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Experimental and Theoretical Investigation of a Model Reactive Armour with Nitrocellulose and Cellulose Composites*)

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Abstract: Monolithic nitrocellulose and cellulose composites (NC-C) were obtained by cross-linking a mixture of nitrocellulose with cellulose using hexamethylene diisocyanate (HDI). They were incorporated into a model reactive armour. An X-ray technique was used to examine the influence of the cellulose content on the accelerating ability of the gaseous reaction products of the composites. The modified Gurney model was used to simulate the process of driving steel plates in the reactive armour after jet impact. Formulae for the determination of the time-space characteristics of the plates' movement were derived. The results of the X-ray recording of the plates driven by the explosion products of monolithic nitrocellulose and cellulose composites (NC-C) were used for the verification of the model.

Keywords: nitrocellulose-cellulose composites, reactive armour, Gurney model

Introduction

In an earlier paper [1], we proposed that monolithic nitrocellulose-cellulose (NC-C) composites were suitable for SLERA (*self-limiting explosive reactive armour*) or NxRA (*non-energetic reactive armour*). These types of armour fulfil their protective function and simultaneously do not cause any damage to the

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protected environment [2]. The composites were prepared by separate cross-linking of the mixed precursors (nitrocellulose + cellulose) with hexamethylene diisocyanate (HDI). The NC-C composites obtained were characterized using TG/DTA thermograms and the results of sensitivity to friction and drop weight impact tests, and were used as a packing in a model reactive armour. The behaviour of the armour after shaped charge jet attack was investigated by the use of X-ray flash photography [3].

In the present work the experimental investigation of the reaction of the monolithic NC-C composites in the model reactive armour on jet penetration is continued. Moreover, the modified Gurney model was used to simulate the process of driving steel plates in a reactive armour after jet impact. Formulae for the determination of the time-space characteristics of the plates' movement were derived. The model was verified by using the results of the X-ray recording of plates driven by the explosion products of the NC-C composites.

Experimental

Materials and test methods

The composites were prepared from commercial grade nitrocellulose (13.13% N) and pure powdery cellulose (for TLC chromatography; Whatman, CF 11). For cross-linking of the polymers 1,6-diisocyantohexane (hexamethylene diisocyanate, HDI 98%, Aldrich D12,470-2), di-n-butyldilauryltin (DBTL 95%, ABCR GmbH&Co KG, AB106543) and methylene chloride (CH_2Cl_2 99.9%, Chempur) were used. The compositions tested are presented in Table 1.

Table 1. Compositions NCC*x*+HDI (*x* – rounded percentage of cellulose in admixture with NC)

Composition	NC [%]	Cellulose [%]	HDI [%]	DBTL [%]
NCC0+HDI	86.5	0	9.6	3.9
NCC20+HDI	69.2	17.3	9.6	3.9
NCC30+HDI	60.55	25.95	9.6	3.9
NCC50+HDI	43.25	43.25	9.6	3.9

The model armour consisted of two square (120×120 mm) steel plates separated by a *ca.* 10 mm layer of NC–C composite. The upper and lower cover plates were of 4 and 8 mm thickness, respectively. Square rods (10×10 mm) made of Plexiglas were used as a lateral enclosure of the NC–C layer. The shaped charge was placed above the upper steel plate at a distance of 70 mm.

The shaped charges for the tests were made of 21.5 g pressed, phlegmatized HMX and included 14 g sintered copper liners with a cone-shape angle of 60° and base diameter of 32 mm. The estimated jet velocity was 5750 m/s. The jet reached the tested armour element within ca. 20 µs after firing the shaped charge. The displacements and shapes of the plates at different time delays were recorded by the use of X-ray flash photography (SCANDIFLASH).

Reaction to shaped charge jet impact

In order to evaluate the influence of the energetic material composition on the motion characteristics of the model reactive armour elements, X-ray photographs of the armour after jet attack were taken. The photographs were recorded 100 and 200 µs after the firing of the shaped charge, so that the plates were driven for *ca.* 80 and 180 µs, respectively, as the jet reached the upper armour plate within *ca.* 20 µs.

Figure 1 shows pictures of the reactive armour containing composition NCC0+HDI recorded at 100 and 200 µs after firing the shaped charge.

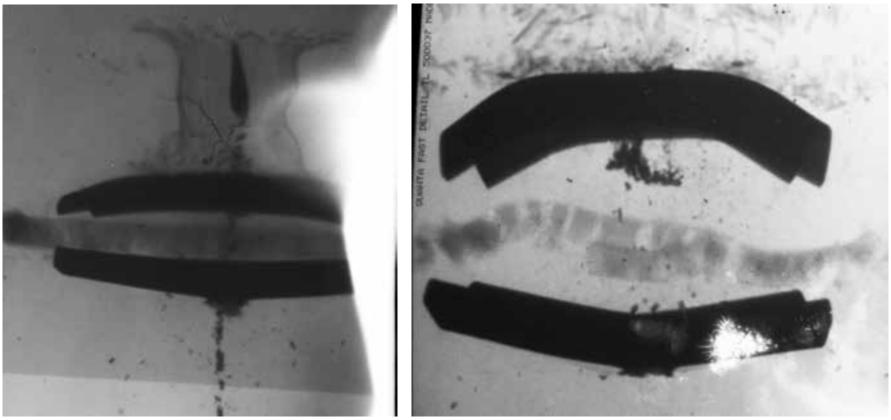

Figure 1. X-ray photographs of the reactive armour plates driven by the explosion of NCC0+HDI composition, taken 100 µs (on the left) and 200 µs (on the right) after firing the shaped charge.

Figures 2 and 3 show pictures recorded 200 µs after firing the shaped charge (*i.e. ca.* 180 µs after the jet impact). The energetic material layers were made of composites containing 20, 30 or 50% of cellulose (NCC20+HDI, NCC30+HDI and NCC50+HDI). The plates were more deformed when more energetic material was used as the armour intermediate layer, but they were never fragmented.

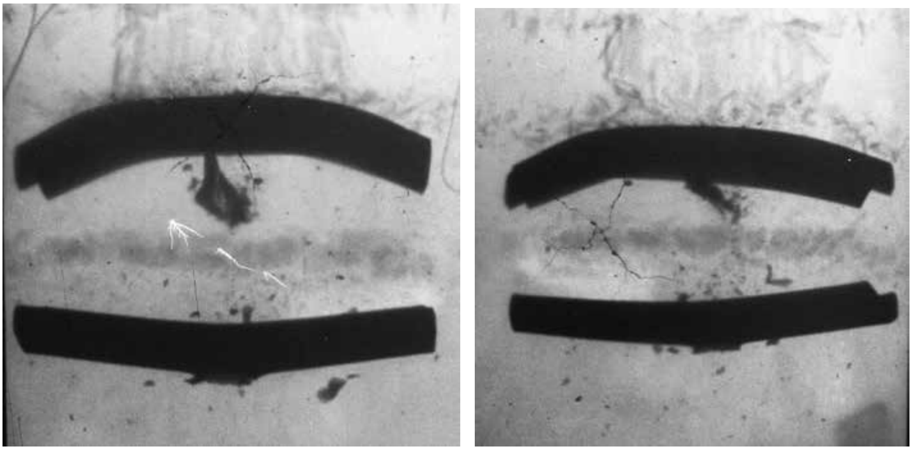

Figure 2. X-ray photographs of reactive armour plates driven by explosion of NCC20+HDI (on the left) and NCC30+HDI (on the right) compositions, taken after 200 μs.

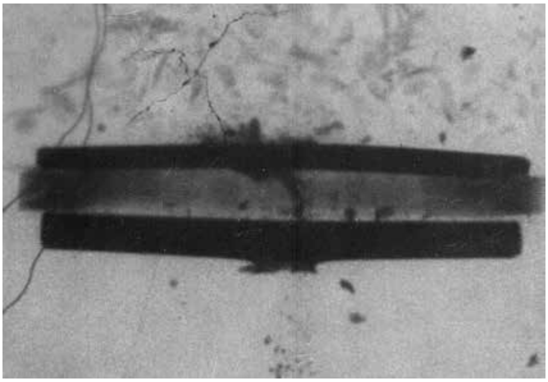

Figure 3. X-ray photograph of reactive armour plates driven by explosion of NCC50+HDI composition, taken after 200 μs.

In order to compare the positions of the steel plates and to estimate their velocity, profiles of the plates were scanned using the Sigma Scan graphics program [4]. The profiles are shown in Figures 4 and 5. Due to the small displacement of the plates in the armour with composition NCC50+HDI, the profiles of these latter plates are not included in Figure 5.

Knowing the displacement of the armour elements and the time of their motion, it was possible to calculate the average velocities of the plates. The mean velocity of the upper and lower plates of the armour containing a layer of NCC0+HDI composite, within a recorded time interval of 100-200 μs, was found to be 320 and 160 m/s respectively. It can be assumed that at this stage of the plate motion, the velocity is constant. The upper armour plates propelled by the explosion of NCC0+HDI, NCC20+HDI and NCC30+HDI within the 180 μs interval move at an average velocity of ca. 305, 260 and 220 m/s, respectively. The estimated velocity of the steel plates is 3-4 times smaller than the velocity of similar plates in a model reactive system in which a plastic explosive was used [3].

The plates were more deformed when more energetic material was used as the armour intermediate layer, but they were never fragmented. This fact and the comparatively low initial velocities of the plates indicate that the tested NC-C compositions explode or deflagrate under the experimental conditions.

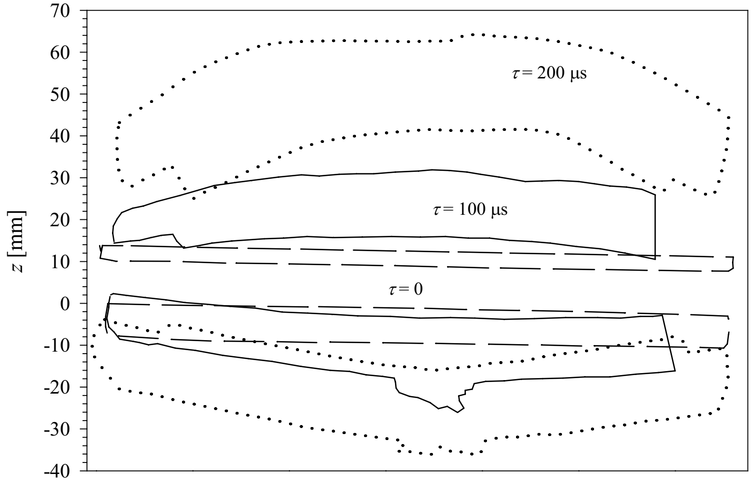

Figure 4. Profiles of steel plates driven by the explosion of composites containing 0, 20 and 30% of cellulose 100 and 200 μs after the firing of the shaped charge.

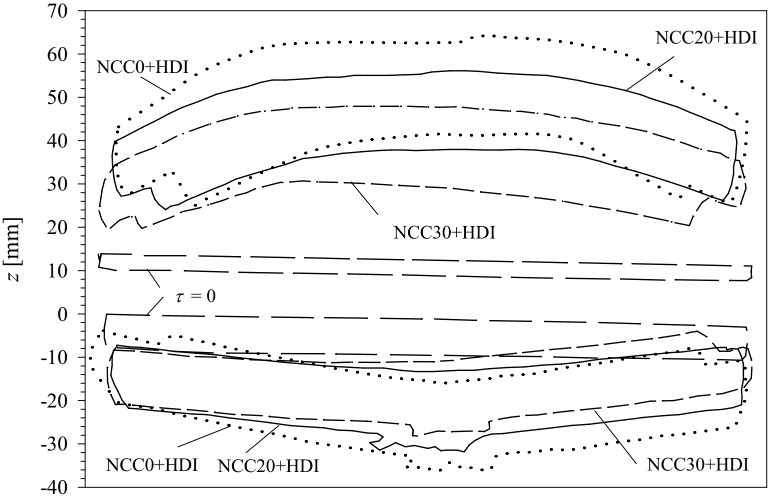

Figure 5. Profiles of steel plates driven by the explosion of composites containing 0, 20 and 30% of cellulose 200 μs after firing the shaped charge.

In order to determine which of these processes is more probable, the velocity of propagation of the chemical reaction's wave in a layered charge was measured. A layer of NCC20+HDI composition with a thickness of 10 mm was placed between steel plates both with a thickness of 4 mm and 250 mm square side. Holes were drilled along the diagonal of one plate, into which four short-circuit sensors were inserted, in order to measure the wave velocity in the composition layer. The distance between adjacent sensors was 40 mm, and the point of jet impact on the plate was situated at a distance of ca. 60 mm from the first sensor.

The following wave velocities were achieved in successive bases (pair of adjacent sensors): 2040, 2270 and 2140 m/s. The close values of the velocity indicate that the process is stable. This fact and the high value of the average velocity (2150 m/s) indicate that there is a stationary explosion (detonation). For this reason the NCC20+HDI composition cannot be treated as a non-explosive material. However, the low density of the test composition makes the detonation pressure low, which does not cause fragmentation of the accelerated plates, but is sufficient to drive them relatively fast. These materials thus meet the requirements for a reactive armour of SLERA or NxRA type.

Modelling

Methods for estimating the velocity of bodies accelerated by gaseous explosion products can be divided into three groups [5]. The first group is based on energy and momentum balances. The Gurney model and its equations are the best known [6-10]. The second group includes methods that use analytical solutions of the equations of the one-dimensional flow of a gaseous medium. They enable gas dynamic effects to be taken into account, however they generally refer to very idealized propulsion models. Examples of such methods are the models of driving bodies which consider the so-called active mass of the explosive charge [9-10]. Computer simulation methods for the process of body driving constitute the third group. Complex models of acceleration and the material properties of the bodies are taken into account, *e.g.*, [11-16].

It is assumed in the Gurney model [6] that detonation of a given explosive releases a fixed amount of energy which is converted into the kinetic energy of the driven inert material and the detonation product gases. It is also assumed that the gaseous products have uniform density and a linear, one-dimensional, velocity profile in the spatial coordinates of the system. Based on this assumption, it is possible to obtain, from the laws of conservation of energy and momentum, simple formulae for the terminal velocity of driven bodies. Jones, Kennedy, and Bertholf [17] applied the Gurney model to describe the process of acceleration of bodies and they obtained the solution of the differential equations in closed form.

The aim of the present study is to use the model proposed in [17] to predict the time-space characteristics of the metal plates driven by the explosion products of the NC-C composites and to compare the results obtained with those achieved from the X-ray recording of the plates of the reactive armour model. To obtain the time-space dependencies, the model described in work [17] has to be extended, because the armour analyzed includes the energetic material which detonates with a relatively low speed. It is shown that the assumption of immediate detonation does not preclude the application of this model for lower velocities as well.

Description of the model

Following the authors of work [13], the Gurney approach was applied in this study to estimate the variations in velocity of the metal plates adjacent to the charge of the energetic material, in the so-called “sandwich” system (Figure 6). The Gurney assumption of a linear profile of mass velocity in the gaseous charge and the use of an equation of state for the gaseous explosion products allows a solution of the metal plates’ acceleration problem to be obtained.

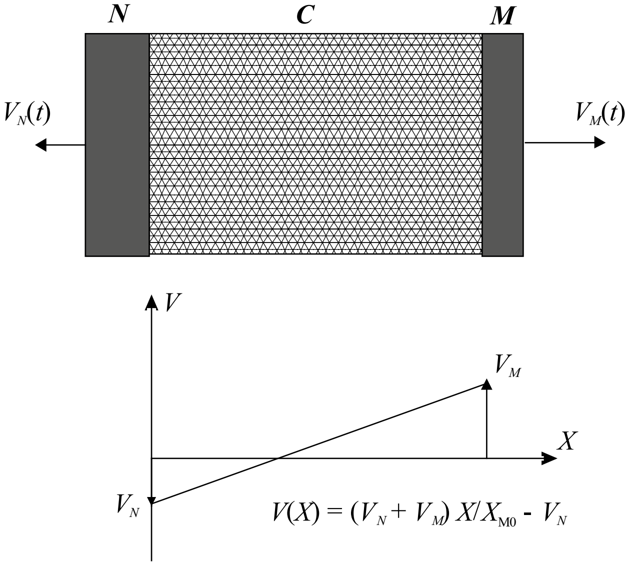

Figure 6. A scheme of the plane system for driving plates, illustrating the Gurney assumption for the distribution of the mass velocity of the gaseous products: N, M and C − respectively the mass of the plates and gaseous charge per unit area; the initial location of the surface of the plate with mass N is the origin of the coordinate Lagrange X.

Assuming that the instantaneous velocities of the plates are V_N and V_M, we can write the energy conservation law in the following form:

$$Ce_0 = Ce + \frac{1}{2}NV_N^2 + \frac{1}{2}MV_M^2 + \frac{1}{2}\rho_0 \int_{X=0}^{X=X_{M0}} \left[\left(V_N + V_M\right) \frac{X}{X_{M0}} - V_N \right]^2 dX \qquad (1)$$

where e_0 is the chemical energy per unit mass of unreacted energetic material, e is the internal energy remaining in the gas charge, ρ_0 is the initial density of the material ($C \equiv \rho_0\, x_{M0}$). It is necessary to note that in this case the velocities V_N and V_M are not the terminal velocities of the metal plates, but are the instantaneous velocities of these plates.

The equation describing the momentum conservation law can be written as follows:

$$0 = -NV_N + MV_M + \rho_0 \int_{X=0}^{X=X_{M0}} \left[\left(V_N + V_M\right) \frac{X}{X_{M0}} - V_N \right] dX, \qquad (2)$$

The ratio of the velocities V_N and V_M can be determined from Eq. (2):

$$A \equiv \frac{V_N}{V_M} = \frac{2\frac{M}{C}+1}{2\frac{N}{C}+1}, \tag{3}$$

and after the integrating Eq. (1) one can obtain:

$$C e_0 = C e + \frac{1}{2} V_M^2 \left(N A^2 + M + \frac{C}{3} \frac{1+A^3}{1+A} \right). \tag{4}$$

From Eq. (4) it follows that for $e = 0$, *i.e.* when all the chemical energy is converted into kinetic energy, the plate of mass M reaches the maximum velocity expressed by the relation:

$$V_M = \sqrt{2 e_0} \left(\frac{N}{C} A^2 + \frac{M}{C} + \frac{1}{3} \frac{1+A^3}{1+A} \right)^{-1/2}. \tag{5}$$

Relation (5) is identical with the Gurney expression of the terminal velocity of the plate driven in the system under study, except that in this expression, the Gurney energy E, where $E < e_0$, is used instead of the chemical energy e_0. This difference results from the fact that not all of the potential energy of the gas charge is converted into kinetic energy in real systems. Since the Gurney energy is determined on the basis of the measured terminal velocity of the driven bodies, so in further considerations the energy e_0 is replaced by the energy E.

Following the authors of work [17], we apply the ideal gas equation to describe the physical properties of the gaseous charge. The formula for the internal energy of the gas then has the form:

$$e = \frac{p}{\rho(\gamma - 1)}, \tag{6}$$

where p is the pressure in the gas and γ is a constant adiabatic exponent. The pressure p acts on the unit surface of the plate.

Let us define B and $\dot{x}$ as follows:

$$B \equiv \frac{N}{C} A^2 + \frac{M}{C} + \frac{1}{3} \frac{1 + A^3}{1 + A} \tag{7}$$

and

$$\dot{x}_M \equiv V_M \,, \tag{8}$$

where x_M is the Eulerian position coordinate of the surface of plate M and the superscript dot denotes time differentiation. Then, using the ideal gas and acceleration relations, we obtain an ordinary differential equation which describes the motion of the plate:

$$\rho E(\gamma - 1) = M \ddot{x}_M + \frac{1}{2} B \dot{x}_M^2 \, \rho(\gamma - 1) \,. \tag{9}$$

Use of the kinematics of the plate and the density expressed in terms of x_M, and the initial conditions, allows us to eliminate ρ from Eq. (9), leaving only x_M and its derivatives as unknowns:

$$2E = B \dot{x}_M^2 + \frac{2M}{C(\gamma - 1)} \ddot{x}_M \left[x_M (1 + A) - x_{M0} A \right] . \tag{10}$$

To solve this equation the identity:

$$\ddot{x}_M = \frac{1}{2} \frac{d\dot{x}_M^2}{dx_M} \tag{11}$$

is used.

Eq. (10) becomes:

$$2E = B \dot{\eta}^2 + F \eta \frac{d\dot{\eta}^2}{d\eta} \,, \tag{12}$$

where

$$\eta = x_M - \frac{A}{A + 1} x_{M0} \qquad F \equiv \frac{M(A + 1)}{C(\gamma - 1)} \,.$$

This is the first-order linear equation for $\dot{\eta}^2(\eta)$ whose solution (for the initial conditions $\dot{\eta} = 0$ and $\eta = \eta_0$ at $t = 0$) is:

$$\dot{\eta}^2 = \frac{2E}{B}\left[1-\left(\frac{\eta}{\eta_0}\right)^{-B/F}\right]. \tag{13}$$

Transforming this to Gurney parameters, the solution for the system in Figure 6 is as follows:

$$V_M^2 = \frac{2E}{B}\left\{1-\left[\left(\frac{x_M}{x_{M0}}\right)(A+1)-A\right]^{-BC(\gamma-1)/M(A+1)}\right\}, \tag{14}$$

where A and B have been defined above.

Relation (14) was derived in Ref. [17]. However, taking into account the definition (8), the time-dependence of a position of the plate M can be also derived from the relation:

$$t = \int_{x_{M0}}^{x_M} \frac{1}{V_M(x_M)} dx_M , \tag{15}$$

by calculating the integral in a numerical way.

Formulae (14) and (15) were derived on the assumption that the transformation of the explosive into the gaseous products occurs instantaneously in the whole volume of the charge. In the case of a detonation running at high velocity this is a reasonable assumption. However, when a lower velocity of explosion takes place in the energetic material, the time of the explosion of the charge may have a significant influence on the acceleration process and profiles of the driven plates. We have assumed that the model described above can be applied independently to the transverse slices (rings) of the steel plates and a slab of energetic material. The chemical transformation and the movement of the plates in subsequent sections of the system begins with a delay associated with the arrival of the explosion wave. However, it is necessary to note that the assumption of planar symmetry in subsequent sections of the system is a coarse approximation, because in real armour the wave propagation at low velocity causes deflection of the steel plates. Thus, the modified model requires experimental verification.

Analysis of the calculation results

The proposed model was used to simulate the acceleration of steel plates by the explosion products of the NCCx+HDI composites described in Table 1. The calculations were performed for the four composites NCCx+HDI. Their density was about 0.35 g/cm^3. The characteristics of the composite components used for the thermochemical calculations are presented in Table 2.

Table 2. Characteristics of the composite components

Component	Chemical formula	Density [g/cm^3]	Creation enthalpy [J/mol]
NC	$C_6H_{7.29}N_{2.71}O_{10.41}$	1.45	-682 000
cellulose	$C_6H_{10}O_5$	0.90	-962 000
HDI	$C_8H_{12}N_2O_2$	1.24	39 300
DBTL	$C_{32}H_{64}O_4Sn_1$	1.05	-1046 000

The thermochemical calculations were performed using the CHEETAH code [18] with a set of parameters of the BKW equation given in [19] (BKWS). It was assumed that cellulose is chemically inert during the explosion of the composite section, because of the negative oxygen balance (approx. -30%) of the nitrocellulose. Initially, the adiabatic exponent γ for the explosion products was determined for each composite. On the basis of the calculated velocity of the standard copper tube, driven by the detonation products of a given composition, the Gurney energy *E* was estimated. Next, it was assumed that this energy is equal to the Gurney energy corresponding to the explosion of the real composite. The parameters obtained and the calculated detonation velocity for the tested composites are given in Table 3.

The explosion process was studied experimentally only for the layer of composite NCC20+HDI. It was found that this process is approximately stationary and the average velocity of the propagation wave is 2150 m/s. This value is lower, by ca. 700 m/s, than the velocity of an ideal detonation (see Table 3). In the cases of heterogeneous, non-ideal, energetic materials, this difference is justified, especially for charges of small thickness.

Table 3. Explosion parameters for compositions NCC*x*+HDI

Composition	γ	*E* [J/g]	D [m/s]
NCC0+HDI	2.20	1324	3494
NCC20+HDI	2.23	1108	3179
NCC30+HDI	2.24	912	2855
NCC50+HDI	2.22	777	2522

The data corresponding to the armour with steel plates driven by the products from NCCx+HDI composites were used in the calculations. It was assumed that the model plate has an area equal to the contact surface of the real plate with a layer of energetic material, and its mass is identical to the actual total mass of the plate. The results of the calculations for the NCC0+HDI composite are shown in Figure 7 as the dependences of plate *M* of 4 mm thickness and plate *N* of 8 mm thickness on the driving distance. The relationship between this distance and time is also shown. The locations of the centre parts of the plates, read from the X-rays photographs taken 80 and 180 μs after the moment of jet impact, are also shown in Figure 7. The maximum calculated velocity of plates *M* and *N* are 345 and 180 m/s respectively. These values correlate well with the experimental average speeds of 320 and 160 m/s assigned to the time range 80-180 μs.

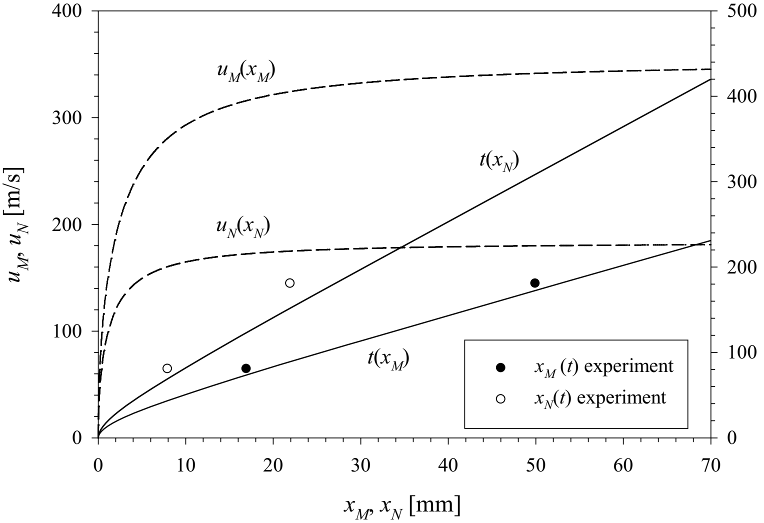

Figure 7. Dependence of the velocities of the steel plates on distance and the relationship between the distance and the driving time for the composite NCC0+HDI.

Similar relationships for plate *M* in sandwiches with the other cellulose contents are shown in Figure 8. The maximum speeds calculated for the composites NCC0+HDI, NCC20+HDI and NCC30+HDI are respectively 345, 290 and 270 m/s. The average speeds of the upper plates determined experimentally for the composites investigated are 305, 260 and 220 m/s, respectively.

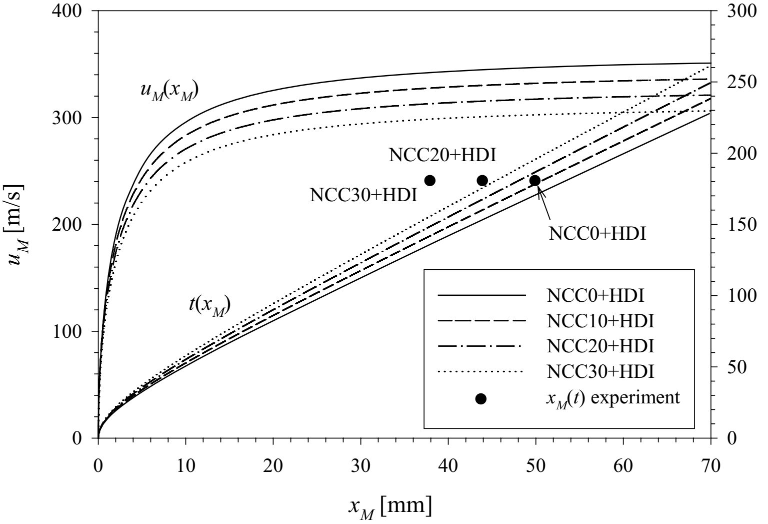

Figure 8. Dependence of the velocities of the thicker steel plates on distance and the relationship between the distance and the driving time for the composites tested.

As previously stated, in the actual system the reaction zone in the layer of energetic material travels with a finite velocity from the point of initiation (jet impact) towards the lateral edges of the layer and therefore the beginning of the motion of the plate varies successively. In order to compare the experimental and calculated profiles, an attempt was made to model the movement considering the delayed start of acceleration of the outer-lying elements (rings). The modified Gurney model was used for each ring layer of explosive and plates. It was assumed that the detonation velocity is constant for all the composites tested and was 2000 m/s. The results of the modelling for composite NCC0+HDI are shown in Figure 9 and they are compared with profiles of the external surfaces of the plates obtained from X-ray observation.

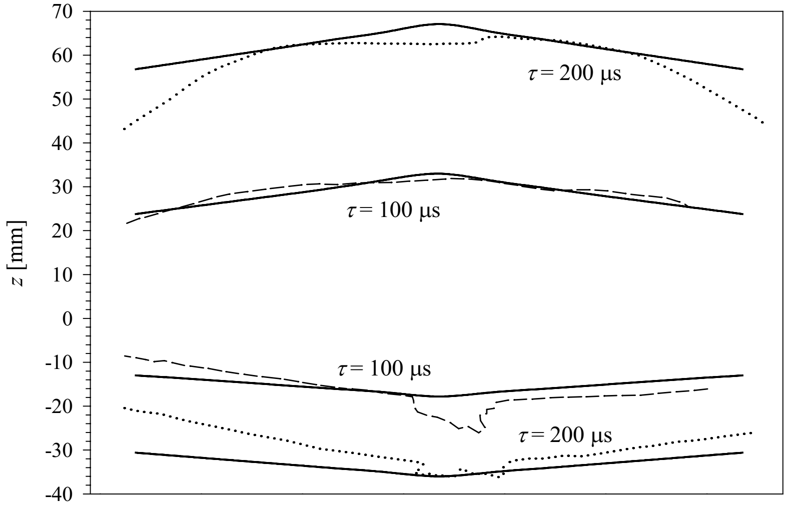

Figure 9. Comparison of the profiles of the external surfaces of the plates obtained by modelling (solid line) and X-ray measurement for the case of composite NCC0+HDI (τ – time delay of triggering X-ray source).

Similar comparisons were made for the other systems (see Figure 10).

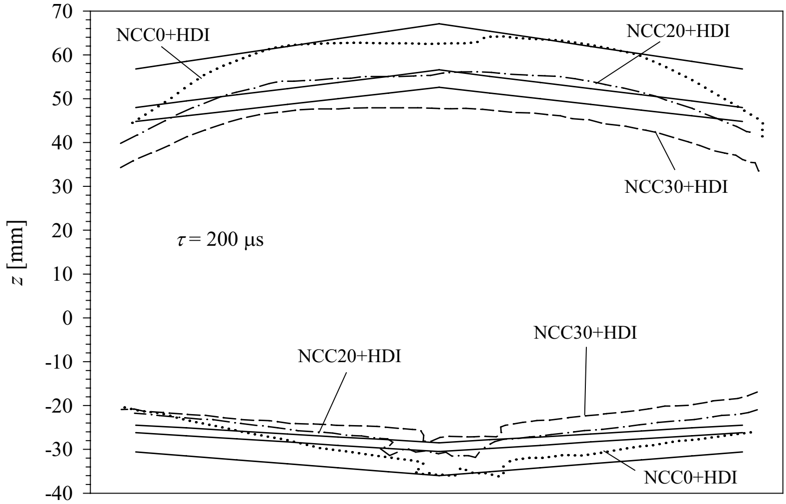

Figure 10. Comparison of the profiles of the external surfaces of plates obtained by modelling (solid line) and X-ray measurement (after time $\tau = 200$ µs) for the composites NCCx+HDI studied.

The model does not take into account the flow of explosion gases through the holes in the plates, caused by penetration by the cumulative jet, and the lateral expansion of the gases. Thus, significant differences in the shape of the calculated and experimental profiles of the plates in the middle and outer parts of the system are observed. Nevertheless, the proposed modified Gurney model enables us to determine the approximate time-space characteristics for the reactive armour with the NCCx+HDI composites.

Conclusions

Nitrocellulose-cellulose monolithic composites, obtained by cross-linking of nitrocellulose-cellulose mixtures with HDI/DBTL in methylene chloride, turned out to be real, tuneable, reactive materials, since their mechanical properties, sensitivity and performance can be easily modified by changing the mass ratio of nitrocellulose to cellulose.

The X-ray pictures of the shaped charge jet interaction with the model reactive armour, taken at different time delays, make it possible to determine the time-space characteristics of the armour plate motion. However, analysis of the images shows that the velocities of the driven plates are much lower than in the case of explosive reactive armour. Consequently the shielding effectiveness against cumulative jet interaction is also much lower.

The study has shown that the modified Gurney model enables the time-spatial characteristics of the reactive armour, in which nitrocellulose-cellulose composites are used as the energetic material, to be determined approximately. It was also found that the decomposition of these composites, after impact of the cumulative jet, is an explosion process of relatively high velocity.

Acknowledgements

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