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A. H. Akbar, R. Köster, G. Rudolph
Institut für Nukleare Entsorgungstechnik
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Kernforschungszentrum Karlsruhe

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A.H. Akbar[+], R. Köster, G. Rudolph

Kernforschungszentrum Karlsruhe GmbH, Karlsruhe

[+]Present address: Pakistan Atomic Energy Commission
Islamabad / Pakistan

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Abstract

A possible way of performing the cement fixation of radioactive wastes is the incorporation into cement pellets on a pan pelletizer, followed by embedding the pellets into an inactive cement matrix. This procedure is suitable for various types of waste, particularly for medium level liquid wastes, and can be used both at drum disposal and at in-situ solidification. This report describes some initial studies on the pelletization technique using a laboratory pelletizer. Formation and size of the pellets have been found to be determined by speed, angle, and load of the pan, ratio and mode of addition of the liquid and solid components, etc. Pellets in various compositions have been produced from cement and water or simulated waste solution, in some cases with the addition of bentonite for improving cesium retention. Some mechanical properties of the pellets such as fall height of fresh pellets, development of hardness (crush test), impact and abrasion resistance, have been determined. Some preliminary experiments were done on backfilling the void space between the pellets - about 40 per cent of the bulk volume - with cement grouts of appropriate compositions.

Verfestigung radioaktiver Abfallösungen durch Pelletierung

Zusammenfassung

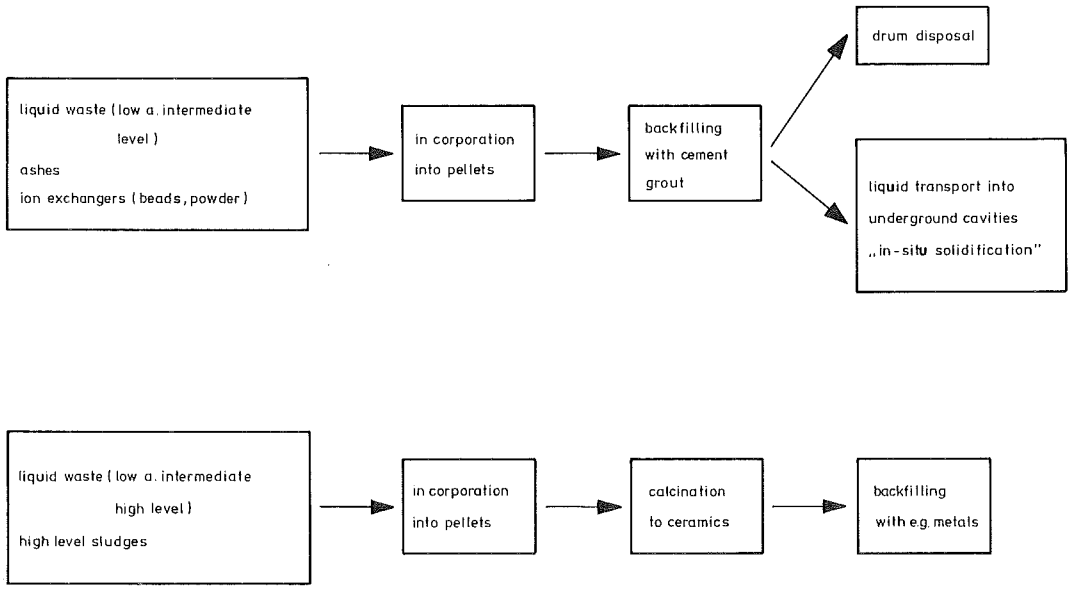
Die Zementierung radioaktiver Abfälle läßt sich in der Weise durchführen, daß zunächst auf einem Pelletierteller Zementpellets hergestellt werden, die anschließend in eine inaktive Zementmatrix eingebettet werden. Dieses Verfahren ist geeignet für verschiedene Abfallarten, vor allem aber für mittelaktive Abfallösungen, und kann sowohl bei der Endlagerung in Fässern als auch bei der In-situ-Verfestigung eingesetzt werden. Dieser Bericht beschreibt erste Pelletierversuche, die auf einem Labor-Pelletierteller vorgenommen wurden. Untersucht wurde die Abhängigkeit der Pelletbildung und der Pelletgröße von Drehgeschwindigkeit, Neigung und Beladung des Tellers, Verhältnis der flüssigen und festen Komponenten, Zugabeart u.a. Pellets aus Zement und Wasser oder simulierter Abfallösung wurden in verschiedenen Zusammensetzungen hergestellt, in einigen Fällen unter Zusatz von Bentonit zur Verringerung der Auslaugbarkeit von Cäsium. Einige mechanische Eigenschaften, nämlich Fallhöhe der frischen Pellets, Härtentwicklung (Bruchtest), Stoß- und Abriebfestigkeit, wurden bestimmt. Ferner wurden einige Versuche unternommen, die Hohlräume zwischen den Pellets, die ungefähr 40 % des Schüttvolumens ausmachen, mit Zementmörtel geeigneter Zusammensetzung zu verfüllen.

Contents

1. Introduction

A concept of a final storage in salt formations, based on experiences in the Asse salt mine and using drum disposal, is considered in the first place for the treatment of low- and intermediate-level waste solutions from the projected German integrated fuel cycle back end center. However, an alternative technique is being investigated in a special R&D program which uses "in-situ" solidification. This means that the cementitious grout shall be pumped directly into a large cavity in a salt formation under the site where the waste originates. Detailed discussions have suggested that a preceeding fixation of the waste solution into cement pellets which then, after mixing with an inactive cement milk, would be transferred into the underground, would offer some advantages over direct mixing of waste solution and cement. These include easy adjustment of the grout composition to the required consistency, low generation of heat of hydration in the underground cavity, and easy quality control. The pelletization technique, however, can be utilized in a drum fixation as well. In either case, the backfilling with an inactive cement grout provides an additional barrier which renders the final product superior to conventional cement blocks (1). A further improvement can conceivably be achieved by coating the pellets, i.e. by applying a protective layer of an organic or inorganic material, or by embedding them into an e.g. organic matrix.

The following scheme shows in principle where the pelletization technique can be used in the treatment of low- and medium level radioactive wastes from a fuel reprocessing plant.



Possible flow sheets for the solidification of low, medium, and high level wastes by pelletization techniques

The term "pelletization" may be used for a number of processes in which a material in powder, or fine grain, form is converted into pellets of fairly uniform size and shape;

these include press granulation, spray drying, rolling, extrusion, and crystallization. Also ploughshare mixers can be used for pelletization. In the work described here the term is used for pellet formation on a rotating pan where the pellets are produced by adding a liquid to a solid powder held in rolling motion in the pan.The particles thus being formed are held together by the cohesive force of the granulating liquid and therefore are of relatively low mechanical strength. As the solids are essentially cement and the liquids an aqueous waste solution, the initially formed pellets undergo further hardening during storage. This process can be enhanced by properly adjusting various parameters such as pellet composition and preparation procedure. Present investigations are aimed at improving the initial and final strength of the pellets, accelerating the hardening process and increasing the amount of waste to be incorporated. This report presents the results of some initial studies on the pelletization technique; further investigations into the chemistry of pelletization will be published in a later paper.

The output of cemented low and medium level wastes from a 1400 tons per year fuel reprocessing plant is about 3500 m^3 per year or 4.5 tons per work hour. As the capacity of a pan pelletizer is proportional to the square of its diameter and the laboratory machine (diameter 0.4 m) can produce 50 kg of cement pellets per hour, the diameter required e.g. for one pelletizer for large scale waste processing can be calculated to be about 5.3 m. Industrial use of pans of this size is a well established technology.

2. Inactive Laboratory Scale Operation of the Pelletizer

2.1 Description of the Pelletizer

The pelletizer used was of the type TR 04, manufactured by Maschinenfabrik Gustav Eirich, Hardheim/Odenwald. It consisted of an inclined pan, 40 cm in diameter which rotated in a clockwise direction. The drive unit is fitted to a thyristor controlled direct current motor (app. 500 Watts) and a maintenance free worm gear drive. The pan revolutions are continuously variable. For the cleaning of the pan wall and pan floor, an adjustable scraper is provided. Fig. 1 shows the pelletizer during operation.

2.2 Mode of Application

The machine can be charged manually or continuously using metering devices of the required kind. The material is fed on the pan and wetted with the solution.
The wetted material is transported to the highest point of the pan and then rolls in an even stream back down to the lower half of the granulating pan. The solutions are added at certain points or places. The rolling movement and repeated tumbling forces and addition of moisture causes pellets to form. The pellets gradually grow in size and with each revolution work their way on to the outer and the upper surface of the granulating mass in the lower quadrant of the dish. As soon as the pan is filled, the pellets will continuously roll over the pan edge.

2.3 Operating Features

Based on working experiments in the pelletization of simulated waste solutions, the features influencing the operation ot the pelletizer were found to be angle of inclination, rotational speed of the pan, location of addition of solution and solids in the pan. Also the size of the pellets was found to be dependent on these factors. The formation of pellets appears to be influenced also by the way in which the liquid is pumped or sprayed onto the solid material.

2.3.1 Speed of Rotation of the Pan

There is a critical speed of the pelletizer, above which there is no movement of material in the pan due to centrifugal forces. It was found to be between 48.5 and 49 rpm. Good pelletization occured when the speed of rotation was from 19 rpm to 36 rpm, i.e. 0.40 to 0.75 of the critical speed. Above 36 rpm the movement of the material in the pan is impaired and at the critical speed the entire wet mass sticks to the walls of the pan. Below 19 rpm poor pelletization is observed as most of the wet mass slides instead of rolling.

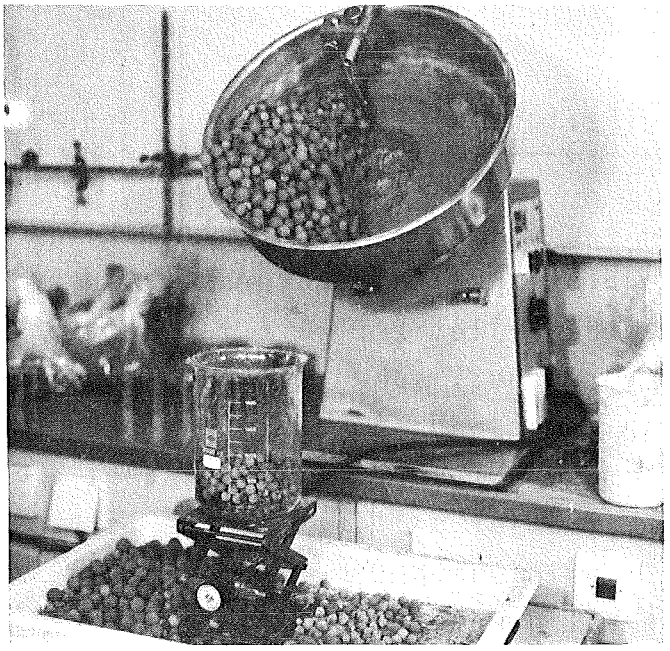


Fig. 1 Laboratory pelletizer during operation

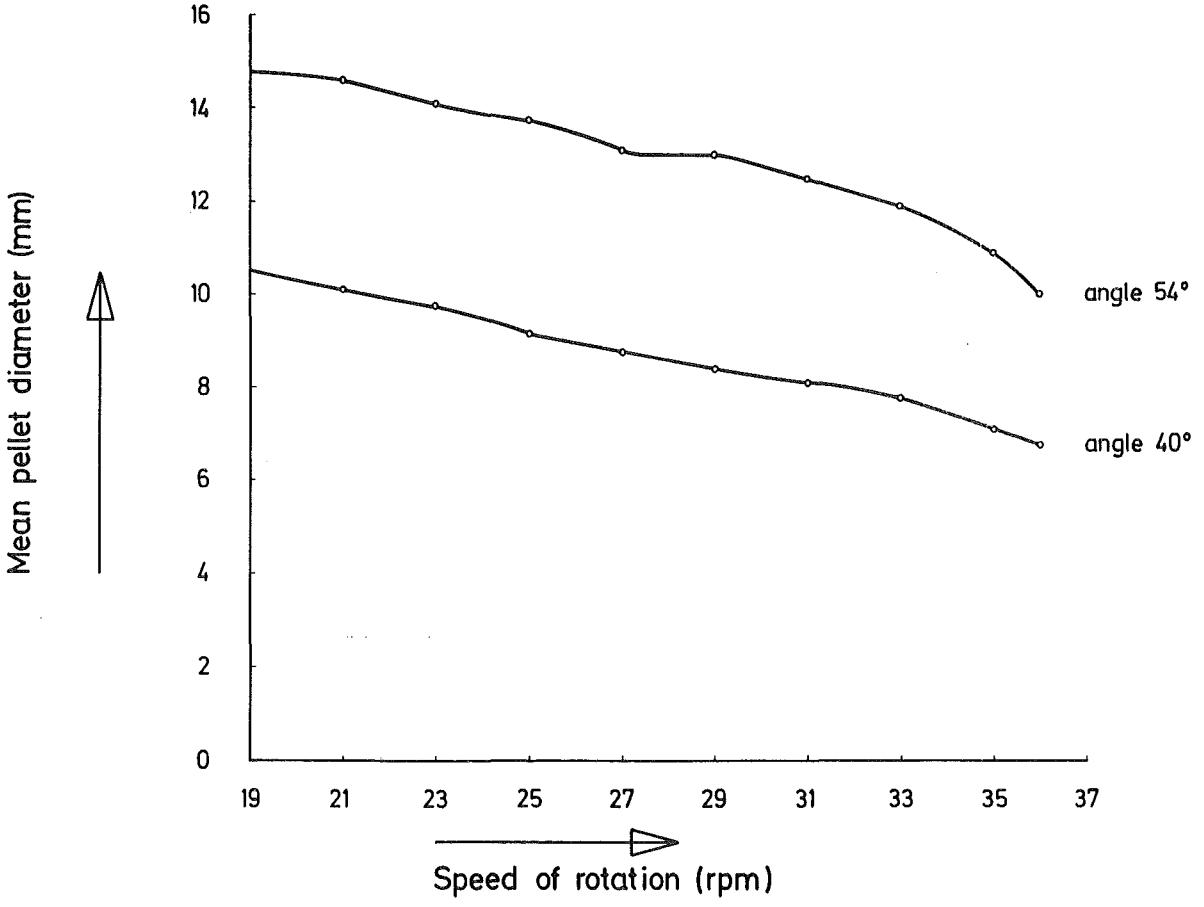


Fig. 2 Pellet diameter vs. speed of rotation (liquid: 40% $NaNO_3$ solution; solids 10 parts of portland cement + 1 part of bentonite)

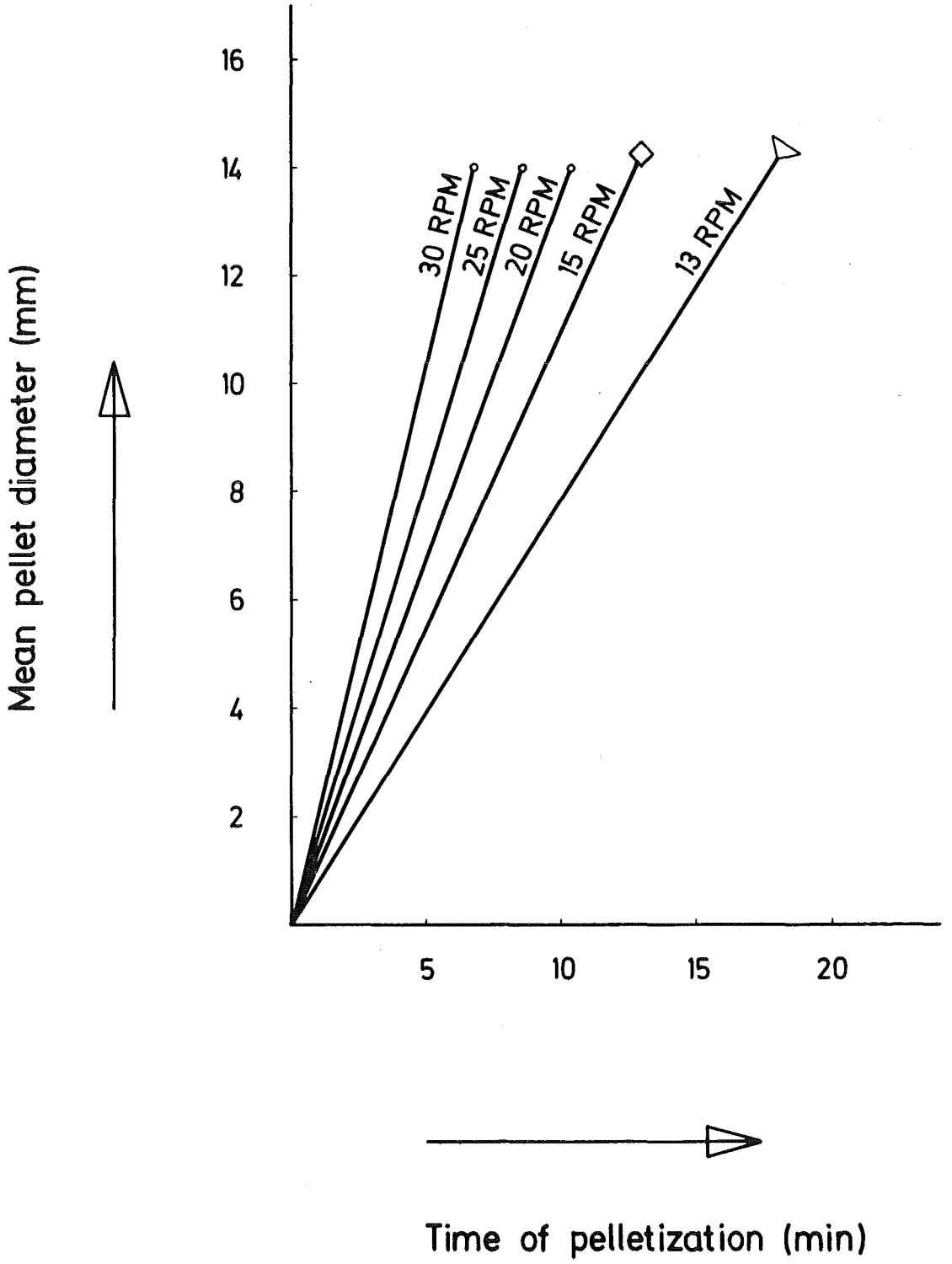


Fig. 3 Interdependence of pellet diameter, speed of rotation and residence time (composition of feed as in Fig. 2; inclination of pan 47°)

Speed of rotation from 26 rpm to 30 rpm gave the best rolling motion to the cement feed. Fig. 2 shows the size of the pellets at different speeds. Fig. 3 shows the effect of pan speed on the pellet growth plotted versus time of pelletization, corresponding to the mean residence time on the pan.
A useful criterion for the optimum speed is to draw a vertical line through the centre of the pan and to adjust the speed in such a manner that the material does not cross this line while tumbling.

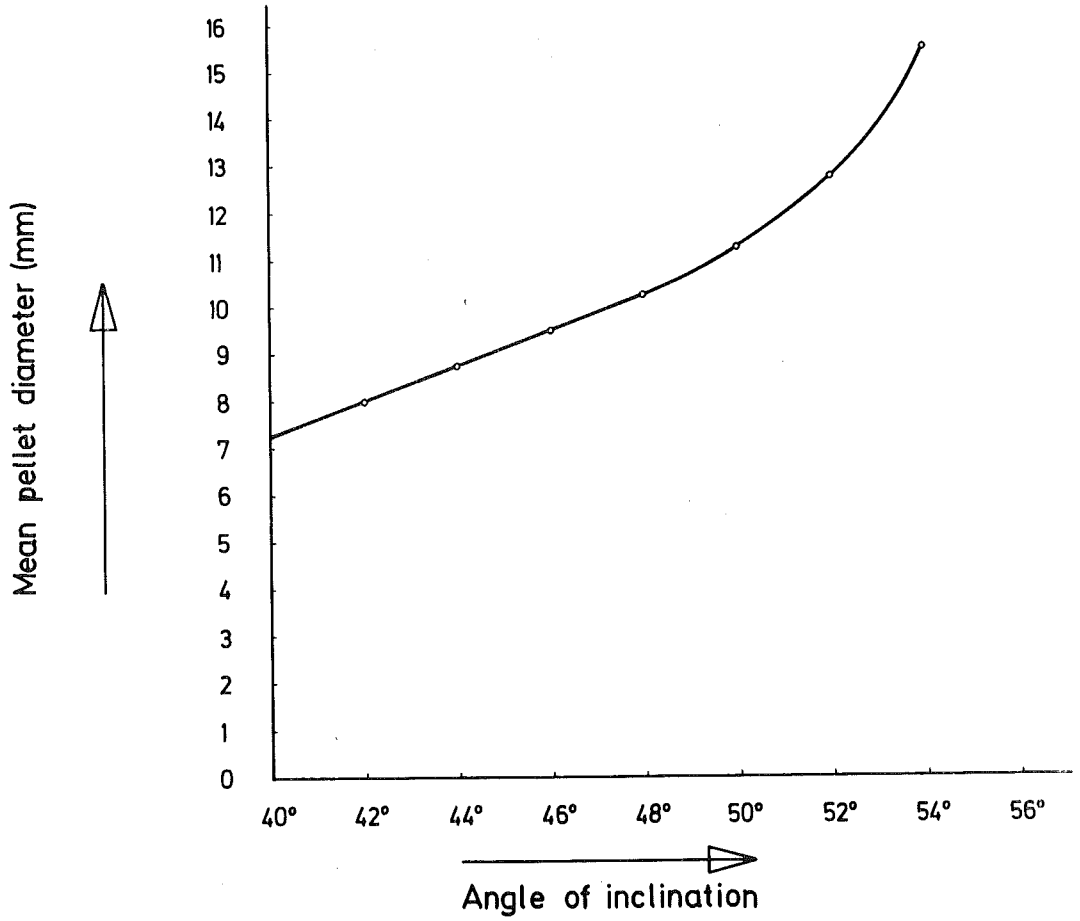


Fig. 4 Pellet size vs. angle of inclination (composition of feed as in Fig. 2; speed of rotation 26 rpm)

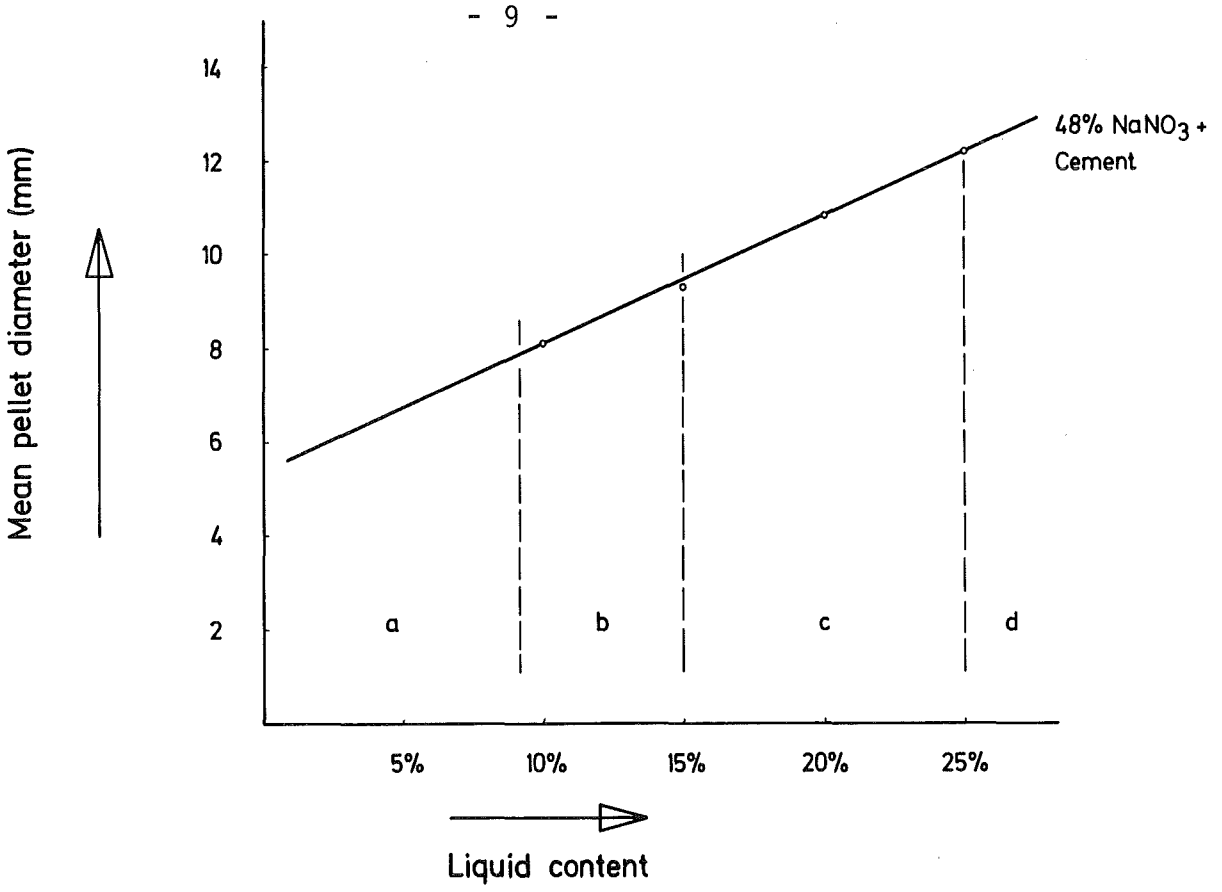


Fig. 5 Size of pellets vs. liquid content (relative to amount of cement) at sodium nitrate solution

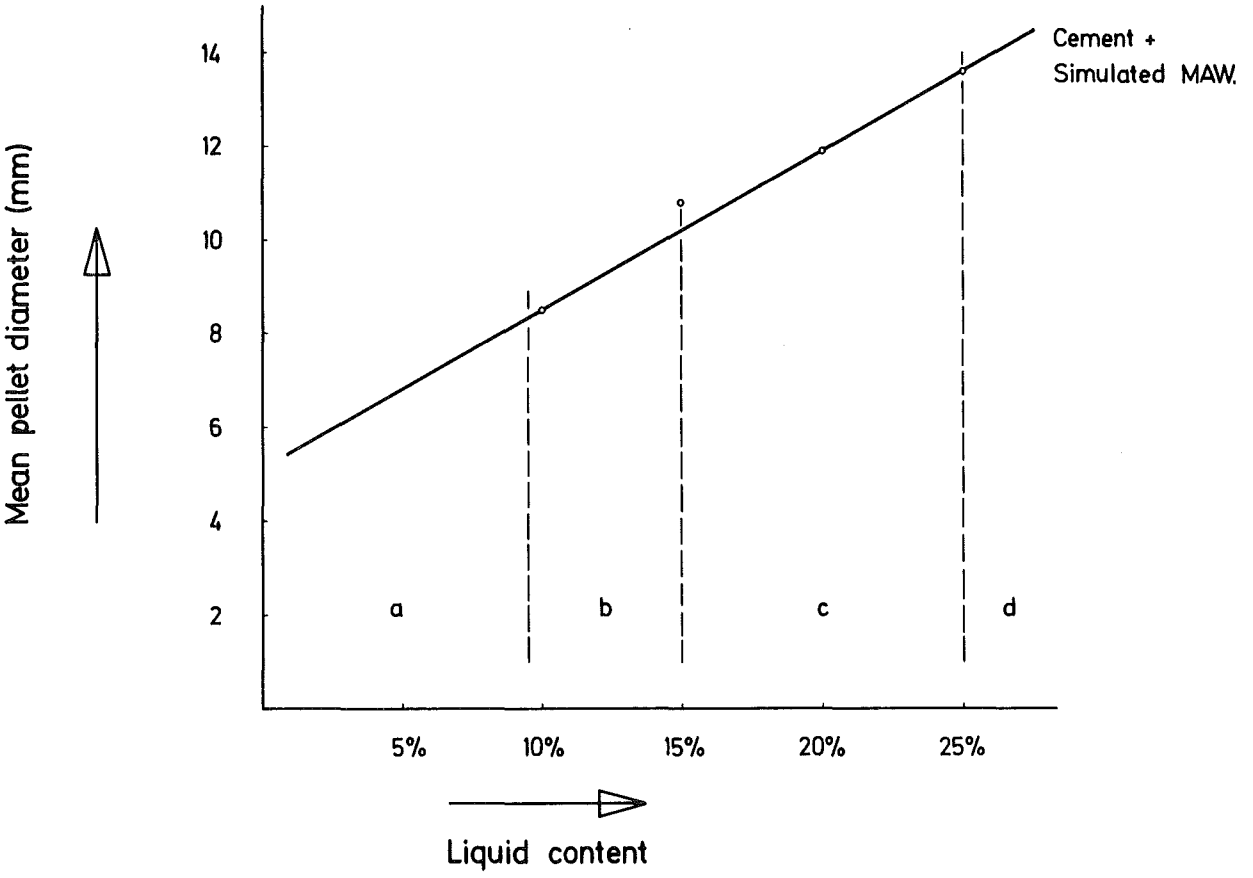


Fig. 6 Size of pellets vs. liquid content (relative to amount of cement) at simulated waste solution

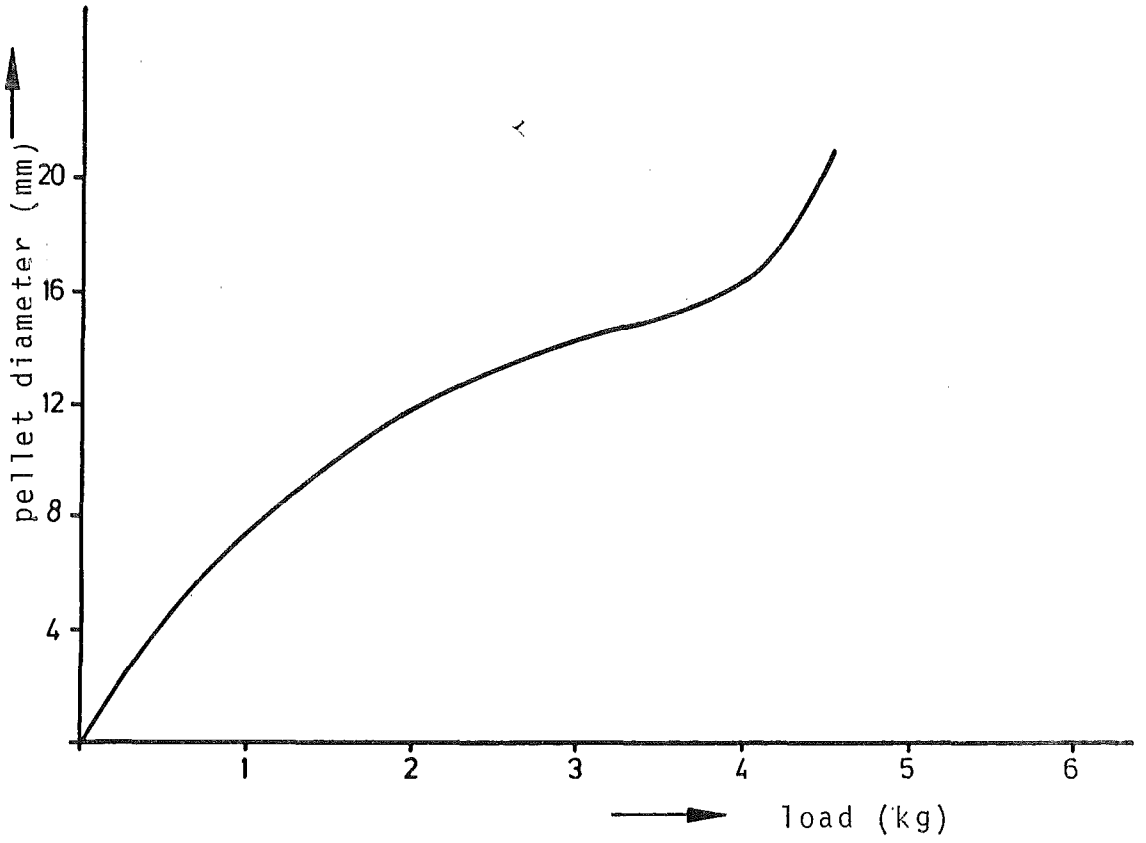


Fig. 7 Pellet size vs. load in the pan

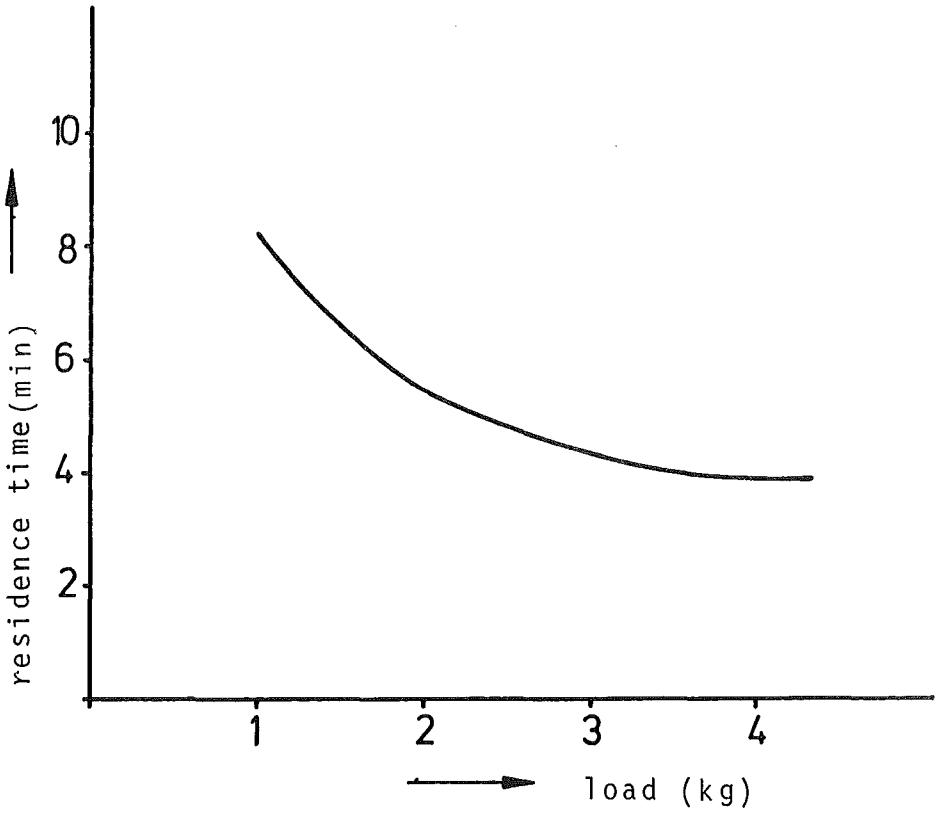


Fig. 8 Residence time vs. load in the pan

2.3.2 Angle of Inclination of the Pan

An angle of inclination of 45^{o} and 47^{o} was found to yield pellets of uniform size. If the angle is 50^{o} or above, there is a significant increase in the diameter of the pellets and pellets of 20 to 22 mm diameter have been obtained. Below 45^{o}, the pellets were irregular in shape. Fig. 4 shows the size of pellets versus change of angle of inclination.

2.3.3 Effect of Addition of Waste Solution

The experiments showed that liquid must be added onto the recycling feed in the upper quadrant of the pan. In this manner pellets of uniform size are obtained. Fig. 5 and 6 show the mean pellet size as a function of the liquid content of the pelletized material for $NaNO_3$ solution and simulated waste solution, respectively. In the section a the liquid content is not sufficient for pellet formation which starts in section b. In section d, the growth rate is very high and the pellets collapse under their own weight due to their high moisture content. Best pellet formation is in section c.

2.3.4 Effect of Load in the Pan

A practical limit was found for the load of pellets in the pan. Above 4.5 kg, the rate of growth is extremely large and the pellets behave like balls in a ball mill. Fig. 7 shows the pellet growth versus variations of the pan load. Fig. 8 shows the residence time in the pan as a function of the load.

3. Production and Composition of Pellets

A wide variety of pellets containing portland cement (PZ 35 F), simulated medium level waste or $NaNO_3$ solution, natural bentonite, high alumina cement and inactive cesium were made on the pelletizer machine. Pellets were also made containing up to 30% of solid $NaNO_3$ salt with and without natural bentonite. Natural bentonite of a particular type was added in order to reduce the leachability of cesium contained in radioactive waste solutions (2). The purpose of blending high alumina cement into portland cement was to attain an early mechanical strength of the pellets.

Table 1 shows the composition of some types of pellets which were used in the later investigations. No. 1 is produced from portland cement and water only for the sake of comparison.

A series of pellets (No. 2 to 5) was made with a simulated MAW solution whose composition was calculated using the flowsheet of a projected 1400 ton per year fuel reprocessing plant and analytical data from the WAK reprocessing plant at Karlsruhe. Its composition is given in Table 2. In these compositions an increasing fraction of the portland cement is replaced by natural bentonite.

The possibility of incorporating solid salts into pellets in order to attain a high salt content was also demonstrated (No. 6 to 8). This was done by giving a mixture of portland cement and solid sodium nitrate on to the pan and spraying water over the rolling mass. The pellets thus formed were bigger in size as compared to other pellets made with a lower salt content. Size, however, can be controlled by changing the angle of inclination of the pan and the speed of rotation.

Table 1 Composition of pellets (% by weight)

No.	Portland cement	Bentonite	water	MAW salts	$NaNO_3$	others
1	81.3	-	18.7	-	-	-
2	83.0	-	9.4	7.6	-	-
3	77.9	3.9	10.0	8.2	-	-
4	74.3	7.4	10.0	8.2	-	-
5	71.0	10.7	10.1	8.2	-	-
6	62.5	-	8.1	-	29.4[a)]	-
7	59.8	-	8.6	-	30.6[a)]	-
8	55.1	5.5	10.1	-	29.4[a)]	-
9	82.6	-	7.0	-	8.3	2.05 Cs
10	73.6	7.4	8.8	-	8.1	2.05 Cs
11	53.8	7.2	12.6	8.5	-	17.9 TSZ[b)]

a) Solid sodium nitrate

b) TSZ= Tonerde-Schmelz-Zement (high alumina cement)

Table 2 Composition of Simulated Intermediate-level Liquid Waste[+)]

Constituent	g/l	Constituent	g/l
$NaNO_3$	450.0	Tributyl phosphate	0.2
$NaNO_2$	5.0	Dibutyl phosphate	0.1
$Fe(NO_3)_3$	0.1	Kerosene	0.02
$Ni(NO_3)_2$	0.01	Sodium oxalate	10.0
$Cr(NO_3)_3$	0.01	Sodium tartrate	10.0
$Ca(NO_3)_2$	0.15	NaF	2.0
$Mn(NO_3)_2$	0.02	Detergents	2.0
$Sr(NO_3)_2$	0.002	Cs	0.004
$Mg(NO_3)_2$	0.2	P as Na_2HPO_4	0.2
$Ce(NO_3)_4$	0.02	Prepared with 1-molar HNO_3	
$Al(NO_3)_3$	0.03	Adjusted with NaOH to pH8.5-9	

+) In later experiments, EDTA (1 g/l) and sodium citrate (5 g/l) was added to this composition.

For later determinations of cesium leachability under inactive conditions pellets were also made containing 2% of Cs in the final product. This was done by dissolving the required amount of $CsNO_3$ in 40% or 48% $NaNO_3$ solutions and incorporating them into portland cement, with and without natural bentonite (samples No. 9 and 10). In order to speed up the hardening of the pellets, a mixture of high alumina cement and portland cement in the ratio of 1 : 3 was pelletized with bentonite (sample No. 10).

4. Mechanical Properties of the Pellets

4.1 Fall Height of Fresh Pellets

Observations were made on various freshly prepared pellets to see up to which height they may fall from the granulating pan without breaking or deformation. For pellets containing simulated MAW solutions (No. 2 in Table 1) this height was 74 cm, for those containing natural bentonite in addition to simulated MAW (No. 5), a height of 65 cm could not be exceeded. Pellets containing up to 30% $NaNO_3$ salts (No. 6, 7, 8) broke beyond 52 cm.

A simple experiment was performed by taking 25 cement pellets about one hour after preparation and dropping them from a height of 130 cm on to a steel plate. Out of 25 pellets of composition No. 2 (Table 1), 11 pellets fractured and four broke into pieces. The 25 pellets containing bentonite (No. 5) all fractured and broke into pieces.

Four different pellet samples were made to fall on a steel plate. The heights up to which they remain intact will be a measure of mechanical strength. All the four samples were tested two hours after preparation.

1. Pellets with simulated MAW and portland cement only (No. 2)	118 cm
2. Pellets with simulated MAW + bentonite + portland cement (No. 5)	83 cm
3. Pellets with 30% $NaNO_3$ + portland cement (No. 7).	61 cm
4. Pellets with simulated MAW, portland cement and high alumina cement and bentonite (No. 11)	141 cm

4.2 Crush Test

Crush tests were carried out on pellets on a special instrument in which the pellet is placed between two parallel plates and a gradually increasing load is applied until the pellet bursts. The load at which the pellet breaks is a measure of its crush resistance. The maximume load the machine can measure is 25 kg. The tests were carried out on pellets having the same size of approximately 13 mm diameter after setting times ranging from 30 minutes to 288 hours.

The composition of the pellets was as listed in Table 1 (p. 11). After preparation, the pellets were allowed to cure in a close container until the crush test was performed, i.e. for a minimum of half an hour.

The relationship between the age of the pellets and the corresponding breaking load is plotted in Fig. 9 and 10. It is evident from the plots that the addition of natural bentonite retards the setting of the cement-waste pellets. This is true for all the samples except No. 11. The incorporation of 30% $NaNO_3$ salt in the pellets (No. 6) does not have a significant effect on the strength of the pellets and the resulting curve is quite close to curve No. 2 (pellets with portland cement and simulated medium-level waste).

A significant increase of the initial strength of the pellets can be achieved by substituting one quarter of the portland cement by high alumina cement (No. 10). As can be seen from Table 2, the rapid setting has the desired effect of increasing the amount of waste liquid which can be incorporated into the pellets.

4.3 Impact Test

Samples of pellets being considered for incorporation of radioactive wastes were subjected to an impact test to determine the relationship between the energy of impact and the resulting increase in surface area of the demaged samples. Compressive strength tests alone are not sufficient for evaluating the safety of

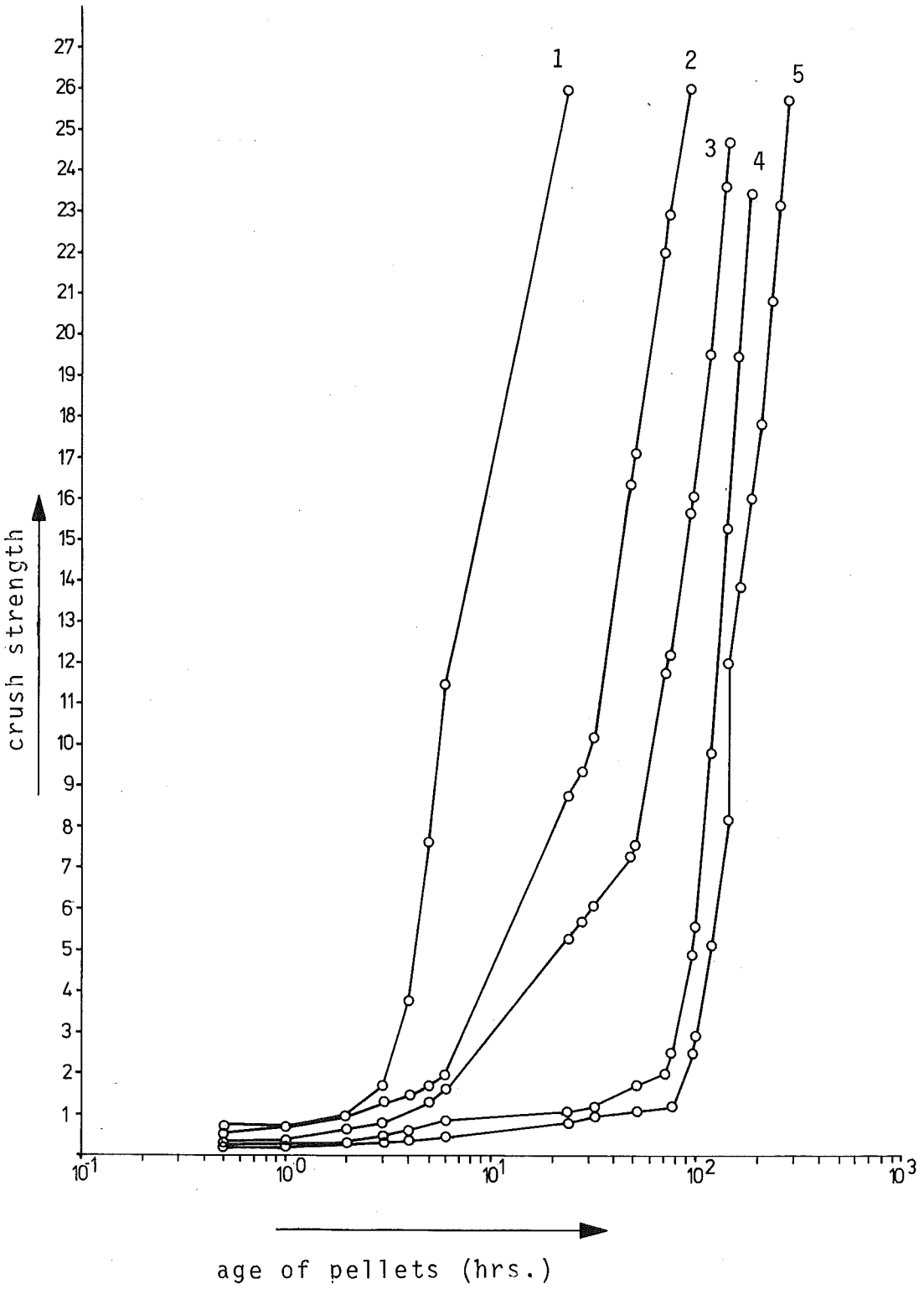


Fig. 9: Development of crush strength (kg) with age of pellets (compositions No. 1-5 of Table 1)

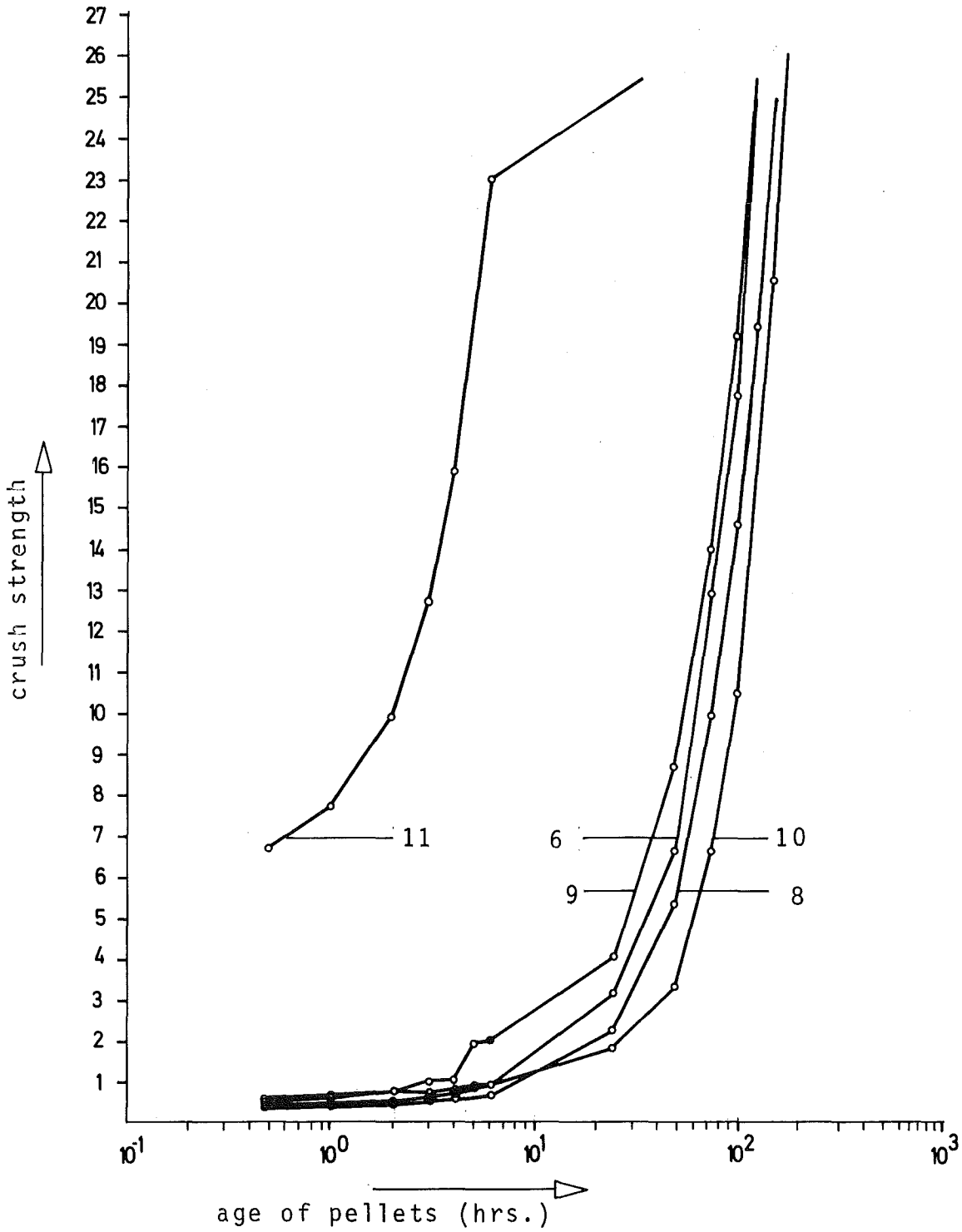


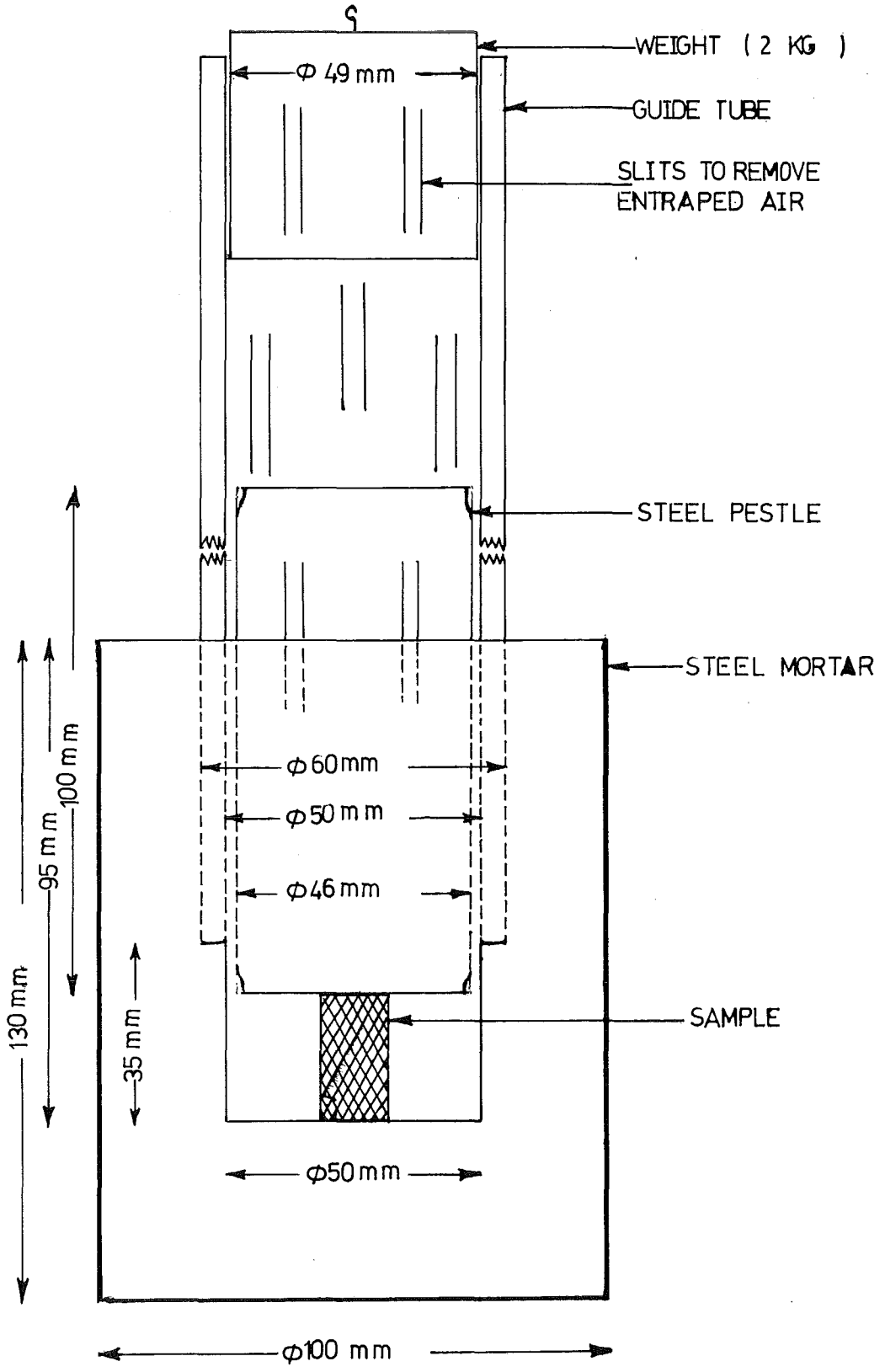
Fig. 10: Development of crush strength (kg) with age of pellets (compositions No. 6, 8 . 11 of Table 1)

waste forms when subjected to impact. A test is therefore required that measures the property of the waste form that is directly affected by impact. The increase in surface area has been taken as a measure of the impact resistance. Pellets with lower values of change in surface area are better in resistance against impact.

Table 1 (p. 11) shows the percentage composition of the samples of pellets. The pellets had approximately the same diameter of 11 mm. All the seven samples were made on the pelletizing machine keeping the parameters of the machine such as speed of rotation and angle of inclination of the pan the same. The pellets were placed in plastic bottles. After sealing with plastic lid, the pellets were allowed to cure for 28 days.

A diagram of the "Impact Test" apparatus is shown in Fig. 11. This apparatus was described in the literature (3); however some modifications were made in the procedure. About 17 pellets of approximately the same diameter were placed in the cavity and the pestle was placed firmly upon them. A weight of 2 kg was dropped through a guide tube from a known height (starting from 50 cm) to hit the pestle near the center. A series of five determinations was made for each type of pellets where the height was 50 cm, 60 cm, 70 cm, 80 cm, 90 cm corresponding to 9.8, 11.8, 13,7, 15,7, 17.7 Joules of energy. The crushed sample was transferred to a stack of 10 sieves (2000 μ to 25 μ) and screened by agitating the stack for 8 minutes by a shaker.

The increase in surface area was calculated using the following assumptions:



IMPACT TEST APPARATUS

Fig. 11

1. The particles were spheres with an average diameter equal to the average particle size within a given fraction.
2. Losses of the sample (due to evaporation of water during the experiment, dust etc.) are negligible.
3. Energy input is homogeneously distributed over the whole sample.

The formula used for calculating the surface area is as follows:

$$A = \frac{6G}{\rho} \Sigma \frac{f_i}{\bar{x}_i}$$

where A = surface area of the crushed sample (cm^2)

G = total weight of the sample (g)

ρ = density of the sample (g/cm^3)

$\overline{X}_i$ = average particle size within a given size fraction (cm)

f_i = mass fraction of the total

The increase in area can then be calculated by substracting from the initial surface area of the sample. Table 3 presents the values of increase in surface area per unit impact energy. The following conclusions can be drawn from these data:

- An increase in natural bentonite from 0% (Sample No. 2) to 10.7% (Sample No. 5) increases the value of A/E, in other words decreases the impact resistance, by 50%.

- In the other samples, the values of A/E are approximately the same, which means that neither the use of a high alumina cement-portland cement mixture nor the incorporation of solid sodium nitrate has any noticeable effect on the impact resistance of the pellets.

WALLACE and KELLEY (3) found values for A/E near 10 for glass samples and between 5 and 10 for neat cement samples. Addition of 40% of a sludge to the cement gave an increase in A/E to 20 to 35 cm^2/Joule.

4.4 Abrasion Test

Pellets must be sufficiently resistant to attrition even before they harden, to prevent any dust formation during handling. A simple procedure was used to measure

Table 3: Increase in surface area per unit impact energy A/E (cm^2/Joule)

Sample No.	Impact Energy E (Joules) 9.8	11.8	13.7	15.7	17.7	Mean
2	24.0	21.6	20.4	17.6	17.4	20.2
3	26.2	23.3	21.3	20.7	22.2	22.7
4	30.6	28.4	24.7	23.0	22.8	25.9
5	27.5	30.6	32.5	34.5	34.5	31.9
7	24.9	22.5	21.2	21.4	21.3	22.3
8	26.7	23.9	21.0	24.5	23.0	23.7
11	27.5	23.3	20.0	17.3	15.5	20.7

the abrasion resistance of pellets of various compositions.

For each test, 1500 grams of pellets were placed upon a wire mesh screen which was fixed on a shaking machine. The dust was collected in a tray under the screen and weighed after 50 and 100 vibrations. For each pellet composition the test was done six hours after preparation of the pellets and repeated in continuing intervals until the losses were negligible. The results are listed in Table 4.

The amount of dust generally decreases with increasing amount of bentonite contained in the pellets. The sample 5, containing more than 10% of bentonite, produces less dust by 66% than sample 2 with no bentonite added. The best results are obtained on sample 11 prepared from the fast-setting mixture of portland cement and high alumina cement.

Table 4: Abrasion Test of Pellets: Dust formation after 50 Vibrations (upper figure) and 100 vibrations (lower figure) in weight percent

Age of pellets (hours)	Sample No. (composition see Table 1)							
	2	3	4	5	6	8	9	11
6	4.2	3.7	1.5	0.7	9.5	6.5	1.9	0.6
	11.4	10.7	6.5	3.8	20.7	18.7	6.4	2.5
24	1.6	0.9	0.3	0.2	5.9	3.7	0.8	0
	5.8	3.9	1.6	0.6	11.3	8.8	1.8	0.5
48	0.9	0.7	0.005	0	2.7	1.4	0.04	0
	3.0	2.0	1.0	0.1	7.9	5.3	0.7	0.08
72	0.8	0.7	0.02		1.4	0.4	0	0
	1.9	1.5	0.2		3.6	2.6	0.04	0.01
96	0.7	0.4	0.006		0.8	0.06		0
	1.2	0.9	0.002		1.6	0.8		0.01
120	0.2	0.1			0.006	0		
	0.8	0.4			0.9	0.1		
144	0.05	0.006			0	0		
	0.2	0.02			0.3	0.02		
168	0.006							
	0.02							

5. Embedding of the Pellets

5.1 Determination of Void Space

The relative amount of void space between the pellets is defined by

$$\text{void space} = 1 - \frac{\text{bulk density}}{\text{true density}}$$

The bulk density was measured by weighing about 100 g of material into a 100 cm^3 graduated cylinder, vibrating the cylinder until the level no longer changed, and reading the bulk volume. The true density was obtained by filling a 100 cm^3 graduated cylinder about half way with water, slowly pouring in about 100 g of material and determining the weight and volume increase.
Table 5 shows the fraction of void space at various types of pellets.

Table 5: Void space between packed pellets

Type of pellets[a)]	Bulk density	true density	void space
2	1.47	2.44	0.39
4	1.49	2.43	0.38
6	1.47	2.42	0.39
11	1.47	2.45	0.40

[a)] cf. Table 1

5.2 Cement Grouts for Embedding Pellets

In order to study the incorporation of pellets into a monolithic block, a number of experiments was performed to pour cement-water grouts, with different water to cement ratios, over various types of pellets prepared fresh or cured some days.

The pellets were filled into plastic cylinders (6 x 5 cm) to three quarters of their height and covered with a lid to allow curing. Water-cement grouts were prepared with water to cement ratios from 0.38 to 0.9 with and without the addition of a water-reducing agent 'BVF' (from Portland Zement-Werke, Heidelberg). It is added to the cement grouts in the amount of 3 ml per 100 g of cement.

These grouts were poured on pellets cured for 2 hours, 6 hours, one day and 7 days, so that all the pellets were completely covered with the cement grout. Two separate sets of samples were made, one with shaking the mixture and the other one without shaking. Then these samples were allowed to cure for several days after closing them tightly with lids. After 14 days the cylinders were cut into two halves by a diamond-lined saw.

In case of pellets aged for 6 hours or less, there was some deformation of the pellets if the water-to-cement ratio was above 0.5.

Generally the cement-water grouts poured over pellets cured 3 days or more did not dissolve or deform the pellets and all the spaces between the pellets were filled by the grouts even without

shaking if the water to cement ratio was above 0.5. By the addition of 'BVF' to the grouts with a lower water to cement ratio (0.38 to 0.5) it was also possible to obtain products where the spaces were filled by the cement grouts, however with shaking.

References

(1) R. Köster, G. Krämer, R. Kroebel, IAEA-SM-243/16 (1979)

(2) G. Rudolph, R. Köster, KfK-2842 (1979)

(3) R.M. Wallace, J.A. Kelley, DP-1400 (1976)