QUALITY ASSURANCE IN WASTE TREATMENT

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ABSTRACT

The licensing procedure for the final storage of radioactive waste in the former KONRAD ore mine has advanced far enough for preliminary repository storage conditions to be published for use as a basis in waste conditioning. These conditions contain a number of criteria with respect to the qualities of the container and the waste form as well as the kinds of conditioning and documentation to be met by the sources delivering waste packages and by conditioning operators. These criteria have a bearing on the conditioning processes, the equipment installed in conditioning plants and, especially, the way in which waste is treated, on the associated quality assurance measures, and on documentation. The report contains an outline of the measures taken at the Karlsruhe Nuclear Research Center to meet these criteria.

CRITERIA FOR FINAL STORAGE

Two projects on the final storage of radioactive waste are at present being pursued in the Federal Republic of Germany: KONRAD, a former ore mine to be expanded so as to accommodate waste packages with negligible heat generation, and GORLEBEN, a salt dome currently under examination for its suitability to accommodate any kind of waste packages (those generating heat and those which do not). Planning and safety analyses of planned final storage in the KONRAD mine have advanced far enough for preliminary repository storage conditions (1) to have been published, which are also constituent parts of the documents filed for the licensing procedure. The German Federal Minister for the Environment has decided that, pending completion of the licensing procedure, these criteria shall be used as a basis in producing repository type waste packages.

These preliminary criteria specify two classes of waste containers, six groups of waste forms, and fourteen characteristics of waste forms and waste containers, and also include guidelines for their descriptions:

(a) Waste container classes I and II are distinguished by quality. Class includes containers without specified leaktightness which, under an impact at a velocity of 4 m/s, must merely retain their integrity. Class II includes containers of a specified leaktightness, whose properties must be preserved even after the container has been dropped from a height of 5 m and subsequently exposed to a fire.

(b) Waste form groups 01 to 06 correspond to the different qualities of waste forms with respect to potential releases of radioactive substances. Classification must take equal account of the primary waste and the conditioning process (see Table I).

(c) The fourteen characteristics of waste forms and waste packages, namely:

activity	setting condition
inventory of nuclides	residual moisture
dose rate	thermal behavior
surface contamination	masses of parts of waste products
chemical composition	total mass of waste packages
quality of binder	quality of waste packagings
mixing dates	stackability of waste packages

These imply that criteria must be met as a function of the waste form group and the waste container class (see Table I).

One of the most important characteristics is the permissible inventory of radionuclides per waste package. This has been derived from safety analyses including normal operation of the repository, accident management in the repository, heat development and criticality. In a simplified manner, it is correct to say that the higher the quality of the containers or the waste forms, the higher will be the permissible nuclide inventory (see Fig. 1).

(d) The history of radioactive waste materials must be documented completely, with the data available from processing and analyses, through all stages of processing; residue, primary waste, intermediate product, waste form and waste package. Each waste package must be described by a data set when delivered to the repository (see Table II).

TRANSPORT CRITERIA

All waste packages must be taken to the repository on public roads and ways. They therefore must meet the appli-

TABLE I

Product Criteria for Various Waste Form Groups in Class 1 With Containers Without Specified Leaktightness.

	Product criteria	Waste form groups 01	02 Non-immobilized	02 Immobilized	02 Compacted	03	04 Dimensionally stable	04 Met. compact	05	06
Primary Waste	Random mixture of substances	o	o	o	o	-	o	-	-	-
Primary Waste	Metal with less than 0.1act.% of non-metal	o	o	o	o	x	o	o	-	o
Primary Waste	Metal with 0.1 to 1 act.% of non-metal	o	o	o	o	-	o	x	-	o
Primary Waste	Liquid or bulk material, respectively, in cement stone	o	o	o	o	-	-	-	x	-
Primary Waste	Scrap in cement stone	o	o	o	o	-	-	-	x	-
Primary Waste	Solids (Compressive strength > 10 N/mm^2)	o	o	o	-	o	-	-	-	x
Additions	No materials self - igniting below 70 °C	x	x	x	x	x	x	x	x	x
Additions	Fissile content up to 50 g per 100 l of product	x	x	x	x	x	x	x	x	x
Additions	Content of explosible substances up to 3 g per 200 lof product	x	x	x	x	-	x	-	x	x
Additions	Content of free liquids limited to 1 vol.%	x	x	x	x	-	x	-	x	-
Additions	Gases in empty vessels with max. 1.5 bar at 20°C	x	x	x	-	-	-	-	x	-
Additions	Content of burnable substances limited to 1%	o	x	o	o	-	x	x	x	-
Additions	Content of burnable substances more than 1%	o	-	o	o	-	-	-	-	-
Properties	Nuclide inventory limit	x	x	x	x	x	x	x	x	x
Properties	Product solid or solidified	x	x	x	x	x	x	x	x	x
Properties	Gas evolution up to permissible operating pressure	x	x	x	x	x	x	x	x	x
Properties	Compressive strength of product ≥ 10 N/mm^2	o	o	o	o	o	o	o	x	x
Fabrication	Ampoules, flasks, gas cylinders open	x	x	x	x	x	x	x	x	-
Fabrication	Voids in waste container filled up	x	x	x	x	x	x	x	x	x
Fabrication	Waste/immobilization agent ratio < 1.5	o	-	x	-	-	-	-	o	-
Fabrication	Cartridge compacted at ≥ 30 MPa	o	o	o	x	o	-	o	-	-
Fabrication	Cartridge (wall thickness ≥ 0.75 mm) compacted at ≥ 30 MPa	o	-	-	o	o	x	o	-	-
Fabrication	Metallic product prepared at ≥ 30 MPa	-	-	-	-	-	-	x	-	-
Fabrication	Radioactive substances evenly distributed throughout product	o	o	o	o	o	o	o	x	o
Fabrication	In case of Rn-220 release, product enclosed all around	x	x	x	x	x	x	x	x	x

x = required — = irrelevant o = permitted

TABLE II

Waste Data Sheet

1	Waste data sheet for package No.:
2	Waste delivered by:
3	Address:
4	Waste conditioned by:
5	Address:
6	Conditioning technique:
7	Certificate No. of process qualification:
8	Date of conditioning:
9	Type of primary waste:
10	Immobilization agent:
11	Waste packaging:
12	Canister:
13	Additional inner liner (material, thickness):
14	Annual permeability factor (waste package):
15	Water content and residual moisture, respectively (%):
16	Weight of fissile materials (g):
17	U-235 enrichment (< 5%; > 5%):
18	Fraction of burnable substances with melting points below 300°C (%):
19	Compaction pressure (MPa):
20	Compressive strength (N/mm^2):
21	Volume fraction of cement stone/concrete matrix (%):
22	Waste form group:
23	Waste class:
24	Total activity of alpha-emitters per package (Bq):
25	Total activity of beta/gamma-emitters per package (Bq):
26	Reference data of activity data:
27	Activity data were measured: estimated: derived:
28	Radionuclide spectrum code:
29	Radionuclide-specific activity per package, alpha-emitters (Bq):
30	Radionuclide-specific activity per package, beta/gamma-emitters (Bq):
31	Guaranteed level exceeded (normal operation) (Y/N):
32	Sum factor of accident S_A:
33	Sum factor of thermal influence S_H:
34	Local dose rate, surface (Sv/h):
35	Local dose rate at 1 m distance (Sv/h):
36	Local dose rate at 2 m distance (Sv/h):
37	Neutron dose rate (Sv/h):
38	Permissible limits of surface contamination by alpha-emitters observed (Y/N):
39	Permissible limits of surface contamination by beta/gamma-emitters observed (Y/N):
40	Weight of package (Mg):
41	Packaging factor:
42	Product surveillance completed (Y/N):
43	Clearance of waste package by BfS (Y/N):
44	Clearance date:
45	Signed by Radiation Protection Officer (waste deliverer):

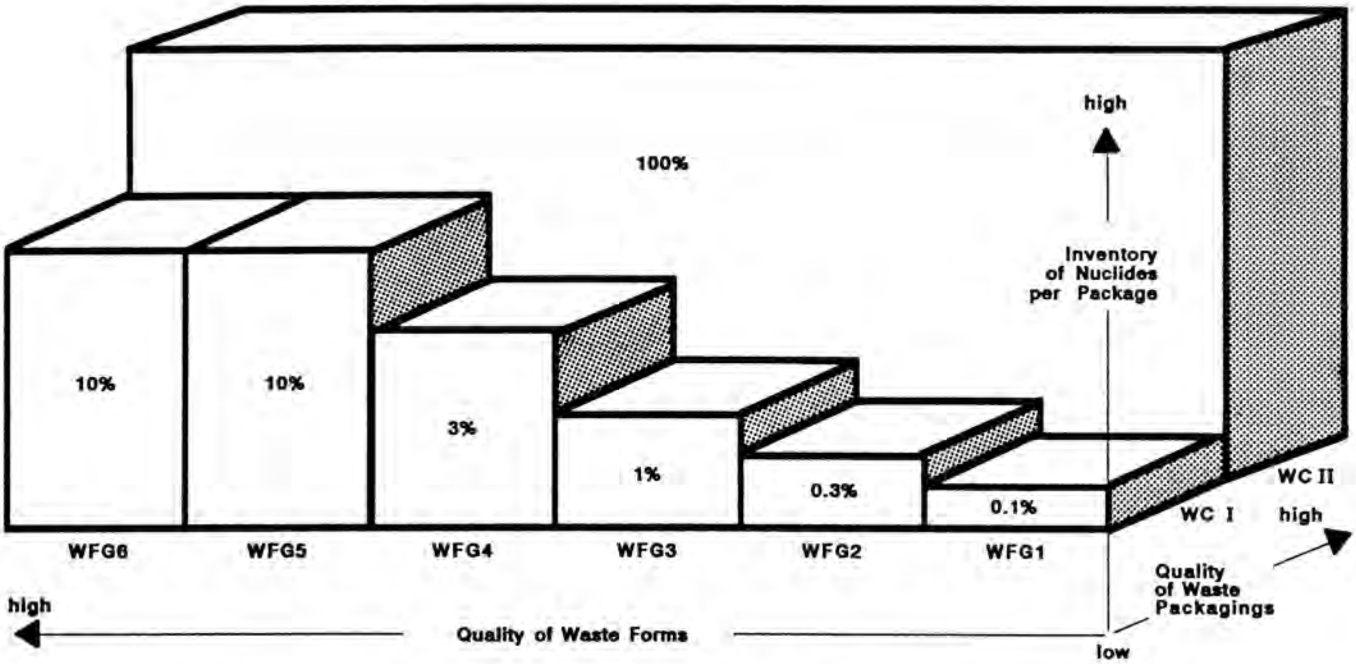


Fig. 1. Interdependence of the Waste Form Groups, Waste Classes and Nuclide Inventory.

cable codes and regulations, namely the Ordinances on Transports of Hazardous Goods on the Road and by Rail, respectively (2). These national codes are based on the IAEA transport regulations, as far as shipments of radioactive substances are concerned. The waste packages meeting the preliminary final storage conditions for the KONRAD mine belong in the LSA, LLS or, in special cases, also Type B transport categories.

PUTTING CONDITIONS AND CRITERIA INTO PRACTICE

The preliminary final storage conditions and transport codes and regulations can be applied only if criteria have been elaborated for manufacturing waste containers and making and packaging waste forms.

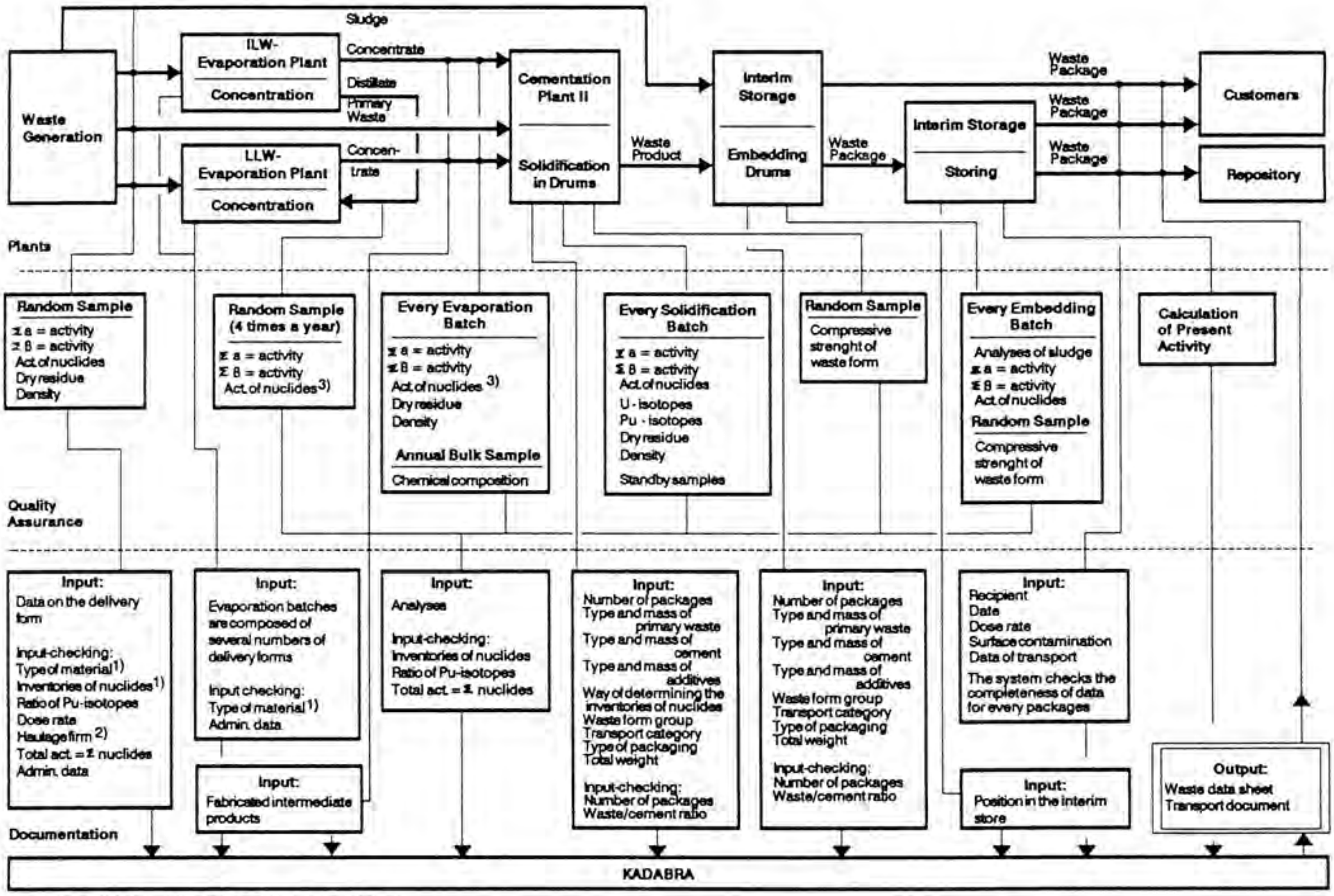
For the first step, so-called quality manuals are compiled, which contain all criteria for the choice of materials, for manufacturing and testing containers and for documentation, and also feature a description of the entire quality assurance organization. These manuals constitute the basis of type approval, production and acceptance tests.

The way in which waste forms are produced is described in so-called conditioning manuals. They provide information about the properties of primary wastes, intermediate products, and waste forms; they also include descriptions of conditioning techniques and conditioning plants, and definitions of quality assurance measures, such as process instrumentation and laboratory checks, and of the documentation of flows of materials and their conditioning. The different steps are interconnected and presented in clear schematic diagrams (see Fig. 2).

ORGANIZATION OF RESPONSIBILITIES AT KfK

The residues delivered for waste management must first be inspected for potential reuse. The processing installations and the General Safety Department cooperate in this check. Where possible, the material is cleared by the Radiation Protection Officer of the Central Decontamination Operations Department (HDB). If the preconditions for reuse are not met, the residues become primary waste to be disposed of in an orderly fashion. To meet the criteria applicable to the disposal of radioactive primary waste, i.e., in the generation of waste packages fit for final storage, the different activities to be carried out are assigned to different administrative units (see Fig. 3). This makes for effective in-house inspection.

- Conditioning: The preliminary treatment and completion of waste packages as well as their intermediate storage are responsibilities of two departments; one of which is responsible for liquid, the other for solid primary wastes, and which operate the installations listed in Table III.
- Quality assurance: Analytical data (operations or product analyses) and formulae, especially those for solidification, are elaborated by a working group independent of the operating departments. This group,



1) according to HDB-instructions for use and delivery conditions
2) according to the dataset of haulage firms
3) fuel nuclides are calculated

Fig. 2. Quality Assurance in Processing Liquid Non-Burnable LLW and LLW.

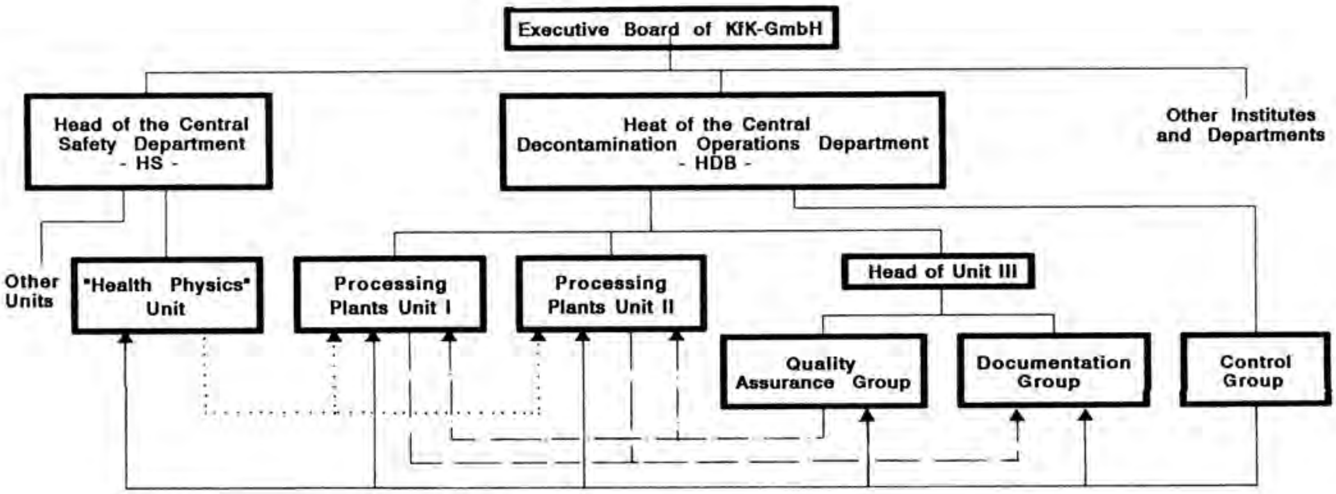


Fig. 3. Organization of In-House Inspection.

TABLE III

Plants of HDB

Plant	Throughput	Capacity	Date of commissioning
LLW evaporation	6,000 m^3/a	2x4 m^3/h	1968
ILW evaporation	500 m^3/a	1 m^3/h	1976
Kerosene purification	20 m^3/a	0.5 m^3/h	1976
Cementation I	2,000 drum/a	3 drum/h	1977
Cementation II	2,000 drum/a	3 drum/h	1986
Incineration (solids)	200 Mg/a	50 kg/h	1971
Incineration (solids)	200 Mg/a	50 kg/h	1989
Incineration (liquids)	40 Mg/a	30 kg/h	1986
LLW scrapping	3,000 m^3/a	-	1984
ILW scrapping	50 m^3/a	-	1971
Decontamination	700 m^3/a	-	1968
Interim storage:			
- Residue hall	-	8,000 drums	1968
- Component hall	-	2,000 m^3	1976
- Drum hall	-	39,000 drums	1978
- Container hall	-	5,152 containers	1984
- Drum vault I	-	1,400 drums	1971
- Drum vault II	-	2,100 drums	1988

for waste packages with negligible heat generation
for waste packages generating heat

in addition, advises and supports the conditioning plants in their activities.

- Documentation: A separate working group provides the hardware and software for the central bookkeeping system, updates the programs and develops them further in line with current requirements.
- Radiation protection: Work station monitoring and clearance measurements are responsibilities of the Central Safety Department.
- In-house inspection: The large number of criteria to be met, the volume of declared or measured data, and the many possible variations in conditioning could easily give rise to non-specification products or to products not matching descriptions. An independent in-house inspection unit verifies the data and orders those corrections to be made that may be required.

QUALITY ASSURANCE MEASURES

One of the most important characteristics is the inventory of radionuclides contained in the waste form and in the waste packages, respectively. Nuclides must be determined by different methods of technical measurement, which require the following distinctions to be made:

(a) Easily measurable nuclides constituting a major fraction of the inventory, such as Co-60, Zr-95, Ru-106, Sb-125, Cs-134, Cs-137, and Am-241.

(b) Nuclides also constituting a major fraction of the inventory, which must be isolated before being measured, such as H-3, Sr-90, U-isotopes and Pu-isotopes.

(c) Nuclides constituting an insignificant fraction of the inventory, but relevant to the safety of the repository, such as Be-10, Cl-36, Ni-63, Rb-87, Te-99, Sn-126, Pb-210, Ra-226, Ac-227, Ra-228, Th-230, Pa-231, Th-232, Cm-245, Cm-247, Cm-248, Np-237 and Am-242.

The nuclides in the first group are determined by gamma-spectroscopy, while U- and Pu-isotopes are measured by means of mass spectroscopy. Inventories of the nuclides of the third group are very small in the primary waste processed at KfK (see Table IV). These inventories

TABLE IV

Repository Tracer Nuclides With Reference Nuclides and Weighting Factors (Examples Taken From the Practice of KfK-GmbH).

Nuclide	Reference nuclide	Factor
Be - 10	Cs - 137	6.3 E-13
Fe - 55	Cs - 137	0.03
Ni - 63	Cs - 137	4.3 E-3
Rb - 87	Cs - 137	1.9 E-10
Nb - 94	Cs - 137	1.4 E-9
Tc - 99	Cs - 137	7.4 E-4
Ag - 108m	Cs - 137	5.9 E-4
Sn - 126	Cs - 137	5.9 E-5
I - 129	Cs - 137	9.6 E-7
Pb - 210	Cs - 137	8.1 E-16
Ra - 226	Am - 241	6.5 E-13
Ac - 227	U - 235	5.8 E-6
Ra - 228	Cs - 137	2.3 E-16
Th - 230	Am - 141	7.1 E-10
Pa - 231	U - 235	4.1 E-5
Th - 232	Pu - 239	3.2 E-13
U - 233	U - 238	8.1 E-5
U - 234	Pu - 242	0.15
Np - 237	Cs - 137	1.8 E-5
Am - 242m	Am - 241	0.01
Pu - 244	Pu - 242	1.8 E-13
Cm - 245	Pu - 242	0.46
Cm - 247	Pu - 242	7.8 E-7
Cm - 249	Pu - 242	3.5 E-6
Cm - 249	Pu - 242	3.5 E-6

are calculated on the basis of data determined by blended annual samples and specific techniques.

Prior to conditioning, the minimum number of packages must be determined which must be produced from an existing volume of primary waste with its nuclide inventory so that all limits pertaining to final storage and transport are met (1438 individual limits). This question can be answered by specific application of the summation formula:

$$n \in \frac{A_{(i)}}{L_{(O,i)}} \quad n \sum_i \frac{A_{(i)}}{L_{(A,H,C,i,g,c,T,D)}}$$

where

n	number of waste packages,
$A_{(i)}$	primary waste activity inventory,
L	limit value,
O,A,H,C	repository safety criteria,
i,g,c	nuclide, waste form group, waste container class,
T	Ordinance on Transports of Hazardous Goods,
D	dose rate.

Manual application is possible, but time-consuming. Making use of a data processing program offers the advantage of the results being available immediately for all possible waste form groups.

The nuclide inventories of the waste packages are calculated in each case for homogeneous and quasi-homogeneous primary waste, by distribution,

$$I_{(n,i)} = \frac{A_{(i)} W_{(n)}}{\sum_n W_{(n)}} \quad ; \quad I_{(n,i)} = \frac{A_{(i)} D_{(n)}}{\sum_n D_{(n)}}$$

where

I(n,i)	inventory of radionuclide i in waste package n,
A(i)	primary waste activity inventory,
W(n)	weight of waste material in the waste product in package n,
W(n)	weight of waste material packed in n waste packages of one batch,
D(n)	dose rate of package n measured in a defined geometry,
D(n)	dose rate of packages produced in one batch.

For inhomogeneous primary waste, they are obtained by calculation with dose rates previously determined and a well-known nuclide composition which may have been derived from operational data, calculated by correlation or measured from samples in the laboratory:

$$I_{(n,i)} = F_{(i)} (C_1 W_{(n)} + C_2) \sqrt[m]{D_{(n,1)} \cdots D_{(n,m)}}$$

where

$I_{(n,i)}$	inventory of radionuclide i in waste package n,
$F_{(i)}$	factor describing the part of nuclide i of the total activity,
C_1	factor describing the gradient of the function,
$W_{(n)}$	total weight of package n,
C_2	factor describing the part of the ordinate of the function,
$D_{(n)}$	dose rates of package n measured in defined geometry at m points of the drum circumference.

While the distribution of nuclide inventories by weights of waste or dose rates of packages is trivial, a number of conditions must be met in calculating nuclide inventories. The nuclide composition, $F_{(i)}$, must be known. Then it is used to do a shielding calculation (we use PROMAX [3]) for various weights, W. The result is a straight line, $I = f(D)$, by means of which C_1 and C_2 are determined. To compensate for the inhomogeneous distribution of the activity inventory, a mean value must be formed of the dose rates, D, measured over the circumference of a drum, most appropriately the geometric mean over all measured values.

PHYSICAL PROPERTIES

Before the waste form is classified in one of groups 01 to 06, certain physical properties must be met. As listed in Table I, they include:

(a) Open ampoules, flasks and gas cylinders,

(b) Hollow volumes filled up in the waste container,

(c) Solid or solidified form,

(d) Gas evolution up to the permissible operating pressure of the container,

(e) Radioactive substances evenly distributed in the form,

(f) In case of compaction, compaction pressure >30 MPa,

(g) Compressive strength of waste forms 05 and 06 >10 N/mm^2.

Meeting criteria (a) to (f) is ensured, on the one-hand, by the in-plant preliminary treatment of primary waste before it is conditioned (a,d) and, on the other hand, by the conditioning step proper as agreed upon with the authorities (b,c,e,f). The compressive strength (g) is determined in the analytical laboratory on the basis of a specimen produced.

CHEMICAL PROPERTIES

In analogy with the physical properties also chemical properties must be met for classification in a certain group, which include the following items (sees Table I):

(h) No substances igniting spontaneously below 70°C,

(i) Fissile material content up to 50 g/100 l of product,

(k) Content of explosive substances up to 3 g/200 l of product,

(l) Content of free liquids limited to 1 vol.%,

(m) Gases in empty vessels at max. 1.5 bar at 20 °C,

(n) Content of burnable substances limited to 1 act.%.

Meeting criteria (1), (m), and (n) in solid primary waste is ensured in-plant by preliminary treatment, e.g., emptying, rinsing and sorting out, respectively, while criteria (h), (i), and (k) are met within the analytical examination of the primary waste prior to conditioning (determining the concentration; determining the content of dry residue in liquid primary waste; conducting the furnace test of solid primary waste).

DOCUMENTATION

The condition stipulates that radioactive waste packages must be described by a dataset as shown in Table II and that their production must be documented over all stages of storage and processing back to the source.

For this purpose, KfK has developed the KADABRA bookkeeping system (4) (see Fig. 4). This system meets those conditions and supports the preliminary treatment, treatment, and packaging of radioactive residues, which must be filled in by the polluter, are attached to the residues and whose data are entered into the bookkeeping system. After receipt, the storage data are added. At the same time, administrative data are collected (client, owner of material, criteria for handling and packaging). Now batches are made up in accordance with the types of processing, again with the system assisting the operator. (May material from various sources be blended? May the product be alpha-contaminated? Observe limits. Run blending calculations, etc.) After such preliminary treatments as evaporation or incineration, the intermediate products generated are analyzed. Also these data are entered and are used to replace the data determined by bookkeeping for a certain intermediate product. The system provides assistance in decision-

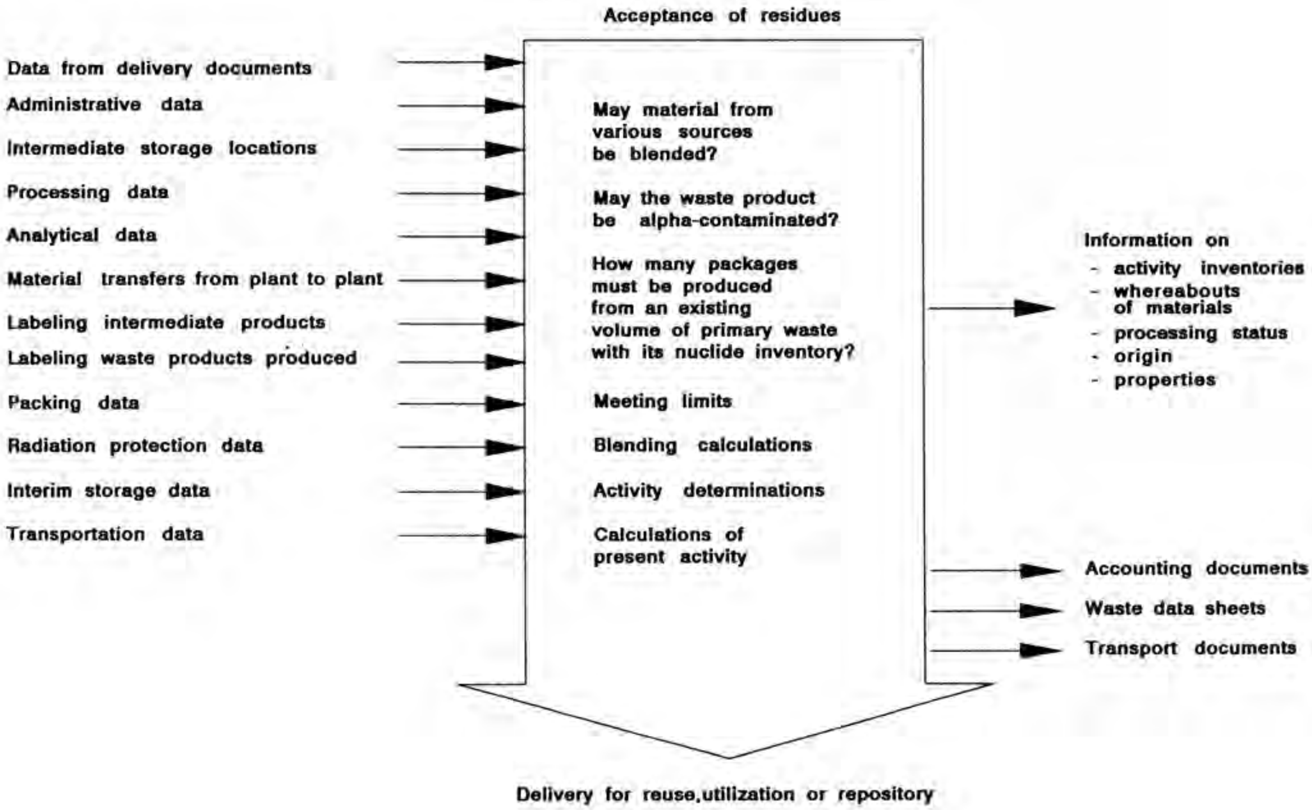


Fig. 4. Bookkeeping System KADABRA.

making, and in checking limits, for further processing, the production of waste forms, and also for packaging into waste containers inner canisters filled with waste forms. The system also calculates the activity inventories, $I_{(n,i)}$, on the basis of weights or dose rates measured. If several stations are passed for these activities, such changes of location are also entered. Blending substances from various sources is technically and economically reasonable and, at the same time, makes safe processing of radioactive primary waste. However, as each package produced can be the property of only one owner, the packages must be allocated by the system on the basis of quantities, so that changes in substances are taken into account. Decay calculations are fun for longer periods of intermediate storage. At the end of the conditioning phase, a dataset is available in the system as shown in Table II. A transport data file providing information on all respective data completes KADABRA.

No specialized knowledge in electronic data processing is required for users of the bookkeeping system. The system guides the user by means of dialogues, both for data entry and for information output.

REFERENCES

1. BRENNECKE, R. and WARNECKE, E., "Requirements on Radioactive Wastes for Disposal," PTB-SE-17.
2. "Verodnung uber die innerstaatliche und grenzuberschreitende Beforderung gefahrlicher Guter auf StraBen," "Verordnung uber die innerstaatliche und grenzuberschreitende Beforderung

gefahrlicher Guter mit Eisenbahnen," Bundesgesetzblatt 1985, pp. 1550 ff.

3. HAUSER, W., Optimierung der Aktivitatsbeladung von schwach- und mittelaktiven Abfallgebinden (PROMAX)," KfK-report 3825 (Jan. 1985).

4. POLSTER, F. I., WEIDEMANN, R. and STEGMAIER, W., "KADABRA" (Karlsruhe Data Base for Radioactive Wastes), KfK-report 3963 (Sept. 1985).