completion of release the system is set back to the sequential mode for continuous cyclic monitoring. Through interfacing between the spectrometry system and the control system for the plant, the switching from cyclic measurement to individual tank measurement or to the fast on-line control and then back to the cyclic mode can be initiated by the plant control software.

The parallel mode

The PC operates one separate MCA for every detector. Spectra from all detectors are measured simultaneously and in parallel with separate timing control in a synchronized mode or without any synchronization. Each spectrum is analyzed immediately after the measurement and the results are stored and provided to the user. By use of extra 20 mA interfaces or through network the actual radioactivity levels can be transferred to a distant monitoring site (e.g. control room).

Fast on-line control

Fast on-line control measurements can be made by software selection with any of the above hardware configurations. The MCA continuously measures the activity and the SODIGAM software reads and analyses the continuously measured spectrum every 30 ms or faster, depending on the speed of the PC. The resulting current activity value for one nuclide is calculated from difference and displayed on the screen; a logical (+5V) output signal or a low-voltage switching relay or a network connection to an external device is activated for control purposes when a user-defined limit value is exceeded. The measured activity profile is internally stored in a file in 1-second increments for documentation.

Fast on-line control measurement is often used during the release of decontaminated water, in order to make sure that the correct tank is actually pumped out (Figure 7). Erroneous and illegal release can thus be avoided.

Figure 7. *Fast on-line control measurement in the release line (flow through measurement)*

CONCLUSIONS

Software for all of the above supervision modes in all different kinds of decontamination plants has been developed allowing quantitative, nuclide-specific measurement shortly before release when activities are very low. The basis of all applications is SODIGAM software for high-precision analysis of NaI(Tl) or other scintillator spectra (e.g. $LaBr_3$, $CeBr_3$, CsI) with its various application-specific control and analysis packages. SODIGAM is the only program which considers the physically correct shape of peaks in scintillator spectra as well as the correct baseline under peaks. Using new programming strategies such as physics-oriented modelling of spectral characteristics, Fuzzy-Logic problem solving and repeated analyses the program allows quantification of peaks very much more correctly and reliably than other programs [6]. SODIGAM has embedded application options for quantitative analysis and operational control that is needed for the most efficient operation of a decontamination plant. For all practically relevant spectrometric applications in decontamination plants the high-precision spectrometry and control software SODIGAM provides adapted operating modes.

The same operating modes of SODIGAM described above for contamination plant applications can also be used for many other purposes. One can, for example, measure the same object with several detectors simultaneously. By use of special software options several individually measured spectra can be manipulated, like e.g. normalization of various energy scales onto one common scale so that spectra can be added or subtracted - maybe considering efficiency corrections individually for each detector - and the manipulated spectra are analyzed. The latter setup is particularly useful for applications in which several locations must be measured simultaneously and results are immediately compared, or where several detectors are used to assay one object. Typical applications are also in multi-detector whole-body counters or in systems for the nuclide detection and quantification in waste barrel scanners where sum-spectra from several detectors are processed for improved statistics.

The fast on-line control mode is also used to continuously monitor processes with fast varying activity gradients like in portal passages, control of flow in tubing or supervision in certain production lines.

There are technical and practical limitations to the application of the above measuring systems:

- The ability to resolve peak-multiplets and to identify peaks in spectra is limited by the detector used. In scintillation spectrometry one normally uses NaI(Tl) detectors, however, $LaBr_3$(Ce) or $CeBr_3$ crystals have significantly better resolution and even smaller peaks or closer multiplets can be resolved in spectra measured with these detectors
- The distance between detector and MCA should not exceed 100 meters; longer distances are possible with more expensive LAN-based MCAs
- The count-rate in a spectrometric scintillator measurement should not exceed 40000 counts per second as the photomultiplier cannot properly resolve single pulses when counting rates are very high. In applications where automatic and unsupervised radioactivity measurements are evaluated this limitation requires additional measures whereas in decontamination plants the count-rate limitation is irrelevant. High counting rates simply mean that more decay time is needed
- The frequency of measurements in the fast on-line control mode is technically limited to approximately 30 spectrum analyses per second. However, for

reason of counting statistics analyses are typically limited to 1 spectrum per second

Advantages of modern systems are:

- Contact of personnel with open radioactivity or infectious material is avoided
- There is continuous monitoring of the status in the tanks
- The spectrometer determines highly reliable nuclide-specific activities even shortly before release, in accordance with federal regulations
- On-line control of activities helps to detect erroneous conditions in the tanks
- Fast on-line activity measurement during pump-out prevents erroneous release
- The complete system status including filling level and current radioactivity in tanks can be shown on screen in the control-room. Figure 8 shows an example of display from plant-control software. Such system visualisation allows the operator to select from his office all components of the plant and to see their current status on screen, like for example:
  - Status of all valves, pumps and motors
  - Filling level of a tank in %
  - Current radioactivity level in Bq/l

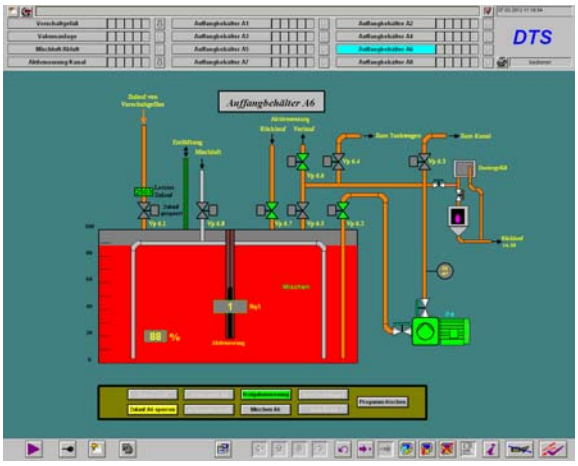

Figure 8. *Tank control screen in the supervisor room*

With the aid of visualisation software the operation of the whole plant can be remotely controlled through networking interfaces.

The key to such powerful applications in all fields of automatic radioactivity supervision is the very flexible software SODIGAM for high-precision spectrum analysis, system control and communication.

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BIOGRAPHY

Wolfram Westmeier was born in Kassel, Germany, in 1947. He received the diploma in Chemistry, and the Ph.D. degree in natural sciences from Department of Nuclear Chemistry, Faculty of Physical Chemistry at the University of Marburg.

His main areas of research include software strategies in gamma-ray and alpha particle spectrometry, applications of spectrometric methods, transmutation of long-lived nuclear waste, and compilation of evaluated nuclear decay data. He is currently working as CEO of his enterprise "Gesellschaft für Kernspektrometrie mbH" in Ebsdorfergrund (Germany).

MONITORING RADIOAKTIVNOSTI U POSTROJENJIMA ZA PRERADU OTPADNIH VODA

W. Westmeier

Rezime: *U radu su predstavljene različite tehničke varijante rashladnih postrojenja za tečni radioaktivni otpad (postrojenja za dekontaminaciju) za objekte nuklearne medicinske. Uz pomoć savremene merne opreme i softvera za sofisticiranu analizu podataka, sa pouzdanošću se može garantovati specifičnost radionuklida, kao i omogućiti monitoring dokumentacije o radioktivnim aktivnostima, bez ikakvog rizika da osoblje dođe u kontakt sa otvorenim radioaktivnim ili biološki kritičnim materijalom.*

Ključne reči: postrojenja za dekontaminaciju, tečni otpad, automatski nadzor, softver, I-131.