A MODEL AND CONDITIONS FOR FRAGMENTATION DURING MOLTEN FUEL/COOLANT THERMAL INTERACTIONS

L. CALDAROLA*, W.E. KASTENBERG**
Institut für Angewandte Systemtechnik und Reaktorphysik,
Kernforschungszentrum Karlsruhe, D-75 Karlsruhe, Germany

SUMMARY

The objectives of this paper are to present the thermal conditions necessary for fragmentation during molten/fuel coolant interaetions to take place, to present models based on these conditions for describing fragmentation and lastly to extrapolate to the case of molten UO_2/Sodium. The conditions and proposed model are based on experimental evidence as reported in the open literature.

Experimental evidence indicates that fragmentation may occur due to one of the following processes:

Process A. Highly superheated coolant vaporizes so quickly, that an energetic high pressure pulse is generated, which fragments the fuel. This high superheating may be caused either instantaneously because the liquid/liquid direct contact temperature is close to the homogeneous nucleation temperature or with a short delay because of the entrainment of coolant droplets inside the fuel.

Process B. Fragmentation is generated by successive coolant vapor bubble growth and collapse. The coolant microjets, which are produced during the coolapse, have sufficient kinetic energy to produce fuel fragmentation directly (similar to a cavitation process).

Process C. Similar to process B, but these microjets may have sufficient energy to penetrate into the molten fuel which leads to dispersion and entrainment of coolant droplets resulting in fragmentation as in Process A.

To initiate this process, several conditions must be met. These conditions are as follows

1. The temperature of the fuel must be higher than its melting point.
2. Liquid/Liquid direct contact must be established.
3. The liquid/liquid direct contact temperature must be higher than the melting temperature of the fuel, or lower but with a small solidification rate.
4. For Process A, fragmentation can take place without entrainment, provided that the contact temperature is close to the homogeneous nucleation temperature.
5. For Process B, a local superheated layer must be established, immediately following direct contact.
6. For Process B, the bulk temperature of the coolant must be lower than the saturation temperature which in turn must be lower than the contact temperature.

Which ever process takes place, there will be fragmentation, enhancement of fuel surface available for new liquid-liquid contact, mixing of fuel with liquid and between cold and hot liquid. For the case of UO_2/Sodium, the liquid/liquid direct contact temperature is lower than the homogeneous nucleation temperature. Hence for UO_2/Sodium, Process A should only occur with entrainment. Recent injection experiments show that high pressure pulses are observed when liquid sodium is injected into molten fuel, but are not present when molten fuel is injected into liquid sodium. This also occurs in the dropping experiments.

As a result of the interpretation of both the dropping and injection experiments, it is proposed that for molten UO_2/Sodium, fragmentation is governed by Process B. A mathematical model, based on the growth and collapse of vapor bubbles, is being formulated.

---

* *Euratom delegate to the fast reactor project Karlsruhe.*

** *Guest scientist from the University of California, Los Angeles, U.S.A.*

1. Introduction

In the theoretical analysis of fuel/coolant interactions, the fuel fragmentation process has generally been neglected because the mechanisms are not well unterstood. For this reason the approach that is generally followed, is to consider the mass of fragmented fuel as an input to the theoretical fuel/coolant interaction models. This mass may be considered to be instantaneously produced for a conservative safety analysis or as a function of the time, to fit experimental data.

The objectives of this paper are to discuss several relevant experiments, to present the thermal conditions necessary for fragmentation to take place, to present models based on these experiments and conditions, and lastly to extrapolate to the case of molten UO_2/Sodium. In addition, an experiment is suggested to verify the proposed mechanism for this letter case.

The equations governing the model will be described in a separate paper.

In the following sections we have called the hot molten materials (UO_2, Tin, Al etc.) by the word "fuel", and the cool liquid by the word "coolant". The symbols used in the text are defined in Section 7.

2. Description of some experiments appearing in the open literature

2.1. Argonne experiments (USA)

Many experiments have been performed at the Argonne laboratories over the past decade, dropping molten fuel into subcooled coolant (Ref. 1;2;4;5;6;9) and by injecting coolant into fuel (Ref. 3).

2.1.1. Tin and Gallium were dropped into water (Ref. 6;1;5) and are illustrative of the results obtained. The important parameters are:

a) Fragmentation "F" of the molten fuel, measured as surface area per unit of mass.

b) Temperature of the molten fuel, "T_f".

c) Temperature of the subcooled coolant, "θ_b"

d) Entrance velocity of the molten fuel into the subcooled coolant, v.

The conclusions appear to be as follows:

1. For given "θ_b" and "v", "F" increases with increasing "T_f" to a maximum and then decreases.
2. For given "v" and "T_f", "F" increases with increased subcooling, that is decreasing "θ_b". If "θ_b" is above 70°C, no extensive fragmentation occurs.
3. For given θ_b and T_f, F increases with "v".

2.1.2. In one series of experiments (Ref. 3 and 7) a small quantity of cool liquid was injected into a crucible filled with hot molten fuel. The materials were subcooled Na into molten UO_2. A general feature of these experiments was a very energetic pressure pulse as contrasted to the low energy pressure pulses oberved when one drops the same molten hot materials into the coolants.

2.2. C.E.G.B. Experiments (Great Britain)

A series of experiments (Ref. 8) involving molten tin and water were carried out. Most of these experiments are similar to those carried out at Argonne and give consistant results. However, there is one experiment which is different and very instructive. The experiment consisted of placing the hot molten material on a crucible (zero velocity) inside the subcooled liquid with "θ_b" higher than the threshold temperature "θ_{th}" of 70°C. In this configuration no interaction occurs. However, if the crucible is given a small mechanical impulse, the interaction occurs with extensive fragmentation.

A film shows that in all the experiments there is a multiple set of interactions with increasing intensity, characterized by the growth and collapse of vapour bubbles.

In another experiment the fuel was pre-dispersed. The interaction was very weak, so that it was concluded that pre-dispersal appears to prevent multiple interactions.

3. Models describing fuel fragmentation due to thermal effects and conditions for initiation of the fragmentation

As shown in the diagram of Fig. 1, it is proposed that fragmentation can occur due to one of the three following processes:

Process 1. Highly superheated coolant vaporizes so quickly, that an energetic high pressure pulse is generated, which fragments the fuel (Block A).
This high superheating may be caused, either instantaneously due to the fact that the liquid/liquid direct contact temperature "θ_c" is close to the homogeneous nucleation temperature "θ_h", or with a short delay because of the entrainment of coolant droplets inside the fuel (Fauske, Ref. 7 and 14).

Process 2. Fragmentation is generated by successive coolant vapour bubble growth and collapse (Block C and B). The coolant microjects, which are produced during collapse, may be shown to have sufficient kinetic energy to produce fuel fragmentation directly (similar to cavitation processes).

Process 3. Similar to Process 2, but these microjects may have sufficient energy to penetrate into molten fuel which leads to dispersion and entrainment of coolant droplets resulting in fragmentation as in Process 1. This path (Blocks C; G; A) is initiated in the same manner as Process 2 (bubble growth and collapse), but leads to the fragmentation Process 1. This path was suggested by the English C.E.G.B. (Ref. 8).

To initiate these processes several conditions must be met. The following conditions are common to all processes (Fig. 1).

1. The temperature, T_f, of the fuel must be higher than its melting point, T_{melt} (condition 1).
2. A liquid/liquid direct contact must be established between the

materials either with entrainment of coolant droplets into fuel (condition 2A) or without entrainment (condition 2B).

3. The liquid/liquid direct contact temperature "θ_c" must be either higher than "T_{melt}", or lower than T_{melt}, but with a solidification rate for the fuel so low, that only a very thin and weak crust at the surface of the unfragmented fuel is allowed to form during the interaction (conditions 3'; 3", 3"').

In addition the following conditions apply for Processes 1, 2 and 3.

4. Process 1 can take place also with liquid/liquid direct contact without entrainment, provided that the contact temperature θ_c is close to the homogeneous nucleation temperature "θ_h" (condition 4A). As discussed by Fauske (Ref. 14) there is experimental evidence that rapid vaporization due to homogeneous nucleation produces a shock wave, which may cause fuel fragmentation.

5. A necessary condition for Processes 2 and 3 is that the thermal energy, which is transferred from the fuel to the coolant, immediately following liquid/liquid direct contact, must not be distributed uniformly throughout the bulk of the coolant but most of it must remain in a local volume about the point of contact. This means that the rate of energy transferred to this volume must be much greater than the rate of energy removed to the bulk of the liquid (condition 4B). When the superheating reaches a given threshold, bubble growth starts. The larger the superheating, the larger the final bubble volume, and the larger the kinetic energy during collapse.

6. Another necessary condition for processes 2 and 3 is that the coolant bulk temperature "θ_b" must be lower than "θ_{sat}" (subcooled coolant), and "θ_{sat}" must be lower than "θ_c" (condition 5B).

   Whichever process takes place, there will be fragmentation, enhancement of fuel surface available for new liquid/liquid direct contact and mixing of fuel with liquid and between cold and hot liquid, so that the coolant bulk temperature "θ_b" will increase.

The enhancement of fuel surface may lead to further rapid heat transfer from fuel to coolant through two different feedback loops (Figure 1):

a) Loop No. 1; where the fuel is dispersed inside the liquid with subsequent bubble growth and collapse.

b) Loop No. 2; where the coolant droplets are dispersed inside the fuel during mixing.

In general this feedback process can occur many times until all the fuel becomes fragemented or one of the conditions discussed above is no longer satisfied.

4. Interpretation of the experiments

The experiments described in section 2 can now be analysed with the model given in section 3. For the Argonne experiments of paragraph 2.1.1. and for the C.E.G.B. experiments (paragraph 2.2.) there is visual evidence (high speed photography) that the interaction occurs with bubble growth and collapse.

In the Argonne experiments of paragraph 2.1.1., condition No. 2 of section 3 can be expressed by saying that the liquid/liquid direct contact is possible only if the fuel temperature "T_f" is lower than the Leidenfrost temperature "θ_{min}".

$$T_f < \theta_{min} \qquad (1)$$

In fact if inequality 1 is satisfied, stable film boiling does not occur and liquid/liquid direct contact becomes possible. Inequality 1 differs from that given by Kazimi (Ref. 13). Here the condition is $T_{melt} < \theta_{min}$, with which one does not succeed to explain some of the experimental results.

θ_{min} is a function of the bulk temperature "θ_b" of the subcooled liquid and of the relative velocity "v" between the two liquids

$$\theta_{min} = \theta_{min} (\theta_b; v) \qquad (2)$$

θ_{min} decreases by θ_b increasing (Ref. 11) and by "v" decreasing (Ref. 11 and 12)

The conclusions 1; 2; 3 of the Argonne experiments of paragraph 2.1.1. may be explained as follows

1. For given "θ_b" and "v", the fragmentation "F" increases first with increasing T_f, because "θ_c" increases with T_f, and therefore more thermal energy will be stored in the superheated liquid, which produces a higher degree of fragmentation. If now "T_f" continues to increase, condition 2 of section 3 is violated, that is inequality 1 of this section is not satisfied any more, and therefore the degree of fragmentation decreases rapidly.

2. For given "v" and "T_f", "F" decreases with "θ_b", increasing, because θ_{min} decreases and inequality 1 is not satisfied. The 70^oC threshold for θ_b can be explained in this way. However, if the vapour film is somehow broken (as in the C.E.G.B. experiments) the interaction takes place also if θ_b is above the 70^oC threshold.

3. For given "θ_b" and T_f, "F" increases with "v", because the contact surface area increases due to pure mechanical effects (Weber number criterion) and because "θ_{min}" increases with "v" favoring liquid/liquid direct contact.

   The influence of "v" on fragmentation is complicated by the additional effect of heat removed to the bulk of the coolant (condition 4B of section 3). Increased velocity tends to distribute the heat more uniformly, and therefore the liquid in the surrounding volume about the liquid/liquid direct contact point will store less energy in the superheated conditions, inhibiting the fragmentation. This effect may perhaps explain the delay between the time of fuel injection into coolant and the time of initiation of fragmentation. As fuel slows down, condition 4B is satisfied and the thermal interaction can occur.

The observation of the C.E.G.B. experiments (paragraph 2.2.) can be explained in the same way as the Argonne experiments, with the exception of the one in which the fuel was placed in a crucible inside the subcooled coolant with θ_b higher than the threshold value of 70^{o}C. This is at a condition in which inequality 1 is not satisfied, that is heat transfer with stable film boiling. In these conditions liquid/liquid direct contact does not occur. Evidently the small mechanical impulse is enough to break the stable film permitting induced liquid/liquid direct contact, which triggers the interaction. This experiment shows very clearly that inequality 1 (given by Fauske in Ref. 7) is not the most general condition for interaction. The most general condition is that liquid/liquid direct contact should occur, which can happen even if inequality 1 is not satisfied. Pre-dispersal of fuel appears to prevent multiple interaction, because condition 4B is violated. This is due to the fact that the thermal energy transferred to the coolant is more homogeneously distributed.

Let us now consider the Argonne experiments in which the subcooled coolant was injected into the molten fuel (paragraph 2.1.2.). Here the reaction takes place because all the conditions for Process 1 (entrainment of coolant droplets into fuel) are satisfied. This is clearly explained by Fauske (Ref. 7.).

Fauske (Ref. 7), explains the high energetic pressure pulses of the Al/H_2O thermal interaction (Refs. 9 and 10) as produced by the rapid vaporization due to the homogeneous nucleation of water, because $\theta c > \theta_h$ (condition 4A). On the other hand this condition is also satisfied for the case of Tin and water, which may be explained (as already seen) with a different model (Block C in Fig. 1). This last model seems more appropriate, because it is based on the visual evidence available to us from the C.E.G.B. More information is needed to decide which one of the two models is right. However, for the case of UO_2/Na interaction, since $\theta_c << \theta_h$, this lack of information should not be vitally important as we shall see in section 5.

5. Application to the UO_2/Na thermal interaction and conclusions

We have shown in section 3 that at present two feedback loops can be used to describe the fuel coolant thermal interaction.

In the case of the UO_2/Na thermal interaction we want to point out the following.

1. The liquid/liquid direct contact temperature "θ_c" is lower than the homogeneous nucleation temperature "θ_h" (Ref. 7), so that process 1 can only occur either through the loop No. 2 or through the loop No. 1 and Block "C" or through the initial entrainment of the coolant droplets inside the fuel.

2. Loop No. 2 postulates spontaneous entrainment of coolant droplets into fuel following mixing, which seems highly improbable, unless artificially induced as in the Argonne injection experiments (paragraph 2.1.2.).

   The high energetic pressure pulse observed during the injection experiments (against the low energetic pressure pulses of the dropping experiments) seems to support the hypothesis that the loop No. 2 can be excluded.

3. Since the high energetic pressure pulse of the injection experiments seems to be connected to the artificially induced entrainment of coolant droplets in fuel, and since no high energetic pressure pulse was oberved during the dropping experiments, one should conclude that Process 3 can be also excluded.

4. In an actual reactor configuration, the Na may be moving with sufficient velocity when it comes in contact with molten fuel (sodium reentry or unprotected overpower transient). For this reason initial entrainment of Na droplets in UO_2 cannot be entirely excluded. This means that Process 1 may only occur as a triggering mechanism.

Taking into account the above four observations, the schematic diagram of Fig. 1 without the dotted paths is proposed for the UO_2/Na thermal interaction. The diagram of Fig. 1 without the dotted paths in shown in Fig. 2.

In order to prove the validity of this proposed model the following experiment is suggested.

To put molten UO_2 at atmospheric pressure in a crucible, in a bath of liquid Na having a bulk temperature θ_b larger than the threshold temperature θ_{th} (which according to ref. 7 would be 770^0C, while according to ref. 15 would be 660^0C). At this condition no thermal reaction should take place, because the fuel temperature T_f is higher than the minimum temperature "θ_{min}" for stable film boiling. This inhibits liquid/liquid dirct contact unless film boiling is disturbed artificially.

This unstable equilibrium can now be broken either by a mechanical impulse, or by raising the ambient pressure or by injecting a small quantity of Na into the UO_2 , which triggers the interaction. If the proposed model is true, the following phenomena should be observed.

A) The reaction should occur with anyone of the three above mentioned trigger mechanisms, when the Na is very subcooled

$$\theta_{th} < \theta_b < \theta_{sat} = 880^0C$$

B) The reaction should not occur when the bulk Na temperature is very near to the saturation temperature of 880^0C, provided that the mechanical impulse method is used. If the injection method is used, only the first interaction, due to the injected Na, should take place.

C) When the Na injection method is used as trigger mechanism, the first pressure pulse should be much more energetic than the following pulses.

The authors are aware of the experimental difficulties to get fuel and Na in the conditions required by this experiment, that is to bring the system to stable film boiling. One of the difficulties is to maintain the surface temperature of the fuel during film boiling at the molten condition because the radiative heat loss through the film may be so large that the fuel surface may solidify in a very short time. However, this may be overcome by increasing the bulk fuel temperature. This temperature above melting may be too high in the case of UO_2. Therefore an alternative material with a lower melting point, such as Al_2O_3, may be used as fuel in order to perform the experiment.

Another indication of the validity of the model can be obtained by performing the experiment at various ambient pressures. Since the energy stored in a collapsing bubble is proportional to the pressure difference corresponding to the bulk temperature "O_b" of the subcooled liquid lowering the ambient pressure should produce less fragmentation.

6. References

1. Argonne National Laboratory Report, ANL-7399, November 1967.

2. Argonne National Laboratory Report, ANL-7152, January 1966.

3. Anderson and Armstrong, "Laboratory Test of Molten Fuel/ Coolant Interaction", ANS Trans. 15, No. 1, June 1972.

4. Anderson and Bour, "Experimental Tests of Vapor Explosions in a Molten Salt/Water System", ANS Trans. 1, No. 1, 1971

5. ANL 7245, July-Dec. 1967.

6. ANL 7800, July 1971.

7. Fauske, H., "On the Mechanism of UO_2/Na Vapor Explosions", ANS Trans. 15, No. 2, October 1972.

8. Borad, S.J., Farmer, C.L., and Poole, "Fragmentation in Thermal Explosions", CEGB Report No. RD/B/N/ 2423, October 1972.

9. S.M. Zivi and R.W. Wright, STL-327-30 (1965) TRW Report.

10. Darby, Pottinger, et al., "The thermal interaction between water and molten alluminium under impact conditions in a strong tube", OECD-Specialist meeting, Grenoble, January 1972.

11. L.C. Witte and P.J. Hennington, J. of Scientific Inst., "Identification of Boiling Regemes with a Reaction - Force Appratus", 1969.

12. F.J. Walford, " Trans Heat Transfer From Moving Nickel Spheres", Int. Journ. Heat Mass Transfer, Vol. 12, 1969, P. 1621.

13. Kazimi, "A Criterion for free-contact Fragmentation of hot molten Materials in coolants", ANS Trans. 15, No. 1, June 1972.

14. Fauske, H., "Nucleation of Liquid Sodium in fast Reactors", Argonne National Laboratory, Internal Report.

15. M. Farahat, "Transient-Boiling Heat Transfer from Spheres to Sodium", ANL-7909, 1972.

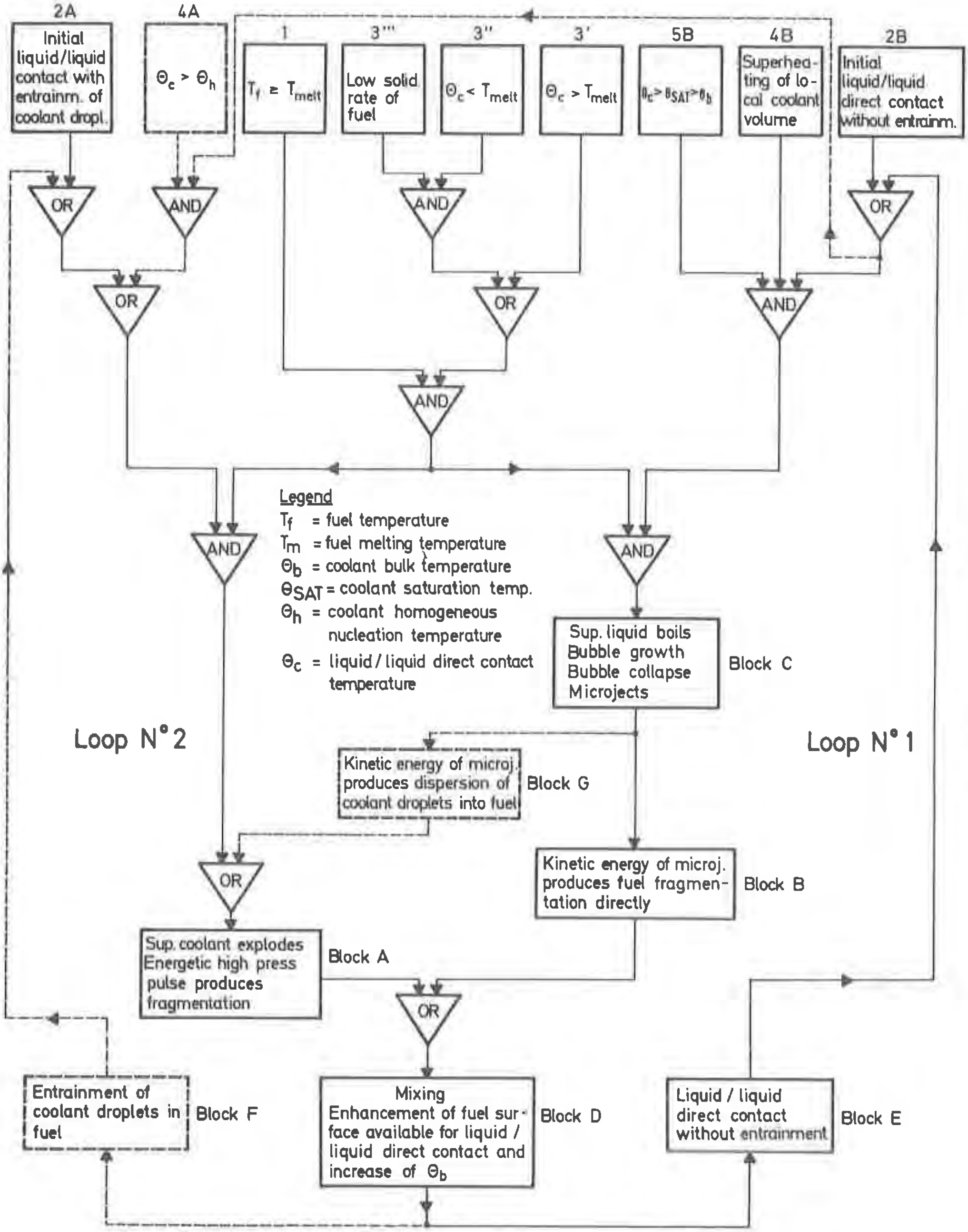

FIG.1 SCHEMATIC DIAGRAM FOR FRAGMENTATION PROCESS

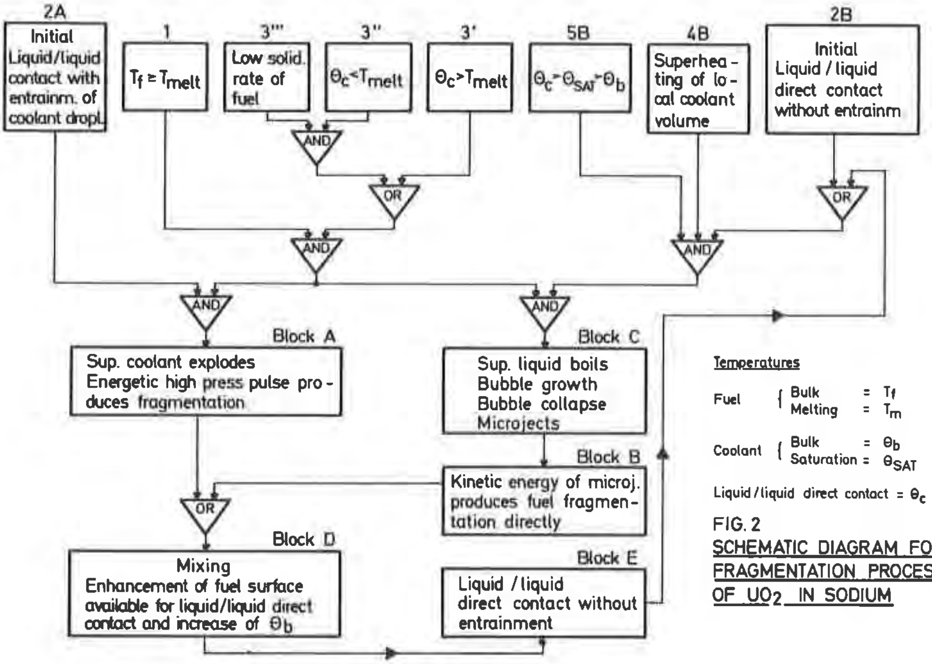

FIG. 2
SCHEMATIC DIAGRAM FOR FRAGMENTATION PROCESS OF UO_2 IN SODIUM

7. List of symbols

F = Fragmentation of the molten fuel, measured as surface area per unit mass

T_f = temperature of the hot molten material (fuel)

Θ_b = bulk temperature of the cold material (coolant)

v = relative velocity between fuel and coolant

Θ_c = liquid/liquid direct contact temperature

Θ_h = temperature at which boiling of the coolant fue to the homogeneous nucleation occurs

T_{melt} = melting temperature of the fuel

Θ_{sat} = saturation temperature of the coolant

Θ_{th} = bulk coolant threshold temperature for film boiling, that is the minimum coolant bulk temperature for which the condition $T_f > \Theta_{min}$ is satisfied.