Estimation of Radiation Dose from Uranium Mill Tailings Bricks Used as Construction Material

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Abstract: In this study estimation of radiation dose from the bricks which are made of uranium mill tailings (UMT) was carried out. For this purpose, RESRAD-BUILD software was used for simulation of radiological exposure of occupant in a model room made with UMT bricks of various composition. In this study, standard model room having dimensions of 5 m × 4 m × 2.8 m was considered for computation. Its floor and ceiling are built with concrete, and the walls are made by using UMT bricks. The occupant location was considered at the centre of the room at 1m height from floor. Possible pathways of exposure were external exposure and inhalation of radon and progeny. The excess dose to occupant residing in model house made by UMT bricks had been found out to be 1 mSv for composition of UMT less than 20%. Further, radon concentration inside the room attributable to UMT bricks was found to be 6.5 – 32.2 Bqm^{-3} for UMT composition 10%-50% which is much less than the recommended action level for radon.

Keywords: Uranium mill tailings, Dwellings, RESRAD, Tailings bricks, Radon, Gamma, Dose.

1. Introduction:

Nuclear energy has played a significant role in achieving energy requirements worldwide. With the increase in demand for clean energy sources to reduce carbon emissions, nuclear energy has emerged as the most viable and robust option. Uranium mining and its extraction from natural deposits have been carried out for the past several decades for use in the production of electricity in nuclear power plants [1]. In India, Uranium mining started in the year 1967 in Jaduguda, Jharkhand [2]. At present, there are seven Uranium mines at Jaduguda, Bhatin, Narwapahar, Bagjata, Mohuldih, Turamdih and Banduhurang in Singhbhum East district in the eastern state of Jharkhand and one underground mine at Tummalapalle in Kadapa district of Andhra Pradesh. Additionally, two Uranium ore processing plants are operational at Jaduguda and Turamdih in Jharkhand which use acid leaching for extraction of U from ore, whereas one Uranium ore processing plant is operational at Tummalapalle in Andhra Pradesh which uses alkaline leaching for extraction of U from ore.

Upon extraction of U, low specific activity waste, known as Uranium mill tailings (UMT) generated in large volume is transferred in the form of slurry on a near surface engineered, designed tailings pond. The liquid part of the slurry is decanted and pumped back to the plant for reuse. The solid part of the slurry is impounded on the tailings pond for long term containment. Due to the presence of low ore grade (0.04% U_3O_8) Uranium deposits, almost the entire volume of processed ore emerges as waste. UMT contains residual amounts of U and all other radionuclides of the U decay series. Tailings ponds can contribute to public exposure through radon exhalation and wind erosion of particulates. If not managed properly, it can also contribute to the contamination of adjoining groundwater bodies. Thus, long term monitoring is necessary to ensure the stability and integrity of the tailings pond. The tailings pond covers a large area of land which becomes unsuitable for any other purpose. To avoid these issues, and to ensure the sustainable operation of the Uranium industry, utilization of UMT for other purposes is required. UMT contains residual amounts of U and other natural radionuclides, so it is necessary to assess the radiological risks associated with its utilization. This present study was carried out to simulate the radiological exposure of an occupant residing in a dwelling made from bricks of uranium mill tailings.

2. Material and methods

In this study a model room of dimensions 5 m × 4 m × 2.8 m was considered in which the floor and ceiling are made of concrete and walls are constructed from bricks made by UMT mixed with components of mixed fly ash bricks (MFAB). MFAB is composed of fly ash, sand, stone chips and gypsum. The radioactivity concentration of natural radionuclides in UMT and MFAB was analysed by using NaI(Tl) gamma-ray spectrometer. The data of radioactivity content and various other input parameters were used to simulate the dose inside the model house by using the RESRAD-BUILD computer code.

2.1 Natural radioactivity analysis

A total of 8 Uranium mill tailings (UMT) samples were collected from the tailings pond of Tummalapalle and 3 mixed fly ash brick (MFAB) samples were collected from local vendors. Samples were homogenized to reduce variance and were air dried, minced, crushed, and passed through a 2 mm mesh sieve [3]. The samples were analyzed for activity of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K using NaI(Tl) gamma-ray spectrometer [4,5].

Samples were packed and sealed in plastic bottles of 250ml capacity and stored for 28 days so that the progeny of radium and thorium attain secular equilibrium with their daughter products. The energy calibration of the detector was done by using ^{137}Cs (662 KeV) and ^{60}Co (1173 & 1332 KeV) sources [6]. The efficiency calibration was done by using standard IAEA sources RGU, RGTh & RGK. The detector was kept in lead shielding of 4cm thickness to reduce the background. The samples were counted for 10000s. ^{226}Ra activity measurement was carried out by the gamma energy peak 1764 Kev of its daughter ^{214}Bi. ^{40}K activity was measured by its gamma peak of 1460 KeV. ^{232}Th activity measurement was carried out by the gamma energy peak 2614 KeV of its daughter ^{208}Tl [7,8]. The activity of a radionuclide was then calculated using Eq (1)

$$\text{Activity (Bq/kg)} = \frac{\text{N x 100 x 100}}{(\text{T x } \gamma \text{ x } \eta \text{ x W})} \qquad (1)$$

where, N= Background subtracted net counts, T = Counting time (sec), γ = Gamma emission probability in %, η = Efficiency for the particular gamma energy in % and W is the weight of the sample in kg.

2.2 Simulation of radiological exposure using RERRAD-BUILD Computer code

RERSAD-BUILD computer code was used for the simulation of radiological exposure of the occupant in a model room made with UMT bricks. RESRAD-BUILD developed by Argonne National Laboratory is a pathway model developed to assess the exposure of individuals resulting from occupancy in a contaminated building with radioactive materials [9, 10]. It is a member of RESRAD family computer codes which is used by regulatory authorities and universities in more than 100 countries for dose and risk calculations. RESRAD-BUILD computer code is capable of calculating the external radiation dose arising from radiation sources, inhalation doses from suspended dust and radon gaseous and ingestion doses from suspended radioactive particles. The code allows considering up to 10 sources of contamination within the building which can be point source, line source, area source or volume source (contaminated wall). Building geometry can be chosen as starting from a single room to multiple rooms or storey up to three compartments. Dose for multiple receptors can be calculated at different locations inside the building. The code provides an option for introducing shielding between the source and receptor.

The exposure pathways provided in RESRAD-Build include external and internal exposure which are mentioned below:

- External exposure to radiation emitted directly from the source.
- External exposure to radiation emitted from radioactive particulates deposited on the floors of the room.
- External exposure to radiation due to submersion in air borne radioactive particulates.
- Inhalation of airborne radioactive particulates.
- Inhalation of aerosol indoor radon decay products.
- Inadvertent ingestion of radioactive material contained in removable material directly from the source.
- Inadvertent ingestion of airborne radioactive particulates deposited on the surfaces of the building.

Out of these possible exposure pathways, the major contributors are external exposure to radiation emitted directly from the source and inhalation of radon and its progeny. These two pathways have been considered for the estimation of dose to the receptor.

In this study, a standard model room of dimensions of 5 m × 4 m × 2.8 m was considered. Its floor and ceiling are built with concrete. The walls of the room are made from bricks of UMT mixed with MFAB, which is the volume source of exposure to the receptor. The walls are plastered on both sides with concrete which acts as shielding between the source and the receptor. The thickness of the wall is 26 cm (23cm brick + 1.5cm plaster on both sides). The room has one door of dimensions 81” x 40” and two windows of dimensions 60” x 48”. The receptor location is at the center of the room at a height of 1m from the floor. The indoor fraction of occupation is 0.8. Model realization of the room is shown in Fig 1. The receptor is shown in the middle of the room and the shapes numbered 1 to 4 represent the walls which are the rectangular volumetric radiation sources.

2.3 Estimation of external dose

The model for calculating external dose from the volume source (brick) is based on a semi-infinite slab source, with corrections for geometrical factors. The effective external dose D_{ex} from exposure to

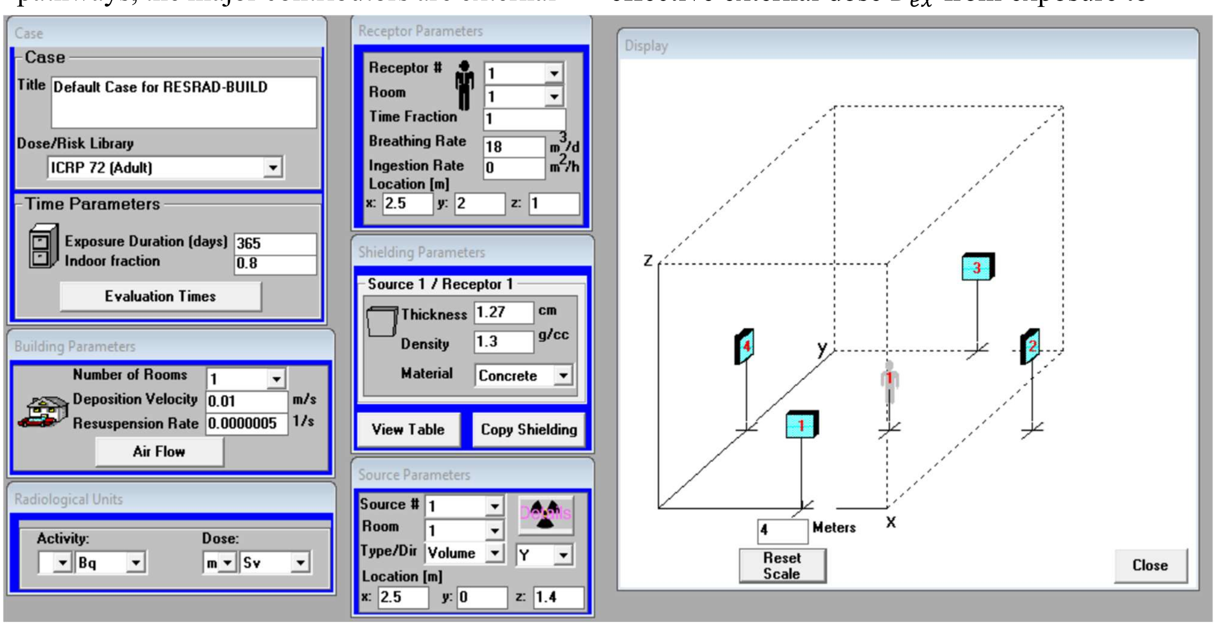

Figure 1. Room model in RESRAD-BUILD.

the source containing radionuclide n, is calculated by Eq. (2) [9]:

$$D_{ex} = ED * F_{in} * C_n * DCF_n * F_G^n \quad (2)$$

where ED is exposure duration (years), F_{in} is the fraction of time spent indoors, C_n is the concentration of radionuclide n (Bq kg^{-1}), DCF_n is dose conversion factor {($mSvy^{-1}$) ($Bqkg^{-1})^{-1}$} and $F_G^n$is geometric factor which takes into account area factor for finite area, source thickness, shielding, source material and position of receptor relative to the source for radionuclide n.

2.4 Estimation of indoor radon concentration and internal dose

A major portion of dose received by the receptor is due to the inhalation of short-lived progenies of ^{222}Rn. Radon is an inert gas which is a daughter product of ^{226}Ra. The radioactive decay of ^{226}Ra present in the UMT bricks leads to the formation of ^{222}Rn, which escapes out from the pores of brick matrix to the room. ^{222}Rn concentration inside the room was modeled by using air quality model. The radon concentration inside the room can be calculated by Eq. (3):

$$V_i \frac{dC_i^{Rn}(t)}{dt} = A_s J(t) - ((\lambda_{rn} + \lambda_v)\, V_i C_i^{Rn}(t) \quad (3)$$

where $C_i^{Rn}(t)$ is concentration of Radon-222 at time t, $J(t)$ is radon flux through the exposed wall area at time t, λ_{rn} and λ_v are radioactive decay constant for radon (0.007 h^{-1}) and air exchange rate (h^{-1}) respectively, V_i is volume of the room (m^3), $A_s$is surface area of the walls (m^2)

The mass balance equation for radon activity in a two-phase medium consisting of solid and gas phase (no moisture) in the brick matrix can be represented by Eq (4):

$$\frac{\partial(\eta C_i^{Rn}(t))}{\partial t} = -\vec{\nabla} x \overrightarrow{J_m}(t) - \eta\lambda_{rn} C_i^{Rn}(t) + \eta\varepsilon\rho_s C_i^{Ra}(t)\lambda_{Rn}(\frac{1-\eta}{\eta}) \quad (4)$$

where$C_i^{Ra}(t)$ is activity concentration of ^{226}Ra in brick at time t, $\overrightarrow{J_m}(t)$ is radon flux through the matrix, η is volumetric porosity, ε is radon emanating factor, ρ_s is the density of solid phase of the porous medium.

In the absence of convective flow of the gaseous phase in the porous medium, the radon flux can be expressed by the Fickian diffusion equation:

$$\overrightarrow{J_m}(t) = -nD_e \vec{\nabla} C_i^{Rn}(t) \quad (5)$$

where D_e is the diffusion coefficient of radon

Eq (5) and Eq (4) are combined and solved for steady state to compute radon concentration profile inside the matrix. The flux density at boundaries can be obtained by substituting the radon concentration profile to Eq (5) and solving at boundary condition. The obtained radon injection rate can be substituted to Eq (3) to obtain the radon concentration inside the room which can be used to calculate radon progeny concentration by mass balance equation. Working level month (WLM) can be calculated by radon progeny concentration and the effective internal dose (D_{in}) can be computed by Eq (6):

$$D_{in} = K * WLM * DCF \quad (6)$$

Where DCF is the dose conversion factor and K is multiplication factor to account for the extrapolation of doses from uranium mines to homes.

Total effective dose can be calculated by taking the sum of external and internal doses. The details of input parameters used for simulation in RESRAD-BUILD are presented in Table 1.

3. Results and Discussion

The average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in UMT was found to be 2657±131 $Bqkg^{-1}$, 15±3 $Bqkg^{-1}$ and 458±15 $Bqkg^{-1}$ respectively, whereas average activity of ^{226}Ra, ^{232}Th and ^{40}K in MFAB was found to be 68±2 $Bqkg^{-1}$, 51±3 $Bqkg^{-1}$ and 376±9 $Bqkg^{-1}$ respectively. In this study a total of six different compositions of brick were considered for dose simulation, as presented in Table 2.

Table 1. RESRAD-BUILD input parameters used for dose calculations.

Parameters	Values
Dose library	ICRP 72 (Adult)
Indoor/time fraction	0.8
Room dimensions	5 m × 4 m × 2.8 m
Thickness of wall	26cm (23cm brick + 1.5cm plaster on sides)
Density of brick	1.60 gcm^{-3}
Total surface area of walls (excluding doors and windows)	44.5 m^2
Volume of model room	56 m^3
Number of room/occupants	1/1
Breathing rate	18 m^3d^{-1}
Deposition velocity	0.01 ms^{-1}
Resuspension rate	5 x 10^{-7} s^{-1}
Occupant location in the room	Centre of the room at 1m height from floor
Shielding type	Concrete
Shielding thickness	1.27 cm
Shielding density	1.30 gcm^{-3}
Type of source	Volume
Source geometry	Rectangular
^{222}Rn diffusion rate	2 × 10^{-5} ms^{-1}
Radon emanation factor	0.1
Building air exchange rate	1 h^{-1}

The activity concentration of bricks is based on activity concentration of tailings and fly ash. Radium equivalent (Ra_{eq}) is a widely used parameter to assess the gamma radiation hazard to humans. Ra_{eq} is defined according to the estimation that 1 $Bqkg^{-1}$ of ^{226}Ra, 0.7 $Bqkg^{-1}$ of ^{232}Th and 13 $Bqkg^{-1}$ of ^{40}K produce the same gamma dose rate [11].

Ra_{eq} is calculated by using Eq (7) and is presented in Table 2.

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.07\ C_K \qquad (7)$$

Where C_{Ra}, C_{Th} and C_K are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $Bqkg^{-1}$ respectively. Recommended limits for building **materials** are also based on Ra_{eq} activity.

The bricks with UMT fall under Class 2 (Ra_{eq} between 370 – 740 $Bqkg^{-1}$) and Class 3 (Ra_{eq} between 740 – 2220 $Bqkg^{-1}$) as per recommendations for building materials based on Radium equivalent activity [12]. Thus, the UMT bricks are suitable as construction materials for industries, roads, bridges, and foundation of non-residential buildings.

The radiological dose to the receptor residing inside the model house with brick compositions mentioned in Table 2 and input parameters mentioned in Table 1 have been evaluated by the RESRAD-BUILD code. Additional dose to the receptor residing in the model room made by using UMT in bricks can be calculated by subtracting the background dose, i.e. dose due to MFAB. The estimated annual effective external, internal, and total doses to recipient in model house made by using MFAB are 0.13, 0.04 and 0.17 mSv respectively. Excess effective external, internal, and total dose are presented in Table 3. The excess dose rate and radon concentration above background are also provided in Table 3.

Table 2. Activity concentration in various brick composition and radium equivalent

Sl No.	Composition of brick	^{226}Ra ($Bqkg^{-1}$)	^{232}Th ($Bqkg^{-1}$)	^{40}K ($Bqkg^{-1}$)	Ra_{eq} ($Bqkg^{-1}$)
1	50% UMT + 50% MFAB	1363	33	417	1438
2	30% UMT + 70% MFAB	845	40	400	930
3	25% UMT + 75% MFAB	715	42	396	803
4	20% UMT + 80% MFAB	586	43	392	675
5	15% UMT + 85% MFAB	456	45	388	548
6	10% UMT + 90% MFAB	327	47	384	421

Table 3. Excess radiation dose from tailings bricks used in construction of dwelling.

Sl No.	**Composition of brick**	**External dose ($mSvy^{-1}$)**	**Dose rate (μSvh^{-1})**	**Internal dose ($mSvy^{-1}$)**	**Radon conc. (Bqm^{-3})**	**Total dose ($mSvy^{-1}$)**
1	50% UMT + 50% MFAB	1.66	0.24	0.72	32.2	2.37
2	30% UMT + 70% MFAB	1.00	0.14	0.43	19.4	1.43
3	25% UMT + 75% MFAB	0.83	0.12	0.36	16.1	1.19
4	20% UMT + 80% MFAB	0.66	0.09	0.29	12.9	0.95
5	15% UMT + 85% MFAB	0.50	0.07	0.22	9.7	0.71
6	10% UMT + 90% MFAB	0.33	0.05	0.14	6.5	0.48

The increase in radon concentration inside dwelling due to use of UMT bricks ranges between 6.5 – 32.2 Bqm^{-3}, which corresponds to internal dose of 0.14 – 0.72 mSv, for indoor equilibrium factor of 0.4 [13].

Radon concentration is well within the recommended action level of WHO [14] and ICRP [15] of 150Bqm^{-3} and 200 Bqm^{-3} respectively. The increase in dose rate inside the dwelling due to use of UMT bricks ranges from 0.05 – 0.24 μSvh^{-1} which corresponds to external dose of 0.33 – 1.66 mSv. The additional total annual dose due **to** use of UMT bricks in place of mixed fly ash bricks ranges from 0.48 mSv to 2.37 mSv for the given compositions. The measured background average gamma dose rate and radon conc. in dwellings around Tummalapalle are 0.16 μSvh^{-1} and 19.9 Bqm^{-3} respectively which corresponds to a dose of 1.16 mSv and 0.44 mSv respectively. For UMT composition of 25%, the dose rate including background comes out to be 0.28 μSvh^{-1} which is comparable to indoor gamma radiation levels of 0.30 μSvh^{-1} for dwellings as per the recommended a limit of the Swedish National Board of Housing, Building, and Planning [16]. External dose to dweller is found to be less than 1mSv when the percentage composition of UMT is less than 25% and total dose, when percentage composition of UMT is less than 20%, is less than the public dose limit of 1mSv.

4. Conclusions

The average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in UMT was found to be 2657±131 $Bqkg^{-1}$, 15±3 $Bqkg^{-1}$ and 458±15 $Bqkg^{-1}$ respectively, whereas average activity of ^{226}Ra, ^{232}Th and ^{40}K in MFAB was found to be 68±2 $Bqkg^{-1}$, 51±3 $Bqkg^{-1}$ and 376±9 $Bqkg^{-1}$ respectively. The radium equivalent of the six compositions of bricks ranges between 421 – 1438 $Bqkg^{-1}$. As per the recommendation based on radium equivalent, UMT bricks fall under Class 2 (Ra_{eq} between 370 – 740 $Bqkg^{-1}$) and Class 3 (Ra_{eq} between 740 – 2220 $Bqkg^{-1}$), thus, the UMT bricks are suitable as construction materials for industries, roads, bridges, and foundation of non-residential buildings. Additional dose due to the use of UMT bricks ranges from 0.48 mSv to 2.37 mSv. The additional radon concentration ranges from 6.5 – 32.2 Bqm^{-3}, which is well within the recommended action level. For computational purposes, 1 air change per hour was considered. Air changes per hour can be increased to further decrease the internal dose. External dose to dweller is found to be less than 1 mSv when the percentage composition of UMT is less than 25% and total dose is less than the public dose limit of 1 mSv when percentage composition of UMT is less than 20%. Thus, based on the input parameters considered for computation, bricks with UMT composition less than 20% are safe for use as construction material.

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References

1. Bhattacharjee B, In 14thIndian Nuclear Society Annual Conference (INSAC), IT-1, 1–27, 2003
2. Gupta, R. and Siddique, S, Management of the tailings and liquid effluents in uranium mining and milling operations. Radiation Protection and Environment, 26(3 & 4), (2003), 506 -515.
3. Operating Procedure, Soil Sampling. IAEA, Soil sampling for environmental contaminants. IAEA TECDOC-1415, International Atomic Energy Agency, Vienna, 2004
4. M. Condomines, O. Loubeau and P. Patrier, Recent mobilization of U- series radionuclides in the Bernardan U deposit (French Massif Central). Chem. Geol., 244 (2007) 304–315.
5. V.K. Shukla, S. Chinnaesakki, A.A. Shanbhag, S.J. Sartandel, G.K. Srivastava, A.H. Khan and V.D. Puranik, Proceedings of the National Symposium on Environment (2004) 445–448.
6. Knoll, G. F.,Radiation Detection and Measurement, 3rd edition, Wiley, ISBN 978-0471073383, 1999.
7. García-Talavera, M., Evaluation of the suitability of various γ lines for the γspectrometric determination of 238U in environmental samples. Appl. Radiat. Isot., 59, (2003), 165-173.
8. Morillon, C., Bé, M.M., Lamé, J., Jean, C., Nucléide- Lara. ISBN 2-7272-212-1, CEA-LNHB, Saclay, 2000
9. Yu C., LePoire, D.J., J.-J. Cheng, E. Gnanapragasam, S. Kamboj, J. Arnish, B.M. Biwer, A.J. Zielen, W. a. Williams, a. W. Iii, H.T. Peterson Jr., User's Manual RESRAD-BUILD Version 3, 2003.
10. Pepin, S., Using RESRAD-BUILD to assess the external dose from the natural radioactivity of building materials, Construct. Build. Mater. 168 (2018) 1003e1007.
11. UNSCEAR, Sources and Effects of Ionising Radiation, United Nations. Report toGeneral Assembly with Scientific Annexes. United Nations, New York, 2000
12. Oresegun, M.O., Babalola, A.I., In: Proceedings of an International Conference on Radiation Protection in Nuclear Energy, vol. 2. 18–22 April, IAEA, Vienna, Austria, pp. 159–166 (1988). Available at: https://inis.iaea.org/collection/NCLCollectionStore/_Public/20/026/20026283.pdf. (last accessed 20 September 2023).
13. UNSCEAR, United Nations Scientific Committee on the Effect of Atomic Radiation. Sources and biological effects of ionizing radiation. United Nations, New York, 1993
14. WHO Handbook on Indoor Radon. A Public Health Perspective. World Health Organization, Geneva (2009).
15. Lecomte, J.-F.; Solomon, S.; Takala, J.; Jung, T.; Strand, P.; Murith, C.; Kiselev, S.; Zhuo, W.; Shannoun, F.; Janssens, A., ICRP Publication 126: Radiological Protection against Radon Exposure. Annals of the ICRP, 43(3) (2014), 5–73.
16. Building Regulations, Mandatory provisions and general recommendations, Section 6 Hygiene, health and the environment, Code of Statutes of the Swedish Board of Housing, Building and Planning. BBR 2006:22.