TESTING THE PROTECTIVE EFFICIENCY OF PERSONAL RESPIRATORY PROTECTION DEVICES IN RADIOLOGICALLY CONTAMINATED ENVIRONMENTS

by

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The use of ammunition primed with depleted uranium is one of the hallmarks of modern combat operations, resulting in environmental contamination by particles of depleted uranium and uranium oxide, scattered around in the form of submicron-scale aerosols. This paper examined the protective effectiveness of the Serbian military's M3 protective face mask in relation to the presence of airborne depleted uranium and its by-products. Sodium chloride in solid aerosol form was used as a test substance and adequate physical simulator of such radioactive aerosols because its granulometric (particle) size distribution met the requirements of suitability as a simulator. Determination of aerosol concentration was carried out by flame photometry method, whilst granulometric distribution was determined by an electric particles analyzer. It was established that the total internal leakage of the M3 protective mask was as much a function of the penetration of particles through the combined M3 filter as of the leaks along the fitting line of the user's face mask and the inhalation valve. In terms of its protective effect against aerosols of depleted uranium and associated oxides, the Serbian M3 protective mask was determined to be of high efficiency and physiological suitability.

Key words: depleted uranium, aerosols, protective mask, filter

INTRODUCTION

This issue is increasingly coming to prominence due to the widespread application of radioactive isotopes and the possibility of their abuse. The increased presence of airborne radioactive aerosols may have resulted from nuclear accidents [1, 2], traffic accidents [3], war operations [4-6] and terrorist actions [7]. Radioactive aerosols containing radionuclides of uranium, primarily depleted uranium (DU) can be found in the air. DU is suitable as a raw material for applications in many fields owing to its high density, low cost, and availability; in medicine (as a means of protection from γ-radiation), in the civilian aviation industry [8] (in airplanes as a counterweight for purposes of balance, in containers for the transport of radioactive material) and in defense industries [9, 10] (in ammunition with greater penetrating power; in "Tomahawk"-type cruise missiles designed to penetrate tank armor, *etc*.). DU is a by-product of the process of enriching natural uranium with fissionable isotope ^{235}U for use as a nuclear fuel element. It contains 0.2% isotope ^{235}U, which is much lower than the concentration found in natural uranium (0.72 wt.%). DU radioactivity is at most 60% of the radioactivity found in natural uranium, and therefore it is considered a low-radioactive waste [8]. There is a significant economic interest in countries that have huge amounts of radioactive waste to dispose of in an effective way. It is estimated that during processing of natural uranium for the nuclear and military industries in the U. S. 700 000 tonnes of waste has been accumulated [11].

The use of DU-enriched ammunition, because of the radioactivity of ^{235}U, ^{238}U, and their descendants, leads to serious and far-reaching consequences in the health of humans contaminated with aerosol particles of uranium oxides [12, 13], as well as long-term contamination of the environment – land [14-16] and water [17, 18], where operations are conducted. Uranium's isotopes and descendants become highly dangerous only after entering the body, because they are α and β emitters [19]. As a heavy metal, depleted

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uranium presents both a radiological and chemical risk to health. The level of risk depends on the route of exposure and solubility. In DU chemical toxicity presents a greater risk than radiological toxicity. The biological half-life of uranium in a body is up to 15 days for the kidneys, as a critical organ, and 100 days for the whole body [20, 21]. Health problems such as damage of kidneys, bones and other organs, lung cancer and breathing problems [22] can appear years after exposure, through the original ingestion and inhalation of DU particles.

The many measures to reduce the risk of personnel contamination by micron and submicron-scale radioactive aerosols, include the use of special clothing (to protect against external radiation) and respiratory protection devices (protective against internal radiation). The Serbian army uses military protective masks for personal respiratory protection. It should allow high combat ability of troops in all the circumstances of sudden environmental contamination (chemical, radiological, and biological). According to the European standard [23], a protective mask is defined as a filtering device for respiratory protection of the respiratory organs, eyes and face from radiological-chemical-biological (RCB) contamination in the form of gases, vapors, solid, and liquid aerosols. It consists of a facepiece and combined filter, fig. 1(a). Combined filters are designed to remove gases, vapors, and particles from the air stream that passes through it [23]. The composition of the combined filter includes gas filter (active charge of the combined filter) and particle filter (filter against aerosols) placed in a common filterhousing, fig. 1(b). The particle filter is made of filtration material of fibrous structure featuring fibers of natural or artificial origin. Particle filters are classified into classes: P1 (80%), P2 (94%), and P3 (99.95%) according to the differences in their particle-retaining abilities (a measure also applied to the particle filters that are an integral part of the combined filters [24]).

One of the key parameters for assessing the protection effectiveness of a protective mask is monitoring the penetration of aerosol contaminants, *i. e.* determining the internal leakage of protective masks.

Since experiments with real contaminants in laboratory conditions are a high-risk activity, in practice one applies the test method of generating aerosols of a

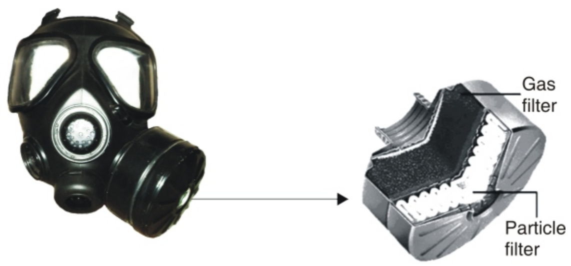

Figure 1. Serbian military protective mask M3 (a) and section view of the combined filter M3 (b)

non-toxic agent which simulates the presence of contamination in the air [25-27].

Previous studies of military protective masks offered no experimental proof of their protective efficiency against radioactive aerosols, only positive hypothetical assumptions. The aim of this study was to investigate the effectiveness of the protection afforded by, and the physiological suitability of, the last generation of personal respiratory protection devices, in circumstances of environmental contamination caused by use of ammunition enriched with DU.

EXPERIMENTAL

The samples of the M3 protective military mask and the combined M3 filter, on which testing was performed, were products of "Trayal" Corporation of Kruševac, Serbia. The particle filter within the combined M3 filter was a type of ULPA filter (manufactured by the French company, Bernard Dumass SAS). It is made on the basis of submicron diameter glass fibers (with a fiber mean diameter of about 0.40 m).

The appearance of the air, including potential contaminants within it, in the dead space of a mask during the inhalation phase is most possible during its passage through the filter and through elements serially connected to it, or on the fitting line of the breathing port (facepiece) on the user's face. In accordance with these factors that can directly affect the protection a protective mask affords against DU contamination, the following was carried out.

Testing of filtration efficiency and inhalation resistance of the combined M3 filter

The method and apparatus for testing with aerosol of sodium chloride per SRPS EN 14387:2007 [27], shown in SRPS EN 143:2007 was used [24]. In accordance with the method:

– the taking of representative samples of the combined M3 filter was performed by random selection of the required number of pieces of individually packed filters (8 samples),
– filters were prepared as follows: 4 samples were exposed to mechanical effect, and 4 samples to the mechanical effect and temperature conditioning,
– using a Collison-type spray, poly-dispersed solid aerosol from a 1% aqueous solution of NaCl, a concentration of C_0 = 8 mg/m^3 was successfully generated, with the following characteristics: particle diameter d_p = 0.02-2.0 m, particle median by weight MMD = 0.40 m, particle median per number NMD = 0.03 m, geometric deviation g = 2.53; the granulometric size distribution of the generated particles was determined by the EAA-3030 electri-

cal particle analyzer (manufacturing company: TSI, USA),

- filters were tested at a test aerosol flow of 30 dm^3/min,
- concentration of test aerosol was measured before and after the tested filter by a Type 1100 Sodium Flame Photometer (manufacturer Moores Ltd., Wallisdown, UK),
- initial penetration was measured 3 minutes after the commencement of the test, as the average response of the instrument in an interval of 30 3 s; precise determination is possible in the area of 0.0001% to 100% penetration through a filter, and
- resistance of the tested filter at a test aerosol flow was measured by differential "U" water meter pressure drop in front of and behind the filter.

Testing of internal leakage of the M3 protective mask

The method and apparatus for testing with aerosol of sodium chloride per SRPS EN 136:2007 was used [26]. In accordance with the method:

- 10 examinee were engaged in the study (shaved persons – without a beard and sideburns), selected according to specific criteria of anthropometric matrix – fig. 2 (covered all the typical characteristics of users' face, except for major anomalies), defined by SORS 8746/05 [28]; these people had already been trained in the use of identical or similar devices for respiratory protection,
- during testing 10 samples of M3 protective masks were used. First, they were tested as untreated samples (in the state in which they were received). Then, they were temperature conditioned, and after returning to ambient temperature, tested again); the taking of representative samples of a M3 protective mask is made by random selection of the required number of individually packaged masks,

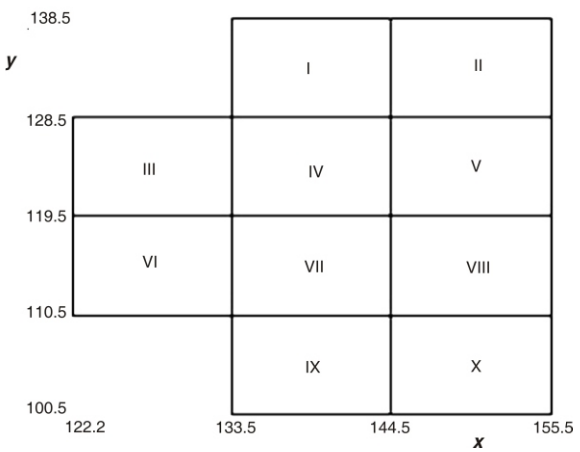

Figure 2. Anthropometric matrix: *x* – width of the face (bizigomatic width) [mm], *y* – length of the face (gnathion-nation) [mm]

- using a Collison-type spray poly-dispersed solid aerosol from a 2% aqueous solution of NaCl, a concentration of C_0 = 11 mg/m^3 was successfully generated, with the following characteristics: particle diameter d_p = 0.0-2.0 m, particle median by weight MMD = 0.60 m, particle median per number NMD = 0.03 m, geometric deviation g = 2.53; the granulometric size distribution of the generated particles was determined by the EAA-3030 electrical particle analyzer (manufacturing company: TSI, USA),
- testing was performed at a test aerosol flow of 100 dm^3/min to the test chamber,
- concentration of test aerosol was measured under the facepiece and in the test chamber by a Type 1100 Sodium Flame Photometer (manufacturer Moores Ltd., Wallisdown, UK) by the following method:
- a concentration sample of test aerosol leaked under facepiece, C_m, was taken through a sampling tube connected to a mask over a water drinking tube by the sequel (the facepiece was not damaged),
- a concentration sample of test aerosol in the test chamber, C_0, was taken by a special system for sampling and determined with the same photometer, and
- a precise determination is possible in the area of 0.0001% to 100% of aerosol concentration.

Remark

The decision for the use of solid sodium chloride aerosol to simulate radioactive aerosols containing DU, when testing filtration efficiency of combined M3 filter and the internal leakage of the M3 protective mask, was made on the basis of the physical properties of radioactive aerosols and generated aerosol of NaCl. In the case of DU, aerosols are formed by rapid cooling of particles generated by burning uranium at high temperatures during the explosion or break through a solid barrier (range 500-1000 °C). It follows that the diameter distribution of aerosol particles obey a log-normal distribution, where part of the DU mass (50% max), exceeds into aerosol particles of uranium oxide. Up to 62% of those particles are aerosols size up to 10 m [29]. Characteristics of size distribution of generated sodium chloride particles are given in SRPS EN 136:2007 [26] and SRPS EN 14387:2007 [27]. Comparing the particle size distribution of both types of aerosols, it was concluded that sodium chloride was suitable as a simulator of DU radioactive aerosols.

RESULTS AND DISCUSSION

Testing of filtration efficiency and inhalation resistance of the M3 combined filter

Results of testing the filtration efficiency and inhalation resistance of the M3 combined filter are given in tab. 1.

Table 1. The values of filtration efficiency and inhalation resistance of combined filter M3 samples at a sodium chloride test aerosol flow of 30 dm³/min

		State of sample	Aerosol penetration [%]	Filtration efficiency [%]	Filter resistance to inhalation [Pa]
Working sample code	1	MD*	0.0003	99.9997	120
	2	MD	0.0007	99.9993	120
	3	MD	0.0003	99.9997	130
	4	MD	0.0003	99.9997	130
	5	MD + TK**	0.0004	99.9996	110
	6	MD + TK	0.0004	99.9996	120
	7	MD + TK	0.0004	99.9996	120
	8	MD + TK	0.0005	99.9995	130
Mean value			0.0004	99.9996	122

MD* – filters exposed to mechanical effect
MD + TK** – filters exposed to mechanical effect and temperature conditioning

The filtration efficiency results of the samples investigated are very high and very homogeneous (standard deviation 0.000136). The mean value of filtration efficiency is 99.9996%, which is seven times higher than the value defined by standard SORS 8829/05 [30] (the penetration of aerosols is 0.0004%, which is seven times less than the limit value 3.0 10^{-3}%). In relation to the penetration limit value given in standard EN 143:2007 [24], which is 0.05% for the particle filter class P3, the test aerosol penetration through the tested combined filters is over 100 times less. So, based on the above, the test results obtained demonstrate that the combined M3 filter, as a vital part of the M3 protective mask, has a great influence on the protective properties of the device in terms of radiological contamination of the environment.

The inhalation resistance mean value of the combined M3 filters, obtained at a flow rate of 30 dm³/min, is 122 Pa, which is over 5% less than the 130 Pa which is the limit value defined by the standard SORS 8829/05 [30]. Not one of the resistance values measured exceeded any given limit, so the combined M3 filter fully meets the quality requirements defined by this standard. Considering that the resistance limit value of the individual P3 particle filter class (standard EN SRPS 143:2007 [24]) is 120 Pa, this is further confirmation of the quality of the combined M3 filter, based on the tested samples. The results of the conducted tests give rise to a positive assessment of the suitability of the M3 protective mask and its combined M3 filter in terms of real usage.

Testing of internal leakage of the protective mask M3

Results of testing of the M3 protective mask internal leakage are given in tab. 2.

In both cases, the results of the internal leakage of the face-piece samples are very homogeneous (standard deviation of the results is 0.000157 for untreated face-pieces, and 0.000125 for treated face-pieces). The obtained mean value of the internal leakage per examinee matrix with untreated face-piece is 0.0006%, which is much lower (>80 times) than the 0.05% which is the limit of leakage given in standard SRPS EN 136:2007 [26]. The situation with the treated face-pieces is very similar, because the mean value of the internal leakage of the treated face-pieces which was obtained was 0.0008%, which is also much less (>60 times) than the standard limit.

Table 2. The values of the internal leakage of facepiece protective mask M3 samples

		Internal leakage of untreated facepieces (mean value per examinee) [%]	Internal leakage of treated facepieces (mean value per examinee) [%]
Ordinal number of examinee in matrix	I	0.0007	0.0006
	II	0.0008	0.0009
	III	0.0004	0.0009
	IV	0.0006	0.0007
	V	0.0006	0.0009
	VI	0.0008	0.0007
	VII	0.0004	0.0007
	VIII	0.0005	0.0009
	IX	0.0008	0.0008
	X	0.0007	0.0006
Mean value per examinee matrix		0.0006	0.0008

According to the facts presented, the test results obtained demonstrate that the M3 protective mask achieves a very good fitting on the user's face and very reliably protects against radioactive aerosols in the surrounding environment.

CONCLUSIONS

For modern masks of the fourth generation, such as the Serbian M3 military protective mask, it has been shown that total internal leakage is as affected by the penetration of particles through the combined M3 filter as by the leakage on the fitting line along the user's face and through the inhalation valve (penetration and leakage are very small, almost negligible). Penetration of aerosols through the combined filter is 0.0004%, *i. e*. the filtration efficiency is 99.9996%, which is much higher, *i. e*. of greater efficiency, than the standard required for the combined filters and particle filters. Internal leakage is 0.0006% per examinee matrix with untreated face-piece and 0.0008% per examinee matrix with treated face-piece. These values are significantly under, *i. e*. well within, the limits defined by prevailing standards.

The M3 Military protection mask is physiologically suitable for application in conditions of real-life contamination by radioactive aerosols. Based on results obtained, it was concluded that the M3 combined filter and the face-piece of the M3 mask were functionally compatible when used in combination. For the combined filter as a vital part of the M3 protective mask, a resistance mean value of 122 Pa at a contaminated air flow rate of 30 dm^3/min was obtained, which is less, *i. e*. within the limits permitted by the standard for combined filters, and even within the limit of acceptability set by the independent particle filter standard.

Based on the presented, one can conclude that the Serbian M3 military protective mask is a highly effective and physiologically suitable means of respiratory protection against radioactive agents in the form of depleted uranium aerosols. This confirms the quality of the applied nanotechnologies in the production process of the given subject.

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AUTHOR CONTRIBUTIONS

Theoretical analysis was carried out by D. S. Rajić, N. D. Ivanković, and N. D. Ivanković, and experiments were carried out by D. S. Rajić, M. S. Ilić, Ž. B. Senić, and N. D. Pajić. All authors analysed and discussed the results. The manuscript was written by D. S. Rajić, N. D. Ivanković, and Ž. B. Senić, and figures were prepared by N. D. Ivanković, M. S. Ilić, N. D. Ivanković, and N. D. Pajić.

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ИСПИТИВАЊЕ ЕФИКАСНОСТИ СРЕДСТАВА ЛИЧНЕ РЕСПИРАТОРНЕ ЗАШТИТЕ У УСЛОВИМА РАДИОЛОШКЕ КОНТАМИНАЦИЈЕ ЖИВОТНЕ СРЕДИНЕ

Употреба муниције лаборисане осиромашеним уранијумом представља једно од обележја савремених борбених дејстава. При томе долази до контаминације животне средине честицама осиромашеног уранијума и оксидима уранијума које се распршавају у околину у форми аеросола субмикронских размера. У раду је испитивана ефикасност српске војне заштитне маске М3 у односу на присуство осиромашеног уранијума и његових продуката у ваздуху. Као тест супстанција и адекватан физички симулатор оваквих радиоактивних аеросола коришћен је чврсти аеросол NaCl чија гранулометријска расподела величине честица испуњава услове за његову подобност као симулатора. Одређивање концентрације ових аеросола обављено је методом пламене фотометрије, а гранулометријска расподела електричним анализатором честица. Показано је да укупно унутрашње пропуштање заштитне маске зависи подједнако од продирања честица кроз комбиновани филтер М3, као и од пропуштања на линији налегања образине на лице корисника и кроз вентил издисања. Утврђена је висока ефикасност и физиолошка подобност српске заштитне маске М3 у заштити од аеросола осиромашеног уранијума и његових оксида.

Кључне речи: осиромашени уранијум, аеросоли, заштитна маска, филтер