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Thermodynamic Consequences of Sodium Spray Fires in Closed Containments

Part 1 — Experiments

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THERMODYNAMIC CONSEQUENCES OF SODIUM SPRAY FIRES IN CLOSED CONTAINMENTS

Part 1 - Experiments

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Summary

With respect to core disruptive accidents in LMFBR's liquid sodium might be sprayed with high pressure through the head of the tank into oxygen-containing atmosphere.

A series of large spray fire experiments has been performed under accident conditions in the FAUNA facility of LAF I in the KfK.

The experimental results showed that the overpressure did not exceed 1.8 bar at the experiment, spraying 60 kg Na in 1.5 seconds.

Zusammenfassung

Thermodynamische Auswirkungen von Natrium Spray Bränden in geschlossenen Containments

Bei schweren Unfällen in schnellen Brut-Reaktoren muß die Möglichkeit in Betracht gezogen werden, daß flüssiges Natrium unter hohem Druck durch den Tankdeckel in sauerstoffhaltige Atmosphäre gelangt.

Eine Reihe von Spray Brand Experimenten unter Unfallbedingungen sind in der FAUNA-Anlage des LAF I am KfK durchgeführt worden.

Bei dem größten Experiment, bei dem 60 kg Natrium in 1,5 Sekunden versprüht wurde, stieg der Überdruck im Containment nicht über 1,8 bar.

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Summary

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1. INTRODUCTION

With respect to core disruptive accidents in LMFBR's it can be assumed that relatively large amounts of liquid sodium will be released into the containment. Depending on the course of the accident sodium will be sprayed with high pressure through the head of the tank into oxygen-containing atmosphere. The resulting spray fire leads to temperature and pressure rise and influences the structure of the containment.

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In order to determine whether the integrity of the containment will be preserved, experiments and code calculations are necessary to predict the consequences (e.g. pressure rise and temperature transient) of such an accident. Calculations made up until now evaluate the mass of sodium ejected at 2400 kg in 1 second, and the ejection speed up to 30 m/sec.

The experimental studies carried out in the 220 m³ containment of the FAUNAfacility maintained the same ratio between sodium ejected and the amount of available oxygen, as well as the same relative flow rates as in the case of a core disruptive accident. The conditions are summarized and compared in table 1

2. Experimental set-up

2.1 The FAUNA facility

The FAUNA facility consists of a cylindrical steel containment with a volume of 220 m³, where large spray fires can be performed (Fig. 1). The containment can be evacuated; the overpressure inside can mount up to 3 bar. The wall temperature (max. 150 °C) can be controlled using a water cooling system with a flow rate of 60 m³ water per hour. A gas supply system to set up different atmosphere inside the containment. / 1/.

2.2 The sodium ejection device

The sodium ejection device consists of a stell tank with a capacity of 150

liters and is installed in an axisymmetric way inside the FAUNA-containment, as shown in picture 2. The tank is 150 cm high and is designed for an overpressure of 6 bar and for a sodium temperature of max. 550 °C. The conditions of the spray flow rate and spray pressure required a nozzle consisting of 271 holes of 4 mm diameter each /Fig. 3/. The nozzle was connected to a dip-pipe of variable length, so that the amount of sodium was exactly controlled and limited by the length of the pipe /Fig. 4/.

The Argon gas pressure supply to produce the spray was regulated by a special device which kept the difference between the pressure inside the spray tank and the pressure inside the FAUNA containment constant. This device is shown in Fig. 5. In a tank, 400 liters of Argon were stored at 10 bar. The pressure regulator R provided a pressure according to the pressure signal PS. This signal consists of the sum of the spray pressure chosen, added to the over-pressure inside the FAUNA containment.

2.3 Measurement devices and data recording

A fast data recording system is necessary to achieve a reasonable measurement with this type of experiment. All on-line data were recorded with a computer system, working at a speed of 220 channels per second.

For temperature measurements 19 thermocouples (chromel-alumel) of 1.5 mm diameter were used. In addition, some 0.5 mm thermocouples were used to check differences in response time.

The FAUNA containment pressure measurements were done using three independent pressure sensors which showed no remarkable differences. The overpressure inside the spray vessel had been measured for control only; the pressure regulation system worked automatically.

The oxygen consumption during the experiments has been determined by an oxygen analyser which had been modified to work pressure-independent inspite of the pressure-transient during the experiment.

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For analysing the chemical composition of the aerosols, filter probes were taken during the experiments. Size distribution measurements had been performed using impactors, installed inside the containment.

Two high speed cameras were used to observe the combustion during the spray. Both cameras were running at a speed of 800 frames per second, but the fast aerosol production allowed only a short observation time.

2.4 Experimental initial data

All the experiments had been performed in the containment of the FAUNA facility using the sodium ejection device already described. For all experiments

the	oxygen	concentration	(20.8	Vol.%)
the	sodium	temperature	(773 °	'K)
the	ejectio	on speed	(20 m/	(s)

remained unchanged. The other initial data are listed in table 3.

3. Results

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3.1 Pressure and temperature transient

3.1.1 Gas temperatures

The temperature measurements inside the FAUNA-containment had been done using chromel-alumel thermocouples with stainless steel shell. The positions of the thermocouples are shown in Fig. 6. The positions allow to distinguish three different zones of temperature: In the upper part of the containment the zone at the highest temperatures can be found. In this area, the burning sodium jet impinges the top plate (thermocouples Nos. 1, 2, 15, 16). A medium zone is represented by the thermocouples Nos. 4, 5, 12 and 13. The thermocouples near the bottom of the containment are especially influenced by downfalling (and partly burning) hot particles. In this zone, the lowest temperatures had been measured In figures 7 - 24 the typical gas temperature transients in the three zones are shown for all six experiments. The data are plotted originally without a calculated response time correction.

In the upper part of the containment a strong increase of temperature can be observed in the first seconds of the experiment. The peak temperature is reached in less than seven seconds in all experiments (Fig. 7 - 12). The highest temperature maximum is reached in the 60 kg-experiment after four seconds with nearly 1200 °C (Fig. 12).

During the experiments different thermocouples had been destroyed. In the experiment FS1, the thermocouples T1 and T16 failed (Fig. 7), as well as the thermocouples T4 and T13 in the experiment FS2 (Fig. 8), and the thermocouple T15 in experiment FS6 (Fig. 12).

No significant peak can be found in the medium zone of the containment (Fig. 13 - 18). Only experiment FS6 shows a flat maximum with about 800 °C (Fig. 18). During the transient, the level of temperature is proportional to the amount of sodium ejected (Fig. 31 - 33).

The thermocouples near the bottom of the containment show a temperature increase within some seconds to a relatively low, nearly constant temperature level (Fig. 19 - 24). Fig. 33 shows here also that the temperature level is proportional to the amount of sodium ejected.

From the whole set of figures it appears that the experiments FS2, FS3 and FS4 give results with similar levels. This means that with the experimental set up used the gap between the experiments is not important enough to be significant.

3.1.2 Pressure transient

One of the main goals of the experiments was the determination of the pressure transient and the maximum pressure inside the containment due to the burning sodium spray. Figures 34 - 39 show the course of the pressure transient for all

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six experiments. In the further discussion, experiment FS1 is not taken into account, due to the small amount of sodium ejected. This'experiment was a system test only and due to the very short ejection time (~ 0.12 sec) not really compatible with the ejection device.

In all experiments the maximum of pressure is reached within 4 seconds after the spray started. The dependence of the pressure maximum vs. the amount of sodium sprayed is shown in Fig. 40; the data are listed in Table 3. In average a pressure increase of approximately 0.57 bar per second can be seen in all experiments.

There are two experiments, FS3 and FS4, which were performed with the same amount of sodium, but different results had been observed. This phenomena will be discussed in chapter 4.3.2. In case the pressure course is plotted together for all experiments in one graph only (Fig. 41), it seems that the pressure increase is similar for all experiments. This fact will also be discussed in chapter 4.3.2.

3.1.3 Wall temperature

The outside wall temperture of the FAUNA containment had been measured during the experiments F4 - F6. There were especially two reasons for these measurements: First, the heat content of the wall might be an important data for recalculating the energy released by the sodium spray. Second, the course of the outside temperature of the wall might be useful to distinguish between heat radition and conduction inside the vessel. It should be mentioned that the sensor for the measurement of the temperature had been glued on the wall surface. Considering the general problems in the measurement of wall temperatures, the data might have some uncertainties. The course of the measurements is shown in Fig. 42 - 44. There are large differences in the temperature increase concerning these three experiments. Experiment FS4 shows a temperature increase of 5 degrees within 3 minutes, while FS5 gains 10.2 degrees and FS6 even 19.5 degrees within 3 minutes. The consequences of these results will be discussed in chapter 4.4. This behaviour is in agreement with the temperature level observed inside the vessel vs. the amount of sodium sprayed.

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3.2 <u>Results on aerosol physics</u>

3.2.1 Aerosol size distribution

Sodium spray fires, as described in this report, are a high intensive but short aerosol source. Considering the high aerosol mass concentration, the aerosol system changes rapidly. The real aerosol size distribution exists only a short time after starting the spray. Coagulation and sedimentation influence the size distribution already (in the order of) one minute after the spray. To perform the size distribution measurements two Andersen Mark III-Impactors had been installed inside the containment.

In the experiments with lower amounts of sodium sprayed (7 kg resp. 20 kg) particle sizes - aerodynamic mass median diameters - between 1.32 μ m and 2.15 μ m had been measured. In the experiment with 40 kg sodium sprayed, a particle diameter of 4.8 μ m had been measured 2 min after the spray started.

3.2.2 Aerosol mass concentration

As already mentioned, the high burning rate in spray fires causes a strong increase in aerosol mass concentration up to - in aerosol physics - unusual high values. The maximum value could be found in experiment FS6 (60 kg Na) with 112 g/m^3 two minutes after the spray started. On the other side, the high mass concentrations cause a high coagulation rate resulting in a strong sedimentation. A typcial fast decrease of aerosol mass concentration is shown in Fig. 45 for experiments FS3 and FS4.

3.2.3 Chemical composition

The experiments had been performed in ordinary atmosphere; therefore, there is a small amount of CO_2 and humidity available - during the first moment of a spray - for chemical reaction. But the main reaction products are sodium oxide (Na₂O) and sodium peroxide (Na₂O₂). It can be assumed that on the outside of the burning sodium jet sodium peroxide is formed immediately. In the inner regions of the jet - due to the lack of oxygen, more sodium oxide is formed.

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But the hot sodium oxide particles can react - if there is enough oxygen available - to sodium peroxide. The analyses of the filter probes also show that the main reaction product in these sodium spray fires is sodium peroxide.

4. Interpretation and Discussion of the Results

4.1 Oxygen consumption

Knowing the reaction product - Na_2O_2 - the oxygen consumption can be used to determine the sodium burned. Table 2 shows the oxygen consumption of the single experiments and the ratio of the sodium ejected and the sodium burned (reaction efficiency). The mean value shows that 67 % of the sodium burns. This is in good agreement with results from sodium spray experiments in Cadarache /2/, giving 52 %. An overview on the theoretically possible reaction and the experimental results is shown in Fig. 46. The oxygen consumption as volume percent of the FAUNA vessel is plotted against the amount of sodium reacting to Na₂O₂. The straight line gives the theoretical value, the crosses beneath show the experimental results. For comparison with the theoretical value, the dotted line gives the least square fit of the experimental data. The slope of this line is only 89 % of the slope of the theoretical curve. This reaction efficiency of 89 % by this evaluation is of course different from the above evaluated ordinary mean value. But due to the fact that higher amounts of sodium ejected mean a decreasing amount of oxygen available, this reaction efficiency can be assumed to be an upper limit under the described conditions.

4.2 Chemical composition

From sodium pool fires performed at KfK and CEA /3/ in open and closed containments, the chemical composition of sodium fire aerosols is well known. Usually, a mixture of sodium peroxide and sodium oxide forms the aerosols. The results from the spray fires presented here show that different sodium oxides can exist during the spray, but the main reaction product was found to be sodium peroxide Na_2O_2 . Therefore, sodium consumption, oxygen consumption and energy release can be calculated using the relation

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 $2 \text{ NA} + \text{O}_2 \rightarrow \text{Na}_2\text{O}_2$

with $\Delta H_{a} = 120.6 \text{ Kcal} (\cong 504.1 \text{ kJ}).$

4.3 Pressure and temperature transient

4.3.1 Gas temperatures

The gas temperatures inside the FAUNA containment are very much dependent on the location of measurement. Due to the fact that the main part of the sodium ejected reacts in the upper part of the containment, there is a high temperature gradient between the top of the containment and the bottom (Fig. 25 - 30) The temperature differences can be up to some hundred degrees. The maximum of temperatures in the upper zone is reached always before the maximum of the medium and lower zone. These maxima are obtained when ejection is already stopped. This means that an energy exchange occurs beween the various zones in order to homogenize the temperature. Fig. 47 show that approx. 300 s are necessary for a nearly homogeneous mixing of the temperatures. This fact settles that important convective movements occur in the vessel. These movements could be used for oxygen transportation from rich to depleted oxygen zones.

It is found that the temperature distribution is axissymmetric, according to the axissymmetric set-up. The time resolution for the temperature measurements is mainly given by the thickness of the thermocouples. Due to the severe conditions inside the containment during the spray, the thermocouple diameters cannot be too small. It was found that 1.5 mm ϕ is a suitable diameter, but for comparison some 0.5 mm ϕ thermocouple had been used. In Fig. 48 the data of a 1.5 mm ϕ and of a 0.5 mm ϕ , located in a 1 cm distance, are shown.

In all cases, no higher temperature than approx. 1200 °C could be observed. This is in agreement with the flame temperature of sodium pool fires. But it should be mentioned that the consequences of sodium spray fires are mainly determined by the maximum pressure peak, which occurs some seconds earlier than the temperature peak.

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4.3.2 Pressure transient

For discussion of the pressure transient it is necessary to look again at the experiments FS3 and FS4. In both experiments, with the same amount of sodium sprayed, different pressure transients have been measured. Fig. 40 shows that there is not only a difference in the maximum pressure, but also the pressure increase itself is different. Because there is no remarkable change in performing the two experiments it must be assumed, that this different behaviour is a "system effect". This means, that experiments performed unter the described conditions, have a certain range within which the results can lay. Due to this fact it is valid to plot the pressure transient in this way together; so one general pressure increase is shown (Fig. 50). Using this kind of evaluation it is possible to calculate an average value of the pressure gradient of the experiments FS2 FS6. Some points of this average values are plotted in Fig. 51 together with the least square fit line. This evaluation shows a gradient of .573 bar/s.

4.4 Energy balance

For the energy balance of sodium spray fires, several phenomena have to be taken into account. During the spray time, the burning sodium (flame temperature up to 1350 °C) is leading to a sodium heat up and a heat radiation. From the sodium flow rate ejected the level of the heat flow rate, produced by combustion, can be evaluated assuming an efficiency of 50 % and peroxide formation to be

$$\dot{Q}_{c} = 340 \cdot 10^{6} \text{ J/s}$$

This value must be compared with

- the heat transfer by radiation to the walls
- The heat transfer to the gases
- the heat transfer into the sodium.

The heat exchange by radiation is calculated using the model of two long coaxail cylinder. Under these conditions the heat flow rate is:

$$\varphi = \frac{\frac{S_s \sigma (T_s^4 - T_p^4)}{\frac{1}{\varepsilon_s} + \frac{S_s}{S_p} (\frac{1}{\varepsilon_p} - 1)}$$
 E 1

where

to be

$$S_s = sodium column area$$

 $S_p = wall area$
 $\varepsilon_p = sodium emissivity$
 $\varepsilon_p = wall emissivity$
 $T_s = sodium temperature$
 $T_p = wall temperature$
 $\sigma = Stephan-Boltzmann constant$

In order to evaluate the importance of radiation the heat flow rate is maximised, to be sure it is the envelope of the real heat flow rate. In particular it needs that the denominator of equation (E 1) is minimum, which can be achieved if every term is minimum. At first the ratio of the surfaces s_s/s_p is calculated with

$$S_{p} = 94 \text{ cm}^{2}$$

 $S_{s} = 6.3 \text{ m}^{2}$
 $S_{s}/S_{p} = 0.067$

The inside wall of the FAUNA consists of a rough and dark surface, so the emissivity $\varepsilon_{\rm p}$ is assumed to be 0.9. It follows, that the second term of the denominator is equal to 0.007. The minimum of the first term is equal to 1, that means that the second term (equal to 0.007) can be neglected regarding the first term.

Therefore, the heat flow rate can be expressed by

$$\varphi = \varepsilon_{s} \cdot \sigma \cdot S_{s} (T_{s}^{4} - T_{p}^{4})$$

To get a maximal heat flow rate let $\varepsilon_s = 1$, using $\sigma = 5.67 \cdot 10^{-8} \text{ W/m}^2 \text{ K}^4$ and $S_s = 6.3 \text{ m}^2$

The following table can be derived:

Experiment		FS 4	FS 5	FS 6	
T	°K	1623	1623	1623	
T_	°C	272	279	287	
φ φ	J/S	2.477 • 10 ⁶	2.476 • 10^{6}	2.476 • 10 ⁶	

Compared with the heat flux derived from the sodium combustion the radiation contributes less than 1 %, which means that radiation can be neglected for the energy balance.

The heat transfer into the gases can be derived from the slope of the pressure increase. This slope is similar in all experiments compared and is found to be

$$\dot{Q} = 44.26 \cdot 10^6 \text{ J/s}$$

which is 13% of the combustion energy.

The remaining 87% of the combustion energy is capable of heating up the sodium near to its boiling temperature.

As soon as there is no more burning sodium in the gases, the main phenomena are heat transfer to the walls and equalisation of the temperature inside the vessel. These effects are much slower than the phenomena described before. The heat up of the walls is taken from the wall temperature measurements. It must be taken into account, that during the time to the wall temperature maximum (e.g. 19 minutes in experiment FS6) also heat transfer from the outside of the wall to be ambient atmosphere takes place. This means, that only a part of the energy released can be found from the wall temperature. The calculation shows, that the main part of the energy of combustion (\sim 80 %) is found in the walls, and it can be reasonably assumed, that the remaining part is shared between the inside gas temperature and the outside wall losses (Table 4).

5. Conclusions

The knowledge derived from these experiments is only valid for this type of fire involved (i.e. large sodium flow rates and short ejection time).

It has been shown that the maximum of the overpressure is reached around three seconds after the ejection stops. In the range studied, the value of the maximum overpressure is dependent on the amount of sodium ejected. Moreover this maximum is far away from the theoretical value, estimating that the whole combustion energy is transfered immediately to the gases. The experimental results confirm the theoretical model used in PULSAR. The difficulty remains to express the initial energy release rate to calculate a hypothetical spray fire, for a low relative error in this term may cause a large absolute error on the maximum overpressure.

6. Literature

- /1/ W. Lindner, J. Kind: Beschreibung der Forschungsanlage zur Untersuchung nuklearer Aerosole (FAUNA), KfK 3011
- /2/ J. Charpenel et al.: Feux de sodium pulvérisés, Rapport SESTR N° 80/53 CEA Cadarache
- /3/ W. Cherdron, S. Jordan: Physical and chemical characterisation of sodium fire aerosols, LMFBR Safety Topical Meeting, 19-23 July 1982, Lyon

Table 1: Experimental Conditions

		Reactor HCDA	KfK Experiment	Up Scale
Volume	m ³	6500	220	1/30
Flow rate	kg/sec	up to 2300	56	1/40
Ejection time	sec	1	0,12 - 1,0	1
Mass ejected	kg	up to 2300	7-60	1/40
Ejection speed	m/sec	5-40	20	1
0_2 concentration	Vol%	21	21	1
Sodium temperature	°C	500-600 .	500	1

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Table 2: Oxygen consumption

Experiment	Oxygen concen- tration after Experiment Vol%	Total amount of gas (moles) final	Oxygen (moles) final	Oxygen consumed (moles)	Sodium burnt kg	Ratio Sodium burnt / Sodium ejected	
FS 1	20,05	8943	1814	86	3,96	0,56	
FS 2	18,3	8773	1623	277	12,74	0,64	
FS 3	17,6	8677	1527	373	17,15	0,57	
FS 4	17,2	8635	1485	414	19,08	0,64	
FS 5	13,4	8256	1106	794	36,5	0,91	
FS 6	. 12,2	8144	994	906	41,7	0,70	
mean value = 0,67							

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Experiment	Initial pressure 10 ⁵ Pa	Initial gas temperature °K	Nitrogen % volume	Water vapour % volume	Initial wall temperature ° K	Ejection time s
FS 1	0,998	296	77,7	1,6	291	0,12
FS 2	1,008	297	77,1	2,1	286	0,36
FS 3	0,978	272	78,6	1,8	270	0,53
FS 4	1,004	273	78,7	0,5	272	0,53
FS 5	0,977	279	78,8	0,4	279	0,71
FS 6	0,999	287	78,4	0,8	287	1,0

Table	3:	Experimental	Initial	Data
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For all experiments:

Initial oxygen / 7 volume 7: 0,208

Sodium temperature $/ \ ^{\circ}K / \ : 773$

Ejection speed / m/s_7 : 20

Table	4 :	Energy	balance

Experiment	Combustion Energy E ₁ J_7	Wall temp. increase A T (°K)	Energy E ₂ from wall temperat. <u>/</u> J_7	Ratio ^E 2 ^{/E} 1	gas tem- perature / ⁻ °C_7	Energy E ₃ from gas temperat. <u>/</u> J_7	Estimated wall losses E ₄ J_7	E ₂ +E ₃ +E ₄	Energy balance deviation <u>/</u> 7_7
FS 4	180 • 10 ⁶	8,25	81,5 • 10 ⁶	0,45	130	34 • 10 ⁶	19 • 10 ⁶	135 • 10 ⁶	34
FS 5	240 • 10 ⁶	16,7	163 • 10 ⁶	0,68	130	34 • 10 ⁶	15,5• 10 ⁶	212,5• 10 ⁶	13
FS 6	340 • 10 ⁶	28,5	276 • 10 ⁶	0,81	150	34 • 10 ⁶	21 • 10 ⁶	331 • 10 ⁶	2,5
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FIG. 1 FAUNA-Overview



FIG. 2 SODIUM EJECTION DEVICE



FIG. 3 SPRAY NOZZLE



FIG. 4. SCHEME OF THE SODIUM EJECTION DEVICE



FIG. 5 SPRAY PRESSURE REGULATION SYSTEM









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27 —
















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42 —

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FIG. 27 EXPERIMENT FS3 : COMPARISON OF TEMPERATURES

43 –





— 45 —





















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WALL TEMPERATURE u Մ 10 4 EXPERIMENT ן) לי FIG.







| 6





TEMPERATURE [°C]



FIC. 48 THERMOCOUPLE BEHAVIOUR

— 64 —

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FIG. 51 AVERAGE PRESSURE INCREASE

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