Development, production and characterisation of LOVA gun propellants

W. Karthaus*, A.C. Hordijk*, C.A. van Driel*, and C. Schoolderman*

In developing gun propellants a number of aspects are of importance. Apart from the performance improvement, higher specific impulse and a lower flame temperature, requirements for safe production, storage and storage lifetime can be mentioned.

At TNO-PML gun propellant compositions have been developed and investigated over the years. A number of compositions have been evaluated for their energy content and ballistic performance. A selected number have been produced on a small scale and tested. Some compositions have been produced with a twin screw extruder and underwent full scale testing.

The paper describes process modelling and processing safety aspects of laboratory scale processing. Furthermore the internal ballistic and performance characterisation and safety properties of the LOVA propellants that are under development will be presented.

The paper ends with a summary of future developments at TNO-PML with respect to gun propellants and concluding remarks.

1. Introduction

TNO Prins Maurits Laboratory is one of the institutes of the Netherlands Organisation for Applied Scientific Research (TNO). Within the framework of TNO Defence Research, the principal mission is to support the Netherlands' armed forces in building and maintaining a cost-effective, strong defence by providing scientific and technical advice in areas such as: ammunition technology and explosion safety, protection against NBC and impact weapons, and weapon effects. The research work is mainly pursued for the Netherlands Ministry of Defence (MoD), although other clients such as foreign Defence contractors to fulfil their offset obligations with high-tech research contracts and civilian contractors for contract research, product design, and standard products account for more than 35% of the turnover.

The authors are working in the division Munitions Technology and Protection, where amongst others research on and development of advanced gun propellants takes place.

Three major drives may be distinguished in gun propellant development: a higher impetus, a lower flame temperature and a higher safety level. This led to the development of the new generations of gun propellants applying knowledge from solid rocket propellants. Some fifteen years ago TNO-PML started research and development efforts in this field.

The research and development path at TNO-PML consists of elements such as:

- formulation of the propellants,
- modelling the performance, combustion, internal ballistics
- small scale production for experimental characterisation of components and propellants concerning hazard and compatibility, thermal stability, particle and binder properties and rheology
- laboratory scale production, mixing, casting, pressing and extrusion,

---

Received : May 17, 2002
Accepted : August 26, 2002

*TNO Prins Maurits Laboratory
P.O. Box 45, 2280 AA Rijswijk, The Netherlands
TEL +31 15 284321
E-mail: Karthaus@pml.tno.nl
www.pml.tno.nl

- characterisation and classification of propellants, LOVA, IM, ageing and implications for existing gun systems

A schematic of the development path is presented in fig.1.

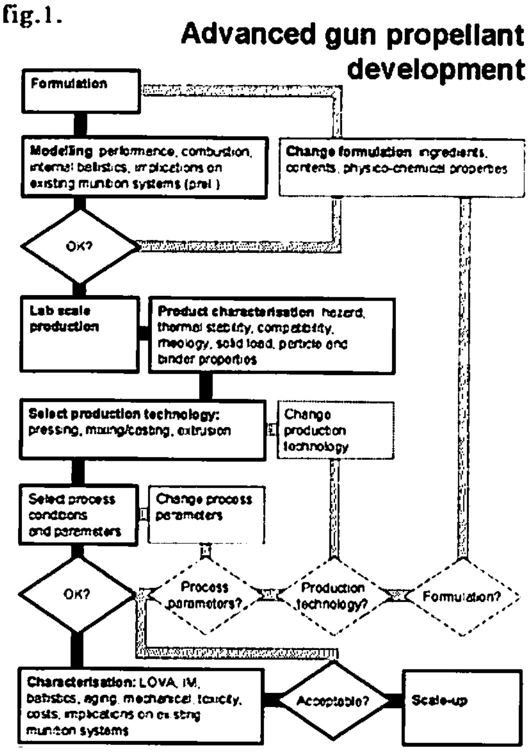

Fig.1 The advanced propellant development path.

The subject compositions in this paper are energetic materials in their paste like intermediate state. The materials tested on and presented in this paper are inert compositions (simulants) and energetic compositions. The energetic compositions are LOVA propellants based on TPE or HTPB as binders and RDX as energetic component.

Aspects of processing and processing safety, internal ballistic characterisation and performance and safety properties of the LOVA propellants that are under development will be discussed.

2. Laboratory scale production; extrusion

TNO-PML has long experience in research on castable energetic compositions such as rocket- and gun propellants and Plastic Bonded eXplosives[2,4]. Seven years ago extrusion was introduced as a production tool for these materials[1,3,5,6].

To build up experience on extrusion, the first products were TPE based propellant compositions. Compared to conventional propellants, this has the advantage that less solvent is needed. TNO-PML uses a small industrial size, twin screw extruder (TSE) to study conditions for safe production of energetic materials and to produce batches up to tens of kgs PBXs and gun propellants for performance and classification experiments.

Computer modelling of the process and essential extruder components supports the extruder work. Commercial available Finite Element Codes such as the Ludovic™ (twin-screw) and the Flow 2000™ (2D and Profile die) are used.

2.1 Processing hardware

Extrusion experiments are performed using a ram extruder, a 100 kN press, for preliminary tests with specific die and energetic composition combinations.

A Theysohn Maschinenbau TSK45 extruder, a 45 mm twin screw co-rotating self wiping machine with a screw length of 1305 mm, 7 barrel sections with independently controlled heating and cooling is used for production (see fig. 3). The throughput can be varied from 2 to a maximum of 45 kg/h. Temperature and pressure are measured at two places along the screw axis above the mixing and kneading blocks and in the extruder head before the shaping element.

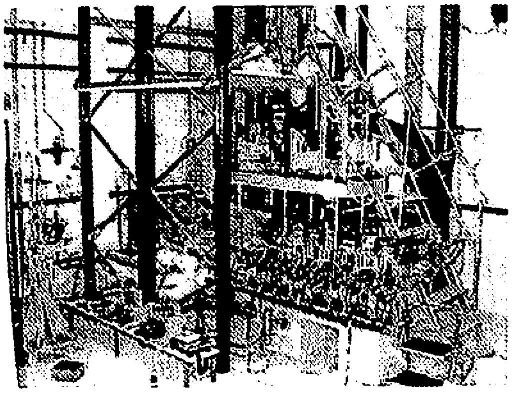

Fig.2 The co-rotating twin screw extruder including flight of steps and safety feeding unit.

2.2 Processing safety

In an extruder the feeding, mixing and shaping of the material is performed in one complex processing step, of which during the kneading and shaping highest pressures and temperatures are expected. This was confirmed during extrusion trials with TPE's and inert filler material. It appeared that pressure and temperature can rise quite high, especially extrudate temperature could rise far above the safety level. Strict safety requirements for pressure and temperature need to be met for safe production of energetic compositions. A method to predict pressures and temperatures of the whole extrusion process before the actually processing of a certain type of energetic formulation can be carried out is mandatory for safety reasons[3].

Material sensitivity tests and computer model simulations are part of the procedure.

In the R&D path of potential compounds or formulations several tests with respect to safety are performed in order to determine the usefulness of the continuation of the research process. Knowing the safety features of the various ingredients a small amount of the product is sufficient to obtain processing (rheological) data and perform small scale tests on: impact and friction sensitivity, thermal stability (DSC) and compatibility (VTS)[1,7].

When satisfying answers have been obtained, the following step is performing substances tests like the shock sensitivity, either by a small scale test like the MAP test (sample mass of about 30 g) or by a larger scale test like the gap test (sample mass hundreds of grams).

If the candidate formulation has successfully passed these experiments the steps towards production by means of the extruder is made.

2.3 Model simulation of the extrusion and shaping process

As part of the method to predict pressures and temperatures of the whole extrusion process before the actually processing, model simulation can be used. Simple models based on the Benbow[10] approach predict only pressure values and are therefore insufficient. Computer models that can simulate the more complex parts of the extrusion process are therefore required. The simulation software in use at TNO-PML are Flow2000™ Version 5.00 of Compuplast International Inc. and Ludovic™ version 2.2 of Sciences & Computer Consultants[11]. The Ludovic code is used to simulate the feeding and mixing processes in the extruder and with the Flow2000 code the shaping process can be simulated.

The rheological properties of the energetic compositions in paste like status, necessary as input data for the computer models, are determined with a twin bore Capillary Extrusion Rheometer (CER, Bohlin-Rosand type RH-7)[5,6,9,11].

The Ludovic™ code is used to study the effects of filler ratio and the solvent fraction on the behaviour of the extrudation process, where pressure and temperature are key parameters. The extruder screw build-up, rheological data, including temperature dependent viscosity was varied[11].

The Flow2000™ code is primarily used for die design studies. The influence of the die land (L/D) on the extrudate temperature and head pressure has for instance been determined[9,11].

As an example of a Flow2000™ application, die pressure simulations, compared with actual measurements will be presented in the next paragraph.

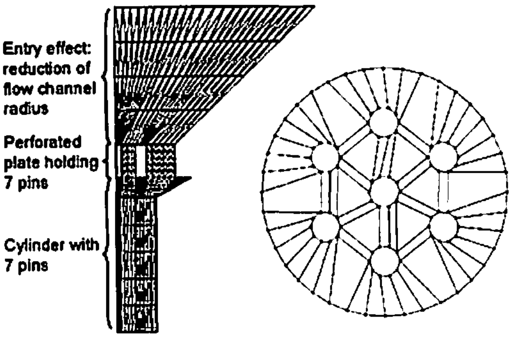

Fig.3 Example of a FEM grid for a 7-hole grain die design.

2.3.1 Die pressure

The flow through the die is simulated with the Flow2000™ code. The flow domain of the die is drawn and divided into large contours. The code generates a FEM grid inside the contours. See fig. 3. In an iterative process the code adjusts the FEM grid slightly during the calculation process until the total calculation error in the balance equations at the nodal points is less than 1%.

The material flow is assumed to be incompressible, Newtonian and isothermal. Gravity is ignored. The total pressure and

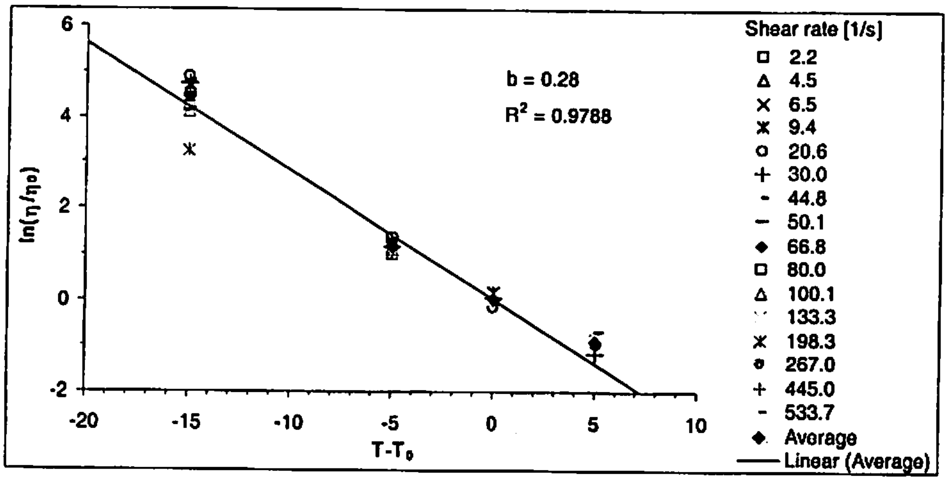

Fig.4 Estimate of the temperature dependency b = 0.28 at a reference temperature of 65°C for Wax from CER rheometer measurements.

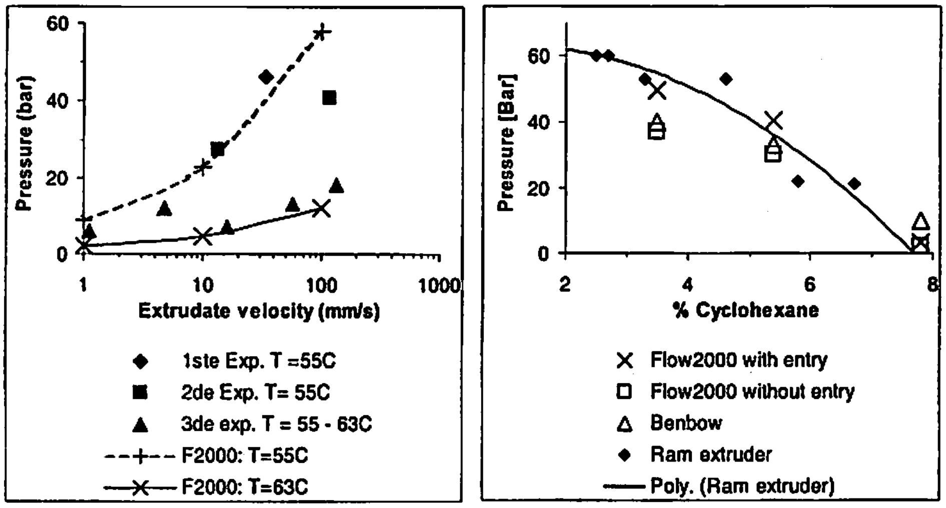

Fig.5 Experimental and calculated pressures for ram extrusion experiments through a 7-hole die as a function of extrudate velocity at different temperatures for a simulator paste (left) and solvent percentages for a TPE-based propellant paste (right).

temperature distribution is calculated in two 2D cross sections. As further input, a model fit of the rheology and the physical properties of the material are required. The rheology, shear rate and temperature dependency of viscosity η, is assumed to be exponential:

$$\eta = \eta_0 \cdot \exp(-b \cdot (T - T_0)) \qquad (1)$$

The temperature exponent b is measured over a wide shear rate and temperature range with the CER. The results for a wax are presented in fig. 4. Estimate of the temperature exponent b=0.28 at a reference temperature of 65°C. The model fits the measured data ±16% average for a temperature range of 50 to 70°C and a shear rate range of 2.2 to 534 s^{-1}. Determined values of b for TPE/sugar and a TPE-based propellant composition (IBK1010) are 0.044 and 0.02 respectively[11].

Ram extrusion experiments have been performed at different temperatures with a wax material to test a 7-hole die design. The ram pressure is measured as a function of extrudate velocity. These experiments have been used to validate the Flow2000™ model assumptions.

The predictions for the wax material correspond quite well with the ram extrusion experiments as can be seen in fig. 5 (left)[9,11].

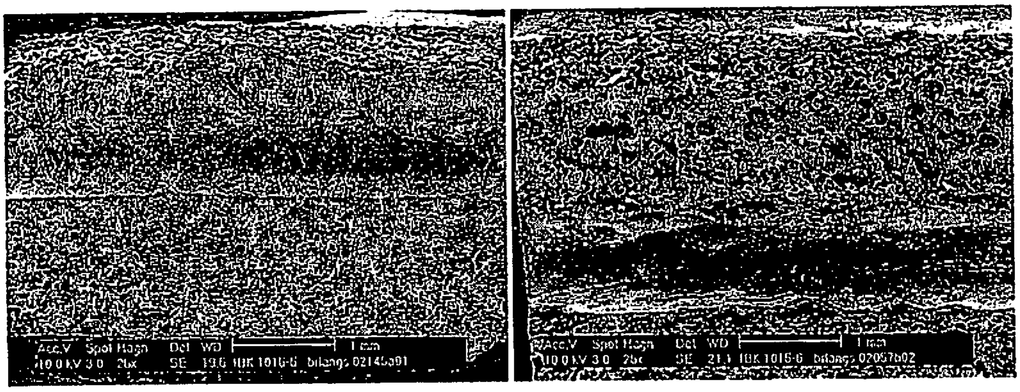

Fig.6 Photographs of propellant grains that are split across the burning channels (left: 'good' product IBK10150-5, right: porous grain IBK10150-6).

2.3.2 Live propellant test and simulation

Ram extrusion experiments with a live TPE-based LOVA propellant IBK1010 (composition TPE (SIS type) 16 wt %, RDX, 5 and 25 micron 83 wt %, additives 1 wt %, solid load 84 wt %) have been performed and simulated as well. The propellant has been shaped into a 7-hole grain through a die having a die-land length of 20 mm, a diameter of 7.50 mm and 7 pins with a diameter of 0.90 mm. During the shaping of the propellant the ram pressure (≈ die pressure) and the mass flow have been measured as a function of the solvent content. The results are presented in fig. 4 (right). The simulation results agree with the experimental results. The Flow2000™ results predict the pressure needed for shaping well and better than the simple Benbow model. The Benbow results are comparable with the simplified geometric model without the entry cone in Flow2000™. The entry cone causes a pressure build up due to a decreasing flow channel diameter.

Three safety criteria are established[3] for TSE extrusion of the live propellant based on TPE and RDX. Die head pressure should remain below 7 MPa, shear stresses must be under 150 kPa (rheological criterium) and the die head temperature increase should be smaller than 10°C. The solvent secures proper paste flow and suppresses excessive heating in the die. The solvent mass input should be about 10 wt% resulting in a product containing a solvent content between 4 wt% and 9 wt% to guarantee the process will occur within the safety limit values[3]. For more details see ref.11.

3. Characterisation

3.1 Performance characteristics

The performance of the produced propellant batches is determined by means of standard 700-ml Closed Vessel (CV) tests. The test results are used to determine the vivacity and the linear burning rate of the propellants. These can be calculated using one of the modules of TIBALCO, the T_{NO} Internal BALlistic COde. This lumped parameter computer code is based on STANAG 4367, like the well-known IBHVG2-code. It is mainly set up to study the effects of various propellant property's on the internal ballistic cycle, like differences in inner and outer burning rate of propellant grains and ignition delays for different sub charges. TIBALCO is currently under development, but is already applicable for several basic grain geometry's[12].

The TIBALCO calculation method can also be applied for various other purposes. As an example the burning surface can be calculated in cases where the linear burning rate is known, for instance from other reliable measurements.

One out of two batches that where produced, which are essentially similar with respect to their composition, showed a rather large porosity, resulting from bad process conditions, see fig. 6.

The major difference between these batches is the length of the 7-hole grains (batch IBK1015-5: 20 mm and batch IBK1015-6:10 mm resp.). The burning characteristics in terms of vivacity are shown in fig. 7 (left).

The vivacity curve for batch IBK1015-5 has a shape, which might be expected for 7-hole

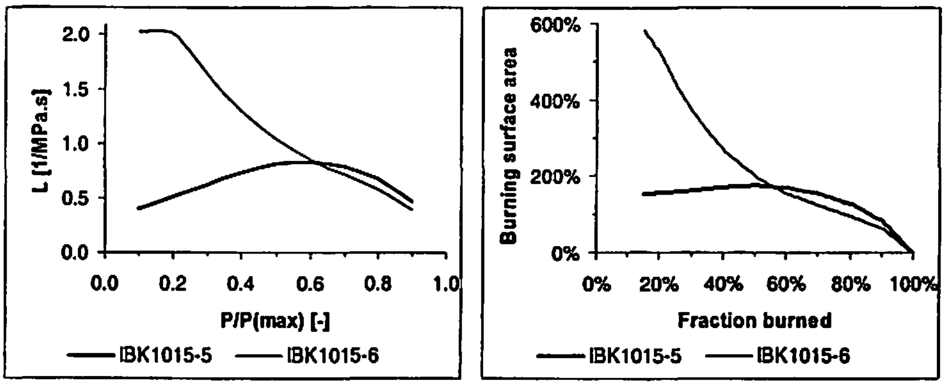

Fig.7 Vivacity curves determined from CV test results for propellant batches IBK10150-5 and IBK1015-6 (left), and Burning surface area A related to the initial theoretical surface area, calculated from the respective grain geometry's (right).

propellant. The vivacity increase at low conversions ($P/P_{max} < 0.5$) is caused by the large burning exponent, α, which is larger than 1. The vivacity of the other propellant batch appears to be much higher at low conversions, which is characteristic for porous propellant grains.

The burning surface area per grain can be calculated from both vivacity curves and the linear burning rate. The results of these calculations are shown in fig. 7 (right). This figure shows that the burning surface area of IBK1015-5 initially slightly increases, showing a progressive burning behaviour, which is characteristic for 7-hole grains. The burning surface area of the porous batch is initially much larger, showing the effect of zones with large pores in the propellant material.

The burning rates that were calculated for the HTPB- and TPE-based TNO propellants are plotted as a function of pressure in fig. 8 (left), together with some data from the literature. This figure 8 shows the improvement of TNO's TPE-based propellant compared to the HTPB-based one and some reference compositions. The problems with carbon deposit during burning of the TPE based composition was solved by additives. The composition is in the process of patenting.

The gain in performance over the years is shown in fig. 8 (right), which presents the impetus versus flame temperature for various generations of gun propellants. The impact of the use of high explosives, like RDX as applied from the HTPB-based propellants onwards, can be clearly seen in this figure. The use of energetic TPE's leads to a shift towards the upper right corner, implying an increase both in flame temperature and in impetus. The increase of impetus results generally in an

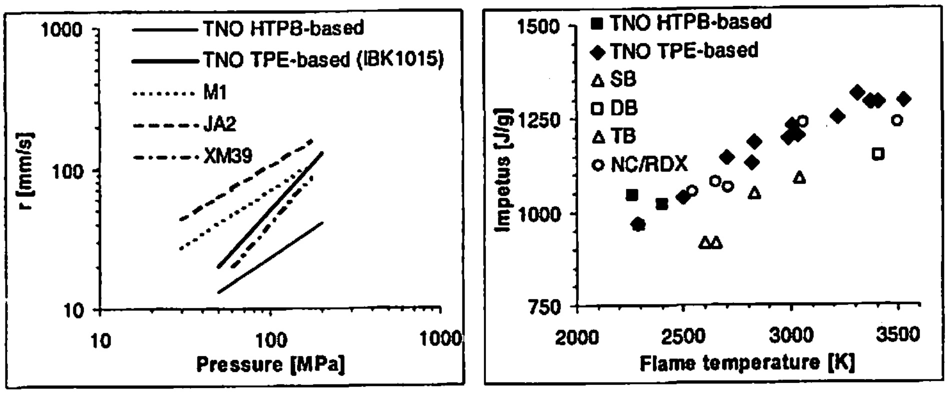

Fig.8 Properties of various gun propellants: Linear burning rates, r, as a function of the pressure (left) and Impetus as a function of flame temperature (right).

Table1 Summary of Gun propellant test data.

type	M1 single base	JA2 double base	M30 triple base	TN$^{O1)}$ HTPB-based	XM-39 NC/CAB-based	TN$^{O2)}$ TPE-based	Thiokol ETPE-based
performance							
Impetus (J/g)	920		1150	1060	1050	1070	1050
flame temp. (K)	2510	3420	3040	2400	2670	2560	
gun (mm)/cal		120	155/55			155/55	
v_0 (m/s)		1550	830			825	
Pmax (MPa)		400	320			325	
safety							
impact (Nm)	3	< 2.5		7.5	9.6	7.5	+
friction (N)	192			252		168	+
ESD (J)	4.5			4.5		@	> 8
VST (ml/g)	2.27			0.26		@	
DSC - T_0 (C)	177			190	192	215	151
TGA/DTA	153			200		221	
HOExpl. (J/g)	3580	4650		±3180	3400	@	
IM-related							
Ballistic	+			+		+	+
EMech. (MPa)	2900	310		40	6200	@	4700
/Ðm (MPa)	74	30		4.8	92		12.9
/Ãm (%)	6		48	37	2.7		1
gap test [3]	8%			3%		@	
slow cook-off				+		@	
SCB	strong			mild		@	
crit. diam. (mm)	> 73	25		> 73	30	@	
bullet attack	+			+		@	
EIDS extern fire	+			+		@	
Shaped charge	+			+		@	

1) The experiments were carried out with M1 propellant as a reference. The results of the various tests were in most cases expressed as better than and sometimes worse than the M1 results.

2) The TPE-based gun propellants presented in this table are in the process of production scale up. Therefore the IM related tests have not yet been performed.

3) gap test - given is the EIDS test result in 100 % (L0 -L)/L0.

@ To be determined this year.

increase of the flame temperature. This is a negative result as bore erosion is proportional to the flame temperature.

3.3 Safety aspects

Safety issues concern the whole route from formulation via processing to transportation, storage and operational use. This chapter deals with the safety aspects Transport, Storage and the Military operations.

3.2.1 Transport and storage

In general transport and storage safety is regulated by United Nations test series 6. Some years ago agreement has been achieved upon the Test Series 7 for Extremely Insensitive Detonating Substances (EIDS), a prerequisite for assignment of ammunition into hazard division 1.6. The test series 7 consists of ten tests; six substance and four tests on articles containing an EIDS(s). To day no full UN test series 6 have been performed with the TNO compositions.

3.2.2 Operational aspects, Insensitive Munitions

From a military view the focus is on Insensitive Munitions. Specific IM tests have been developed, which are typically more realistic and practice oriented article tests such as fast and slow cook-off, bullet and fragment impact, shaped charge and sympatic detonation. Some tests, the substance selection for instance, are similar to the UN test series 7 (b, c, d, and f).

A number of tests according the UN test series 7 and the IM tests have been executed on the propellants under development and some typical reference propellants such as JA2 and M1[1,8]. The results are summarised in table 1.

From the results it can be concluded that the TNO HTPB-based gun propellant is evidently a less sensitive propellant than the reference M1. In fig. 9 typical test results of the Slow Cook off test are presented, showing that this HTPB-based propellant produces less fragments than M1.

The TPE-based TNO propellant is developed, produced and partly tested within a Common European Priority Area international collaboration program. Further testing and life firings will be performed in the second half of this year.

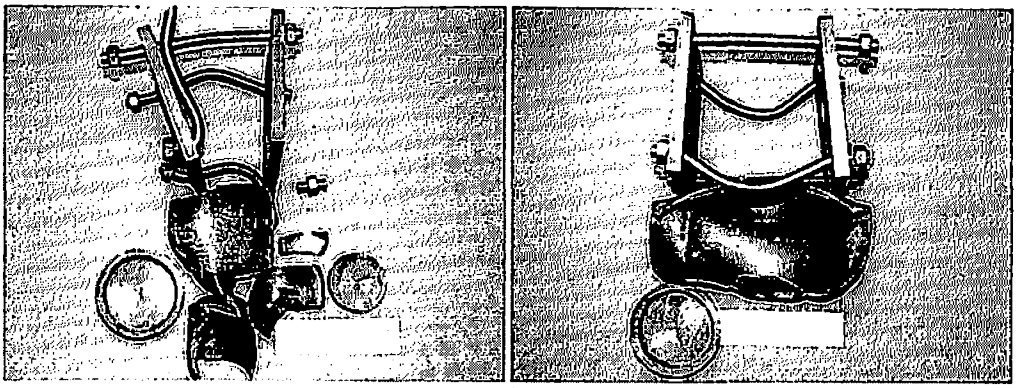

Fig.9 Slow Cook off test results for M1 (left) and TNO HTPB-based propellant (right).

4. Future developments at TNO-PML

At TNO-PML the following developments are considered:

- Application of Energetic Thermoplastic Elastomer (ETPE's) in gun propellant formulations. To date characterising of ETPE's takes place and the first small-scale batches of a PBX will be produced for sensitivity testing.
- Research on temperature (in)dependent gun propellant.
- Research on ignition with emphasis on plasma ignition of LOVA gun propellants.
- Continuous effort on safety issues of the extrusion processing of energetic materials.
- Extension of the research on lifetime assessment of propellants focussed on safety and functional aspects. Safety aspects are related to thermal stability (HFC) and functional aspects to the mechanical integrity of propellants. Recently a method based on the deterioration of NC chain length for conventional propellants has been developed[12].

5. Concluding remarks

- A systematic method is presented to develop advanced gun propellants. The development path comprises formulation; performance modelling; small scale production for experimental determination of compatibility and stability; and laboratory scale production for product characterisation and classification concerning operational and safety aspects.

- Advanced composite gun propellants for artillery applications are currently under development.

Acknowledgement

The authors would like to acknowledge the support of Mr. Jan van Gool (NL – MoD). The contributions of John Koman and Rudie Krämer in the extrusion work, of Louis Broekhuijsen, Wim de Klerk and Ellen la Haye for carrying out the safety tests and the research group EM for carrying out the IM related tests are appreciated.

References

1) Meer, B.J. van der, "Safety tests on seven new LOVA formulations", (TNO-PML report, 1994)
2) Hordijk, A.C.; Sabel, H.W.R.; Schonewille, E., "The application of Rheological equipment for improved processing of HTPB based PBXes", Procs 27th Int. Annu. Conf. ICT (1996)
3) Sabel, H.W.R.; Schonewille, E, "Extrusion trails with a TSK045 twin screw extruder", Procs 29th Int. Annu. Conf. ICT (1998)
4) Keizers, H.L.J.; Hordijk, A.C.; Vliet, L.D. van, "Modelling of composite Propellant Properties", AIAA (2000)
5) Hordijk, A.C.; Velde, I. v.d., "Capillary extrusion rheometry and extrusion of propellants and propellant simulants", Procs 31th Int. Annu. Conf. ICT (2000)
6) Hordijk, A. C.; Schoolderman C.; Van der Heijden, A. , "Highly filled TPEs – processing and rheology", Procs 3rd RAPRA Polymer Rheology Conf. (2001)
7) "PE - Protocol for initial assessment of Performance and sensitivity", TNO-PML protocol (1999)
8) Hordijk, A.C.; Karthaus, W.; Driel, C.A. van; Bouma, R.H.B., "Safety testing and IM characterisation of LOVA gun propellant', Procs EM & IM Techn. Symp. (2001)
9) Hordijk, A.C.; Schoolderman, C., "Highly Filled TPEs – processing and Rheology", Procs RAPRA Technology Limited (2001)
10) Benbow, J.; Bridgwater, J., "Paste flow and extrusion", Clarendon Press Oxford, 1993
11) Schoolderman, C.; Hordijk, A., "Processing of highly filled energetic compositions", to be presented at 33th Int. Annu. Conf. of ICT (2002)
12) Driel, C.A. van; Klerk W. de, "Burning properties and mechanical integrity of aged gun propellants", to be presented at 20th Int. Symp. on Ball. (2002)