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CRITICALITY SAFETY STUDY OF PU CONTAMINATED CARBON WASTE STORED IN 100 L STEEL DRUMS

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I/ Introduction

A part of recent studies on the minimal critical areal density (Dminca) is briefly exposed.
Its customary conditions of use and its deficiencies are mentioned.
Then the Dminca values for the infinite medium of Pu contaminated carbon waste are precised.
They are confirmed by calculations on infinite plane arrays of several layers of 100 l waste drums.
The drums steel walls poisoning effect is possibly taken into account.
Practically, the criticality safety analysis is also based on other favourable effects.
Thus, the proposed regulatory value of 0.1 g/cm^2 is fullfilled in our storages.

II/ Dminca

1/ Present use

Dminca is the product of the critical thickness by the concentration.
Its minimum allows to calculate the unit cell dimensions of the waste storage arrays. The practice shows it is a very safe but penalizing notion.
For Pu bearing wastes, the french rule refers to $^{239}Pu\text{-}H_2O$ medium. When concrete reflected, its Dminca is 0.20 g/cm^2.

2/ Deficiencies

- Theory : Dminca is independant of the fissile density only for unreflected mediums.
Corrective factors can be established on existing data,
- Practice : other moderating materials than water give lower Dminca (reduced density water, CH_2, C_2F_4, Carbon).

III/ Dminca of Pu-C

By APOLLO-DTF codes and their associated libraries, the calculated (concrete reflected) Dminca is 0.036 g/cm^2.

IV/ Storages in 100l steel walls drums

The reactivity of concrete reflected infinite plane arrays of drums is studied by MORET.
Dminca roughly varies linearly with the steel thickness of walls, from 0.036 g/cm^2 (no steel) to 0.17 g/cm^2 (0.105 cm of steel, totally described) or to 0.137 g/cm^2 (0.105 cm of steel, lateral walls only described).

V/ Safety Analysis

Some factors, as the following ones, improve the storage safety :
- Pu is not 100 % ^{239} Pu,
- the used cubical description of the drum is conservative,
- the steel thickness is larger than 0.07 cm,
- the amount of Carbon is less than 60 kg and the amount of Pu is less than 100 g per drum,
- Pu is not homogeneously distributed, but would be under the form of little spherical specks. Calculations show Dminca increases with the radius of the Pu (or PuO_2) Carbon coated sphere,
- neutron poisons as N and Cl are in the waste bulk.

VI/ Conclusion

For us, in contrary of a recent US analysis, Carbon as moderator, is of a great effect in waste.
The concrete reflected Dminca of Pu-C is 6 times lower than that of $Pu\text{-}H_2O$. Happily, the poisoning effect of drum steel walls is efficient, since for a 0.1 cm thickness, Dminca rises to 0.137 g/cm^2 (when only lateral walls are taken into account).
As they are other favourable safety elements, the regulatory value of 0.1 g/cm^2 is recommended for Pu-C wastes. This norm is practically fullfilled in french storages.

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CRITICALITY SAFETY STUDY OF Pu CONTAMINATED CARBON WASTE STORED IN 100 L STEEL DRUMS

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SUMMARY

The notion of the minimum critical areal density (D_{minca}) used to ensure the Criticality-Safety of poor solid waste is recalled with its deficiencies. D_{minca} is assumed constant, independent of the fissile material concentration. This assumption is only true for unreflected mediums. Corrective factors are established. Furthermore, the usual norm of the Pu-H_2O, which is 0.20 g/cm², (concrete reflected) is greater than that for other mediums, such as Pu contaminated graphite waste (Pu-C), which is 0.036 g/cm². D_{minca} calculated on infinite slabs is confirmed by calculations on infinite planar multilayers arrays of 100 l cubi l waste drums. Moreover, d_{minca} increases linearly with the steel thickness of the drums' walls and goes up to 0.17g/cm² for 0.105 cm of steel. The safety analysis on a real storage case takes into account the limited amount of Pu (100 g) and C (100 kg), the minimum thickness of 0.07 cm of drums' steel, their geometrical arrangment, the heterogeneity and size of contamination and the occurrence of neutronic poison (N and Cl) in the waste. Because of these parameters, the Keff are very less than 0.95 and the taken norm of 0.1 g/cm² for the Pu-C waste is fulfilled.
Finally, it is demonstrated that the mixing of Pu-C waste drums and Pu-H_2O wastes drums is allowed.

I - INTRODUCTION

This paper presents briefly and partly some recent studies [1-4] on minimum critical areal density applied to storages of graphite (C) wastes contaminated by Plutonium (Pu).
After recalling the use and deficiencies of this notion, d_{minca} is theoretically studied on infinite slabs, then on infinite planar arrays of 100 l cubical drums, with determination of the steel wall effect.
Finally, the safety analysis is carried out in more realistic cases, taking into account the favourable effects of the limited amounts of Pu and C, of the heterogeneity and grain sizes of the contamination, and of the occurrence of neutronic poisons in the wastes.

II - D_{minca}

II.1 - Present Use

D_{minca} is the product of the critical thickness by the fissile material concentration.
For Pu bearing waste, the French regular documents [5,6] recommend to take as a reference the ^{239}Pu-H_2O concrete reflected medium : d_{minca} is worth 0.20 g/cm² and the acceptable value is $0.7 \times d_{minca} = 0.14$ g/cm².
The sizing and the acceptability of a storage consist in comparing this value to the projected surface density of the storage

$$d_{ss} = N \times m/S \quad (1)$$

where m is the mass (g) of Pu per drum, S is the projection area (cm²) and N is the number of drums in the direction perpendicular to this area.

If $\quad d_{ss} = N \times m/S \leq 0.7 \times d_{minca} \quad (2)$

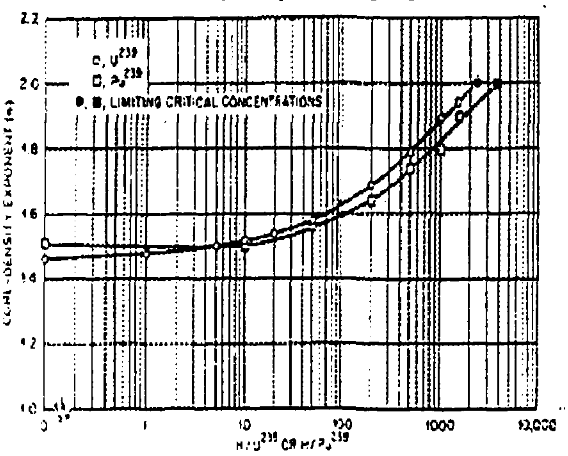
then the storage is safe, all the more it has been checked that the d_{minca} notion is practically very penalizing.

II.2 - Deficiencies

a-Theory

D_{minca} is assumed constant and independent of the density d of the fissile material. This assumption is true only for the unreflected mediums. With the data published by H.C. PAXTON [7a] reproduced in figure 1, it is possible to establish corrective factors for the critical surface density d_{cs} from the critical mass density exponent.

Figure 1
Density Exponent.[7a]



$$M_c = M_{c0}\,(d/d_0)^{-s} \qquad (3)$$
$$\rightarrow d_{cs} = d_{cs0}(d/d_0)^{a} \qquad (4)$$
$$\text{with} \quad a = (2-s)/3 \qquad (5)$$
$$\rightarrow d_{cs(s)} = d_{cs(s=2)} \times Fc \qquad (6)$$

If s=2, we find the expected and usual result $d_{cs}=d_{cs0}$=constant.

The corrective factors Fc are given below, for various ratios d/d_0 versus the moderating ratio H/Pu.

Table I
Example of Corrective Factors for Pu-H_2O

H/Pu	1000	2000	3000	
s	1.8	1.92	1.97	
a	0.0667	0.0267	0.0100	
d/d_0=0.1	0.857	0.940	0.977	Fc
d/d_0=0.01	0.735	0.884	0.954	Fc
d/d_0=0.001	0.63	0.831	0.933	Fc

Fc increases towards 1 when the fissile concentration goes near the infinite critical concentration and/or the medium becomes very large. In the latter case, the reflector's effect is obviously not very appreciable (7b).

b-Practice

There are some moderating materials different from water, which give values of d_{minca} smaller than the regulatory norms in [5, 6]. R.D. CARTER showed this fact for mixed Oxygen and water at reduced density. This is confirmed in our first-mentioned study [1], which also finds the same result for the polyethylen CH_2, the teflon C_2F_4, and the graphite C at the usual densities.

These reduced density mediums mixed with non or little absorbing elements (O, Al) also give small d_{minca}.

III - D_{minca} Of Pu-C In Infinite Slabs

D_{minca} is calculated on homogeneous infinite slabs of Pu-C, for various moderating ratios C/Pu, by using the codes APOLLO-DTF IV (S_4) [9-11] and their associated libraries.

-With both sides reflected by 40 cm of concrete, d_{minca} = 0.036 g/cm².

- If there is 0.1 g/cm³ of Iron homogeneously scattered in the slabs, d_{minca} increases up to 0.189 g/cm².

IV - D_{minca} Of Pu-C In 100 l Steel Drums

The reactivity of planar infinite arrays of many tiers of drums, reflected by 40 cm of concrete, is calculated by the three dimensional Monte-Carlo MORET code [12].

The drums are described as cubes.

The steel thickness of the walls varies from 0 to 0.105 cm.

The variations of some results are shown in figures 2 and 3.

The results of our calculations show that the previous value, without steel, of 0.036 g/cm² is confirmed. In other cases, d_{minca} increases almost linearly with the steel thickness of the walls. D_{minca} = 0.17 g/cm² for 0.105 cm of steel, when all walls are taken into account, and 0.137

g/cm² when the top and the bottom are not taken into account.

Figure 2

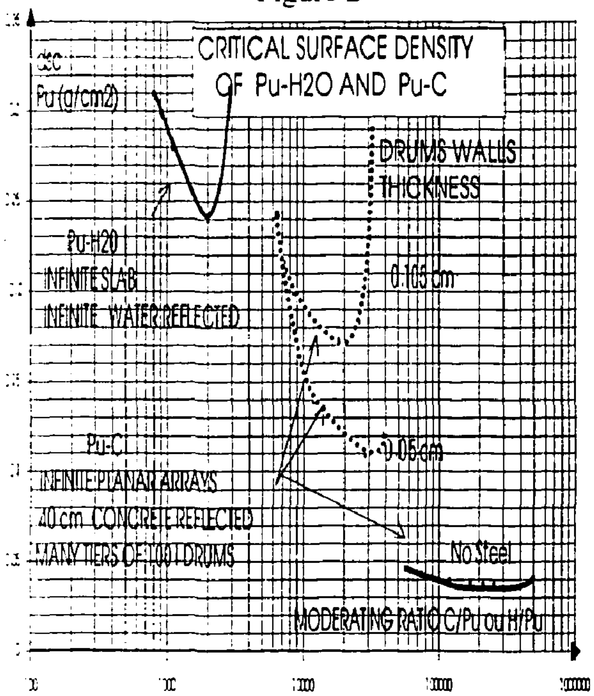
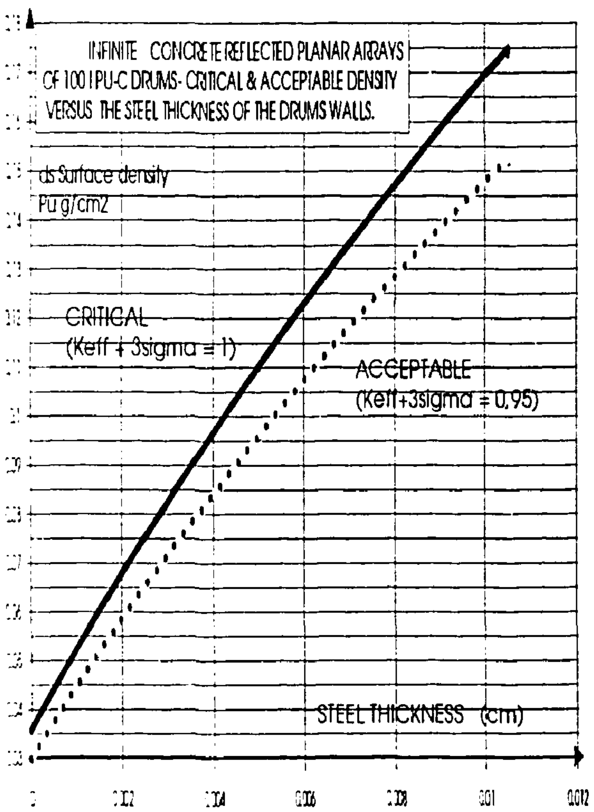


Figure 3



The results of our calculations show that the previous value, without steel, of 0.036 g/cm² is confirmed if all walls are taken into account, and 0.137 g/cm² when the top and the bottom are not taken into account.

It is noticeable that, at the same average concentration, the Iron (or Steel) absorption effect is roughly the same, whatever the form (homogeneously in the Pu-C or heterogeneously in the drums' walls).

This is due to the very large length of the neutron between its birth and its absorption, larger than the side of the cube, which is also the array pitch.

For a steel thickness of 0.07 cm, $d_{minca} = 0.14$ g/cm² and the acceptable subcritical regular value is $0.7 \times 0.14 \cong 0.10$ g/cm².

(According to the French safety usual rule, the acceptable value is the minimum one between $0.7 \times d_{minca}$ and d_s calculated for Keff + 3σ = 0.95).

V - SAFETY ANALYSIS

V.1 - Heterogeneity-Particles' Sizes

The graphite contamination by Plutonium is not homogeneous in the waste, but would rather be located on the outer surfaces of the graphite blocks, heterogeneously, and by little spherical specks of Pu or PuO_2, of variable density, from 19.6 for the metal to 1 for the apparent density of the PuO_2 powder.

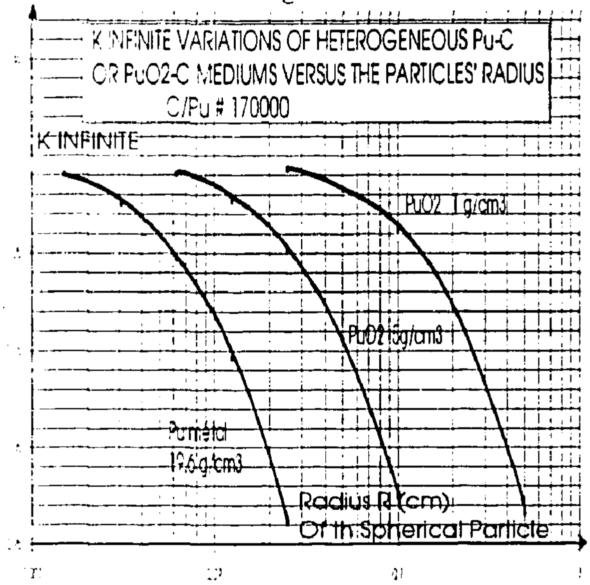
To observe this effect, some calculations were performed by using the APOLLO code to get the K infinite of arrays of little spherical particles coated in a graphite shell.

Figure 4 shows the variations of the K infinite near C/Pu = 170000, the d_{minca} optimum moderating ratio, versus the radius of the fissile particles.

The Kinfinite decreases from the homogeneous contamination (radius = 0) when the particles radius increases.

It also appears that, when the particles radius is larger than a certain value, depending on the density of the fissile contamination, the waste is subcritical (K infinite < 1).

Figure 4



V.2 - Case of a Real Storage

a-General Description.(figure 5)

The outer dimensions of the drums are :

$\Phi = 46$ cm $H = 66$ cm $V \cong 109$ l.

The steel thickness is guaranteed ≥ 0.07 cm.

The drums are gathered in group of five on three tiers on 120 cm x 120 cm pallets.

The maximum amount of Pu per drum is 100 g, giving in these geometrical conditions a value $d_{ss} = 0.10$ g/cm^2 when projected on the surface pallet.

The maximum amount of graphite is limited up to 100 kg.

The waste is wrapped in sheets of PVC or EVA. (Poly Vynil Chloride and Ethyl Vynil Acetate) PVC contains Cl. EVA does not contains Cl.

The practice showed that the wrapping does not exceed 4500 g.

b-Calculations

The calculations are performed by using the MORET code on the general description of figure 5 modelling an infinite planar array of 120 cm x 120 cm pallets bearing 5 drums on three tiers.

The bottom and the top are concrete reflected.

The Keff standard deviation is $\sigma = 0.003$.

Various cases are studied.

c-Wrapping not taken into account

Each drum is completely and homogeneously filled with the Pu-C medium containing 100 g of ^{239}Pu at a density of 19.6.

This description is more reactive than that where the Pu-C is supposed as a sphere in the middle of the drum.

The reflexion is got by the concrete albedo at the ground level and at the top at 2.50 m height.

The results are given in table II versus the moderating ratio for various amounts of graphite.

Table II

Real Drums Storage-Keff versus C/Pu

100 g Pu/Drum

C/Pu	graphite d	graphite Kg/drum	Keff (σ=0.003)
39070	1.8	196	0.806
34700	1.6	174	0.793
21700	1.0	109	0.697
13000	0.6	65	0.580

These calculations are performed with the 16 energy groups Hansen & Roach cross sections.

When the calculations are performed with CEA 86 APOLLO cross sections, the results are lower.

The favourable effect of the 0.07 cm steel thickness is roughly 0.25000.

d-Graphite wrapping and moderation between drums taken into account

In the same description as previously, calculations are performed for 100 g Pu, 100 kg graphite, and 450 g Hydrogen per drum, the Hydrogen amount corresponding to its 10 % maximum ratio in EVA.

The results are given in the table III versus the water mist density between the drums.

Table III

Real Drums Storage-Keff versus d Mist

100 g Pu / Drum

d water	0	0.01	0.05	0.02
Keff	0.696	0.696	0.672	0.52

e-Pu-C Waste Drums with 135 g of Pu

-We have to store in the same place hydrogeneous waste drums. Their acceptable surface density is 0.14 g/cm². We proposed then to create two distinct zones for each specific type of waste. This proposal was not accepted by the fire safety specialists who demanded that the graphite drums, which have a high calorific power, be mixed with the others, less penalizing on this safety aspect.

Thus the following study was conducted with a Pu amount different from that is taken for the norm determination.

Calculations are performed with 135g of Pu per drum, (C/Pu ≅ 14750) corresponding to a surface density d_{ss} = 0.14g/cm² which is the acceptable regular norm for the usual $Pu\text{-}H_2O$ wastes.

(note that this norm does not take into account the steel walls effect).

As formerly, there is a water mist of variable density between the drums.

The table IV gives the results versus the mist density.

Table IV
Real Drums Storage.Keff versus d Mist
135 g Pu / Drum

d Water	0	0.01	0.05	0.10	0.20
Keff	0.760	0.800	0.805	0.750	0.617

- Complementary calculations are performed with the central drum accidentally containing 250 g of Pu.

This mass is described as $Pu\text{-}H_2O$ sphere (radius 12.66 cm) optimally moderated (H/Pu = 900, CPu = 29.4 g/l), placed at the middle of the drum. A water shell of variable thickness surrounds this fissile sphere.

The other drums are homogeneously filled with 100 g of Pu and 100 kg of graphite.

The table V gives the results versus the thickness T of the water shell.

Table V
Real Mixed Drums Storage
Keff versus Water Shell Thickness

T(mm)Water	0	10	20	40
Keff	0.832	0.885	0.89	0.813

Further,in the case of the maximum Keff of the former array,some calculations are performed with a water mist in the free spaces between the drums.

The table VI gives the results versus the density of the water mist.

Table VI
Real Mixed Drums Storage
Keff versus d Mist

d Water	0.00	0.01	0.05	0.10
Keff	0.885	0.877	0.856	0.832

V.3 - Poisoning Elements in the Wastes Bulk

In the bulk, mainly in the wrapping, there are N and Cl as absorbing elements.

It has been shown the favourable effect played by the Iron, which has a thermal absorption cross section σ_a = 2.55 barns [13].

It will be the same, roughly proportionnally with the respective amounts and cross sections, for N (σ_a = 1.85b) and Cl (σ_a = 33.2b). Their presence will decrease the Keff.

V.4 - Oher Favourable Parameters

The Plutonium is not only ^{239}Pu but there is also, in variable amounts, a little of ^{240}Pu which is known to decrease the reactivity of the moderated fissile material.

The norms are got on a cubical description of the 100 l drums, without leakage betwwen them in the arrays, then for a more reactive geometry than the cylindrical drums reality.

Nevertheless, the absorbing effect of the steel is the same in both descriptions because the total wall surfaces have approximately the same value of 13000 cm².

VI-CONCLUSION

VI.1 - Theoretical Study

For us, in contrary of recent US analysis [14], graphite as a moderator is of great importance. Versus the d_{minca}, C plays defavourably as other non absorbing moderators different of the water : in fact, d_{minca} of Pu-C is 6 times lower (0.036 g/cm²) than the Pu-H_2O one, which is ordinary used to get the safe sizes of poor waste storages.

Happily, the drums' steel walls have a favourable absorbing effect, since d_{minca} increases up to 0.137 g/cm² for a steel thickness of 0.105 cm (taking only into account the drums' side walls).

By using the d_{minca}, one should care and take into account a corrective factor, because d_{minca} is not a constant independent of the fissile concentration when the mediums are reflected.

But this corrective factor goes towards 1 for the small concentrations and for the large mediums, where the reflector is meaningless.

Thus, for Pu-C wastes, stored in steel drums, the decided regular norm is 0.1 g/cm², if the minimum thickness of the walls is guaranteed larger (or equal) than 0.07 cm.

VI.2 - Practical Study

The real conditions of the studied storage (Pu < 100 g , C < 100 kg , five 100 l drums in three tiers on 120cm x 120 cm pallets, steel thickness > 0.07 cm) are so, that the norm is fulfilled : The calculations performed for checking the criticality safety show values of Keff + 3σ sensibly lower than 0.95.

The heterogeneity and the sizes of contaminating Pu particles decrease the Kinfinite of the fissile wastes.

Further, the presence of N and Cl, absorbing elements in the wrapping, is favourable for the safety.

Finally the mixing in the described storage of Pu-C drums at 0.10 g/cm² and Pu-H_2O drums at 0.14 g/cm² is acceptable.

ACKNOWLEDGEMENTS

We are pleased to say our gratitude to Sir L. MAUBERT, now retired who had us always given profitable advices.

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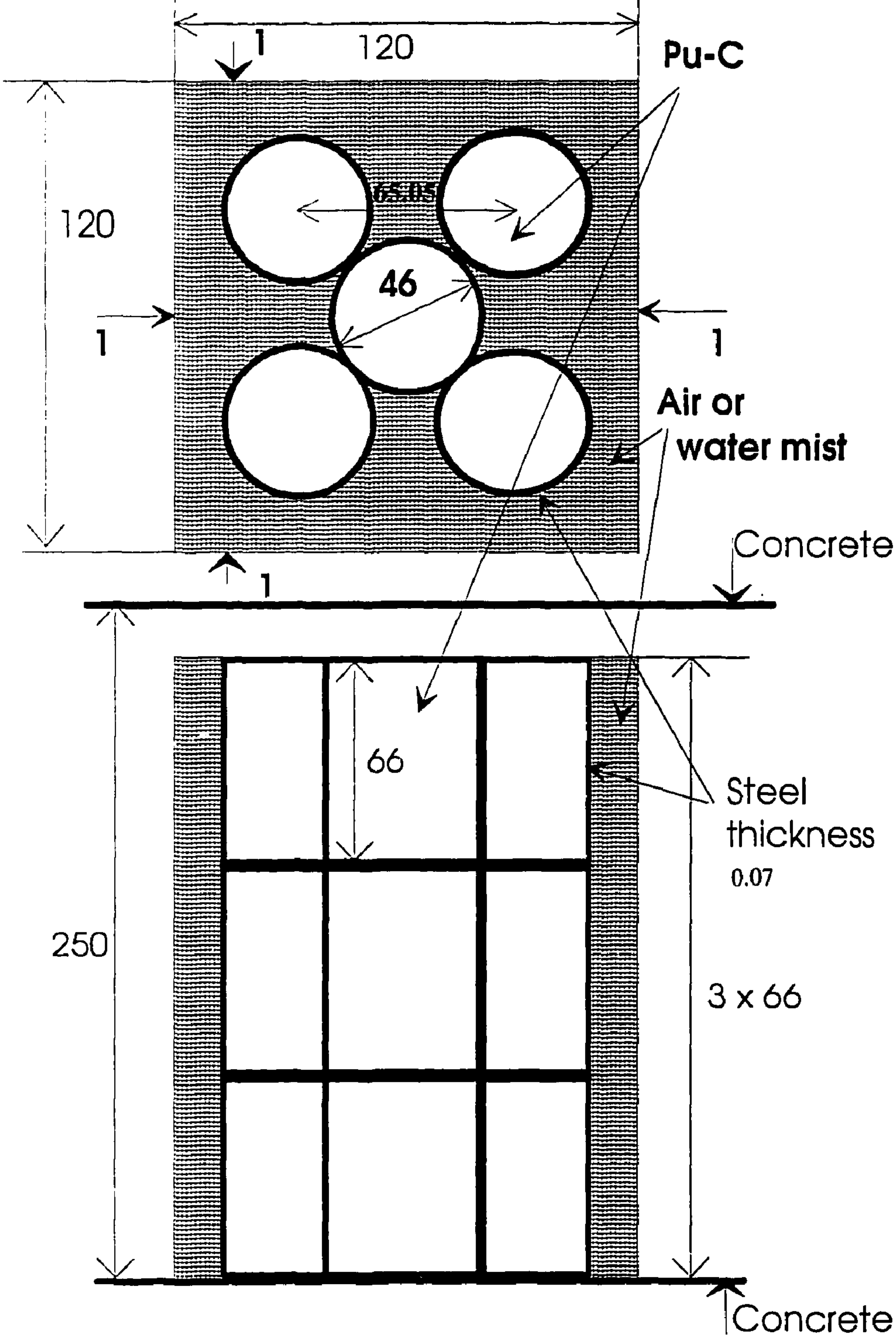
Figure 5

100 L PU-C WASTES DRUMS STORAGE GEOMETRY FOR MORET CALCULATIONS



Length and Thickness in cm