RADIATION DOSIMETRY OF A GRAPHITE MODERATED RADIUM-BERYLLIUM SOURCE

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ABSTRACT

The Brookhaven National Laboratory Sigma Pile is a Radium-Beryllium neutron source imbedded in a cube of graphite blocks. The pile is approximately 2.13 m on four sides and is 3.07 m high. Absolute and relative thermal neutron flux measurements have been made using gold and indium foils, which were both bare and cadmium covered. Thermo-luminescent dosimeters were used to determine the neutron and gamma-ray dose rates in the pile. Gamma-ray dose rate measurements have also been made in the air outside of the pile, while the Radium-Beryllium neutron source was being withdrawn from the pile. The Monte Carlo MCNP code has been used to calculate the coupled neutron-photon transport. Measured dose rates at various locations agreed with the calculated values within 5% to 15%.

1. Introduction

The Brookhaven National Laboratory (BNL) Sigma Pile contains a Radium-Beryllium (Ra-Be) neutron source, which is embedded in a cube of graphite blocks. (The name " pile" comes from the historical manner of construction: the simple process of "piling up" blocks of graphite containing a neutron source into a structure. "Sigma pile" refers to the pile as a facility to measure diffusion properties of the graphite or other material being used for the neutron slowing down and the term sigma was the usual symbol for a neutron cross section). The Sigma Pile was ordered by BNL in 1947 and received in the time frame of 1948-1949. The amount of Radium was recorded as 1.0181 grams. After emission of a high energy α-particle from either ^{226}Ra or from one of its daughter nuclei emitted in the ^{226}Ra radioactive decay chain, neutrons are produced via the (α,n) reaction in Beryllium. This Ra-Be source was double encapsulated in stainless steel. The source was about 1.9 cm high and ~1.27 cm in diameter and it was inserted into the blocks of graphite. ^{226}Ra has a radioactive half-life [1] of 1599 years, so the neutron source strength does not vary with time to any large extent, once the equilibrium of the ^{226}Ra decay chain has been established.

The Pile is a square that is 2.13 m wide and 2.13 m deep. The total height is 3.07 m, as shown in Figure 1. There are seven slots with graphite trays on each of the four faces of the Sigma Pile. Each face of the pile has a number from 1 to 4 corresponding to the direction of the side, where 1 is the north side, 2 is the east side, 3 is the south side and 4 is the west side. Each of the slots and trays are assigned a pair of numbers, where the first number represents the side and the second number corresponds to the height of the slot on that side. 7 is the highest slot and 1 is the lowest slot. On the east side, slot 2-5 contains the Ra-Be source, so there are only 6 movable slots on the east side of the pile. There are 2.44 m between the highest and lowest slot on each face, so each slot is 40.64 cm from the one above and the one below for a given face, as shown in Figure 2.

Each slot position is offset by 10.16 cm as you move around the pile with the east side slot always being the highest. Slot 2-7 is the highest slot and is 11.43 cm from the top of the pile. It is located 10.18 cm higher than slot 1-7, which is 10.18 cm higher than 4-7, which is 10.18 cm higher than 3-7, which in turn is 10.16 cm higher than slot 2-6. This continues around the pile until you reach slot 3-1, which is the lowest slot in the whole pile and is 19.05 cm above the pile bottom.

2. Neutron Yield

^{9}Be can absorb α-particles to create ^{13}C in an excited state above the level for neutron emission and de-excite to an excited state of ^{12}C. Discrete higher energy groups of neutrons are emitted. Gamma-ray emission will accompany the de-excitation of ^{12}C. There will be both a neutron and gamma-ray dose rates as a result. α-particles can also scatter and excite ^{9}Be to a state above the neutron emission level, which leads to a continuum of lower neutron energies. The neutron spectrum has a series of peaks at high energy and a continuum at the low end of the energy spectrum. α-particles [2] in the ^{226}Ra decay chain include the following:

^{226}Ra with a half-life of 1,599 years has 2 major α-particles located at 4.784 MeV (94.5%) and 4.601 MeV (5.5%) and two weaker α's where the α-particle intensity is given in parentheses;
^{222}Rn with a half-life of 3.824 days has an α-particle at 5.490 MeV (99.9%) and a weaker α;
^{218}Po with a half-life of 3.04 minutes has an α-particle at 6.003 MeV (100%) and a weaker α;
^{214}Po with a half-life of 163.7 μ-sec has an α-particle at 7.687 MeV (99.99%) and a weaker α;
^{210}Pb with a half-life of 22.3 years has an α-particle at 3.720 MeV (0.0000019%);
^{210}Bi with a half-life of 5.01 days has 2 α-particles at 4.646 MeV (0.000079%) and at 4.694 MeV (0.000053%);
^{210}Po with a half-life of 138.4 days has an α-particle at 5.304 MeV (100%) and a weaker α;
^{210}Pb and ^{210}Bi are primarily β-emitters with weak α-particle decay branches; note the α intensity.

Since ^{210}Po is the product of 22.3-year ^{210}Pb decay, the neutron yield and the energy spectrum will depend on the amount of ^{210}Po present in the source. Is the Ra-Be old enough for the ^{210}Po to be in equilibrium with other decay products? The neutron dose rate to flux conversion factor varies with neutron energy, so the yield and dose rate will change with age of the Ra-Be source. Anderson [3] measured the neutron yield from a ^{226}Ra-Be source to be 526 ($n/10^6\alpha$). Runnalls [4] derived a formula, which determines a range of yields, from 485 to 552 ($n/10^6\alpha$) depending upon whether ^{210}Po is present or not. The Runnalls' formula had the form:

$$n_{max} = (0.152)\, E^{3.65} \qquad \text{in unit of } [n/10^6\alpha].$$

The Ra-Be source at the Atomic Energy of Canada Limited (AECL) was later renormalized by a factor of 0.95, which lowered Runnalls' effective range to values from 461 to 524 ($n/10^6\alpha$). If we use the Runnalls' formula for equilibrium of the radium decay chain and also with the contribution of the polonium-210 missing due to the holdup of the lead-210, the ratio is 0.879 for the reduction from the maximum.
Geiger [5] measured a Ra-Be source, with neutron yield of 502±50 ($n/10^6\alpha$). If we use measured (α,n) cross sections [6] and the α yield from the ^{226}Ra equilibrium decay chain cited above, calculated neutron yield becomes 454 $n/10^6\alpha$ (decays up to the 22 year half-life of ^{210}Po) and 524 $n/10^6\alpha$ (if ^{210}Po were in secular equilibrium with the whole decay chain), which agrees with the Runnalls' formula. Thus Geiger measured a young source (only 68% of the ^{210}Po α particles contributed).
The BNL Sigma Pile with 1.018 grams of ^{226}Ra would have a specific activity of 3.726×10^{10} dps, corresponding to 1.69×10^{7} n/s for a mature source compared to the new source value quoted

of $1.1x10^7$ n/s. It should be noted that Anderson and Feld [7] found the neutron yield also depended on the relative amount of Beryllium (m_{Be}) and Radium (m_{Ra}) in the ratio of $m_{Be}/(m_{Be}+m_{Ra})$. In our source, this would amount to $0.75x1.69x10^7$ or $1.27x10^7$ n/s. Finally, factoring in the loss of contribution of Polonium for a new source, 87.9%, the result for the neutron yield for our new Ra-Be source becomes $1.27x0.879x10^7=1.12x10^7$ n/s as quoted.

3. Calculational Details

The Monte Carlo code, MCNP-4B2 [8], developed and periodically updated by the Los Alamos National Laboratory is a general purpose Fortran compiled software package, which can model any single particle motion or coupled neutron photon transport in three-dimensional geometry consisting of different material regions. We used continuous cross sections for neutron and photon transport and reaction rate calculations and appropriate thermal neutron scattering function, $S_{\alpha\beta}$, to treat the neutron interactions with graphite in the MCNP package. To track particle histories starting from different source locations, version MCNP-4C was frequently used so data processing could be performed on a parallel computing platform. A continuous run option was used to increase the particle history and reduce the tally statistical uncertainty.

4. Instrumentation

For the neutron flux measurements at the slot positions in the trays, gold foils and indium foils were used. The indium foils were 3 cm in diameter and 0.0254 cm (10 mils) thick and weighed ~1.28 gm. For the neutron and gamma-ray dose rate measurements in the slots locations, thermo-luminescent dosimeters (TLD) were used. They were Harshaw (Saint Gobain Industrial Ceramics, Inc.) TLD-600 series, which contains Lithium which is enriched in ^{6}Li to 95.6% and TLD-700 series which contains Lithium with a ^{7}Li content of 99.9%. Both TLDs have approximately the same density and atomic number and have roughly the same response to gamma rays. The ^{6}Li has a large cross section for thermal neutrons in the (n,α) reaction of $934x\ 10^{-24}\ cm^2$ (934 barns). The ^{7}Li has a thermal neutron cross section of only a very small fraction of a barn.

For measurements of the neutron and gamma-ray dose rate outside the Pile, an RO-2 ionization chamber and a ASP-1 instrument for gamma rays and neutrons, respectively. The RO-2 is an ion chamber manufactured by Eberline, Inc. for gamma-ray dose rate measurements in the range from 0 to 5 R/h. For the dose rates as the source was being removed from the Pile, the neutrons were measured with an Eberline rem-ball, which is a polyethylene-coated Cadmium sphere filled with BF_3 gas, while the high gamma-ray dose rates were measured with a telescoping Geiger-Mueller instrument, a Teletector, produced by Zetex with a dose rate range from 0 to 10,000 R/h.

5. Neutron Flux

The thermal neutron flux was measured in the Sigma Pile on a number of occasions. By 1955, the pile was reconfigured into its present form and since then there were three sets of measurements to determine the flux in various slot locations. The various measurements of the neutron flux have used either indium foils or gold foils or both and the foils were irradiated bare and cadmium covered in the various slots of the Sigma Pile. In 1955, the relative neutron flux values were measured by cadmium difference with indium foils, bare and cadmium covered, while absolute measurements were made at two locations 1-5 and 2-7 with gold foils, bare and cadmium covered [9]. In 1962, both relative and absolute neutron flux measurements were made with indium and gold foils, respectively [10]. These foils were both bare and cadmium covered. In 1977 another set of measurements of the neutron flux at the Sigma Pile were made utilizing indium foils bare and cadmium covered [11]. There was a variation in the results of the thermal and epithermal neutron flux values in the different trays during the three separate measurements. Table I provided a sample of these thermal neutron flux results from the 1955 measurement. The tray locations and the distance of that tray from the Radium-Beryllium source is noted. In the 1962 measurement, indium foils of different thickness were used at the various tray locations. These different foil thickness correspond to a different self-shielding of the neutron flux by the atoms of the foil at a given neutron energy. Activity of the foils of various thickness were measured for both bare and cadmium covered foils so that the difference in the cadmium ratio between the thermal and epithermal neutrons that activated the given foil could be determined.

The cadmium shields that were used had a thickness of 0.0762 cm (30 mils). This thickness corresponds to a neutron energy cutoff of 0.5 electron-volts (0.5 eV), i.e., neutrons of energies above 0.5 eV are transmitted through the shield to the foil, while any neutrons of lower energies are absorbed by the cadmium shield and never reach the foils beneath the cadmium shield. In order to position the foils at approximately the same location in the slot trays for the bare measurements of the foils compared to the cadmium covered measurement, an aluminum cover of thickness 0.0508 cm (20 mils) was used, since the neutron absorption cross section for aluminum is only a fraction of a barn for thermal neutrons and would not appreciably affect the flux.

Table 1. Measured thermal neutron flux within the Pile

Tray	Source Distance	Flux ($n/cm^2/s$)	Tray	Source Distance	Flux ($n/cm^2/s$)
1-1	167.68 cm	146			
1-2	127.08 cm	520.	2-2	116.78 cm	720.
1-3	86.48 cm	1,900.	2-3	76.18 cm	2,600.
1-4	45.88 cm	7,020.	2-4	35.58 cm	9,340.
1-5	5.18 cm	15,520.			
1-6	35.22 cm	9,380.	2-6	45.42 cm	7,100.
1-7	75.82 cm	2,230.	2-7	86.12 cm	1,290.
3-1	187.88 cm	72.	4-1	177.78 cm	102.
3-2	147.28 cm	280.	4-2	137.18 cm	380.
3-3	106.68 cm	970.	4-3	96.58 cm	1,350.
3-4	66.08 cm	3,740.	4-4	55.98 cm	5,050.
3-5	25.48 cm	11,710.	4-5	15.38 cm	13,940.
3-6	14.92 cm	13,570.	4-6	25.02 cm	12,240.
3-7	55.52 cm	5,010.	4-7	65.62 cm	3,470.

For a number of these points, MCNP calculations have been performed. The comparison of MCNP results and the measured values are listed in Table 2. In general, there is very good agreement in the values, 10% or less, with the exception of the tray 2-7 location. At this location, there is a significant difference (66%) in the calculated and measured thermal neutron flux.

From Table 1, it can be noted that the thermal neutron flux falls off as the distance from the neutron source increases. The exception is the change from tray 2-7 at 86.12 cm to tray 4-3 at 96.58 cm where the neutron flux actually increases in traversing more graphite. This would indicate that either the thermal flux value at location 2-7 is too low or the flux at location 4-3 is too high. It can also be seen from Table 1 that there area number of pairs of locations that are at approximately the same distance from the source (with a fraction of a cm). Some examples would be 1-4 and 2-6, 1-6 and 2-4, 3-4 and 4-7, 3-5 and 4-6 and 3-7 and 4-4. For these pair of locations, the cadmium ratios of the activities and the thermal neutron flux values are similar. However, for the pair of locations 2-7 at 86.12 cm and 1-3 at 86.48 cm, the cadmium ratios and the thermal neutron flux values differ significantly. For location 1-3, the cadmium ratio and thermal flux values are 151 and 1,900 n/cm^2/s, while for location 2-7, the cadmium ratio and the thermal flux values are 89 and 1,290 n/cm^2/s. Either location 1-3 is too high or location 2-7 is too low. In 1962, cadmium ratio measurements were performed at a number of locations including 1-3 but not at location 2-7. The cadmium ratio of activities at location 1-3 was 161, in general agreement with the value of 151 and not with the value of 89. If the measured thermal neutron flux value at location 1-3, 1,900 n/cm^2/s is compared to the calculated value of 2,139 n/cm^2/s, the difference is only 12.6% rather than 66%.

Table 2. Comparison of the calculated and measured thermal neutron flux values

Tray Number	Calculated Flux	Measured Flux	Percentage Difference
1-4	6,944 n/cm^2/s	7,020 n/cm^2/s	1.1 %
1-5	15,277 n/cm^2/s	15,520 n/cm^2/s	1.6 %
1-6	9,822 n/cm^2/s	9,380 n/cm^2/s	4.7 %
2-7	2,139 n/cm^2/s	1,290 n/cm^2/s	66.0 %
3-4	3,965 n/cm^2/s	3,740 n/cm^2/s	6.0 %
3-5	11,312 n/cm^2/s	11,710 n/cm^2/s	3.4 %
3-6	13,194 n/cm^2/s	13,570 n/cm^2/s	2.8 %
3-7	5,510 n/cm^2/s	5,010 n/cm^2/s	10.0 %

These facts would seem to indicate that there may have been an error in the bare or the cadmium covered activity measurements at location 2-7, which is the cause for the great disagreement between calculation and measurement of the flux at that location.

6. Neutron Dose Rate

Neutron dose rates have been measured both in the slots of the Pile and external to the Pile at various distances from the source, using TLDs and a rem-ball, respectively. In Table 3, the neutron dose rates are presented in various Pile slots and in Table 4, the neutron dose rates are presented external to the pile as the neutron source was moved from its normal location toward to outer end of the pile. The * indicates the normalizing value of the MCNP calculated dose rate.

In general, the TLD-600 series has a ^{6}Li content of 95.6% and should be more reliable in determining the neutron dose rate since ^{6}Li has the much higher neutron cross section than ^{7}Li. Since the amount of lithium is the same and the atomic weight is about the same, the ^{7}Li can be used to correct for the gamma-ray contribution to the dose rate. The difference between the calculated and the measured dose rates in Table 3 vary between 4.0% and 5.4% at the various tray locations through out the Pile.

For the reported data of Table 4, the calculated and measured neutron dose rates agree between 2.0% and 16%, except for the very low reading in the micro-rem region, where the rem-ball is not too asccurate. In the milli-rem region, there is reasonable agreement in the neutron dose rates.

Table 3. Internal measured neutron dose rate in trays compared with calculation

Tray	TLD Type	Source Distance	Measured Dose Rate	Calculated from ^{6}Li	Calculated from ^{7}Li
1-5	$^{6}Li/^{7}Li$	5.18 cm	108.73 R/h*	108.73 R/h*	108.73 R/h*
2-6	$^{6}Li/^{7}Li$	45.42 cm	6.76 R/h	6.49 R/h	7.21 R/h
3-5	$^{6}Li/^{7}Li$	25.48 cm	10.42 R/h	9.86 R/h	12.12 R/h
3-6	$^{6}Li/^{7}Li$	14.92 cm	16.12 R/h	15.40 R/h	17.30 R/h
4-7	$^{6}Li/^{7}Li$	65.62 cm	3.54 R/h	3.36 R/h	3.73 R/h

Table 4. External measured and calculated neutron dose rate as source was extracted from Pile

Distance (the source moved from Origin)	Measured Dose Rate	Calculated Dose Rate	Detector Distance (30.48 cm in air)
30.48 cm	0.4 mR/h	0.14 mR/h	83.82 cm in graphite
60.96 cm	4.0 mR/h	4.23 mR/h	53.34 cm in graphite
76.20 cm	10.0 mR/h	11.6 mR/h	38.10 cm in graphite
91.44 cm	45.0 mR/h	43.4 mR/h	22.86 cm in graphite
99.06 cm	80.0 mR/h	76.8 mR/h	15.24 cm in graphite
106.68 cm	98.0 mR/h*	98.0 mR/h*	7.62 cm in graphite
114.30 cm	140.0 mR/h	137.2 mR/h	No graphite

7. Gamma-Ray Dose Rate

Gamma-ray dose rates were measured both in the pile and external to the pile, using the TLDs and with an RO2 ion chamber and with a Teletector, respectively. Table 5 provides the gamma-ray dose rates in various slots of the pile, while Table 6 provides the gamma-ray dose rate as the source was removed from the pile.

Table 5. Internal measured gamma-ray dose rate in trays compared to calculation

Tray No.	TLD Type	Source Distance	Measured Dose Rate	Calculated Dose Rate from ^{6}Li	Calculated Dose Rate from ^{7}Li
1-5	$^{6}Li/^{7}Li$	5.18 cm	299.36 R/h	293.0 R/h	297.0 R/h
2-6	$^{6}Li/^{7}Li$	45.42 cm	0.44 R/h	0.45 R/h	0.46 R/h
3-5	$^{6}Li/^{7}Li$	25.48 cm	4.93 R/h	4.91 R/h	5.03 R/h
3-6	$^{6}Li/^{7}Li$	14.92 cm	27.68 R/h	27.68 R/h	27.81 R/h
4-7	$^{6}Li/^{7}Li$	65.62 cm	0.063 R/h	0.054 R/h	0.065 R/h

From Table 5, there is good agreement between the measured and calculated gamma-ray dose rates at all locations. Although the gamma-ray dose rate can be calculated from the MCNP data for either TLD, the result from TLD-700 series should be more accurate. The agreement between the calculated and measured dose rates vary between 0.5% and 4.5% for the reported locations.

Table 6. External measured and calculated gamma-ray dose rate as source extracted from Pile

Distance (the source moved from origin)	Measured Dose Rate	Calculated Dose Rate	Detector Distance (30.48 cm in air)
30.48 cm	1.2 mR/h	2. mR/h	83.82 cm in graphite
60.96 cm	70. mR/h	30. mR/h	53.34 cm in graphite
76.20 cm	200. mR/h	120. mR/h	38.10 cm in graphite
91.44 cm	1,200. mR/h	1,120. mR/h	22.86 cm in graphite
99.06 cm	2,600. mR/h	2,380. mR/h	15.24 cm in graphite
106.68 cm	4,500. mR/h*	4,500. mR/h*	7.62 cm in graphite
114.30 cm	5,300. mR/h	5,510. mR/h	No graphite

8. Discussion

In Table 1, the thermal neutron flux values as provided at various locations in the Sigma Pile. In Table 2, the measured values are compared with the calculated values from the MCNP code for a selected set of positions in the Pile. The absolute thermal neutron flux was determined from coincidence counting of gold foils using the cadmium difference method from the measured activity of the bare and cadmium covered foils at each location. A cadmium cutoff energy of 0.5 eV was assumed for the 30 mil cadmium shield.

In Tables 3, the neutron dose rate as calculated by MCNP at the pile locations was compared with measured values from TLD dosimeters and the neutron dose rates agreed to within 5.4%. In Table 4, the neutron dose rates external to the pile as calculated by MCNP were compared with rem-ball measurements and they were in general agreement with the measured values to ≤16%.

In Table 5, the calculated gamma-ray dose rate in the trays of the pile generally agrees with the measured data to within about 4.5%. However, in the case of Table 6, when the Ra-Be source was being withdrawn from the pile, the calculated gamma-ray dose rate disagrees *significantly* with the measured dose rates at those locations where the gamma-rays from the source were traveling a long distance through the graphite and the measurement were being taken at the low end of the range of the instrument. It is not clear where the problem lies in this disagreement.

9. Acknowledgement

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10. References

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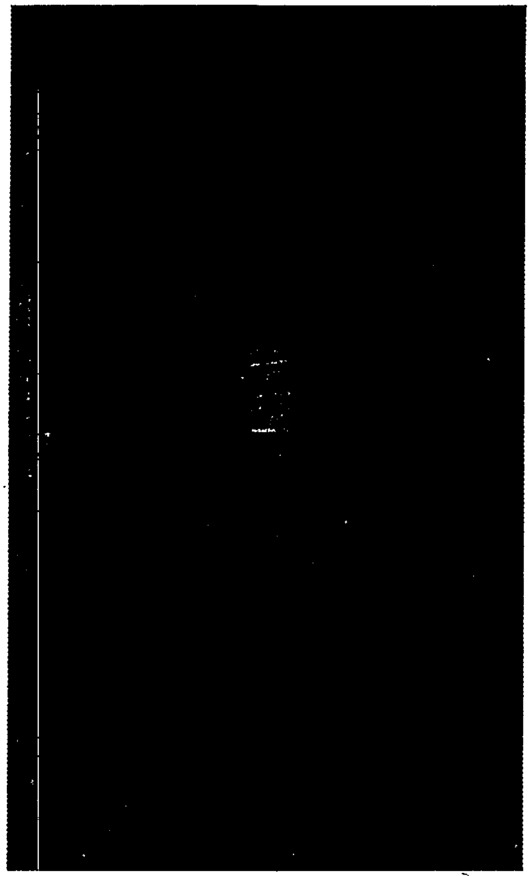

Figure 1. BNL Sigma Pile (Radium-Beryllium Neutron Source)

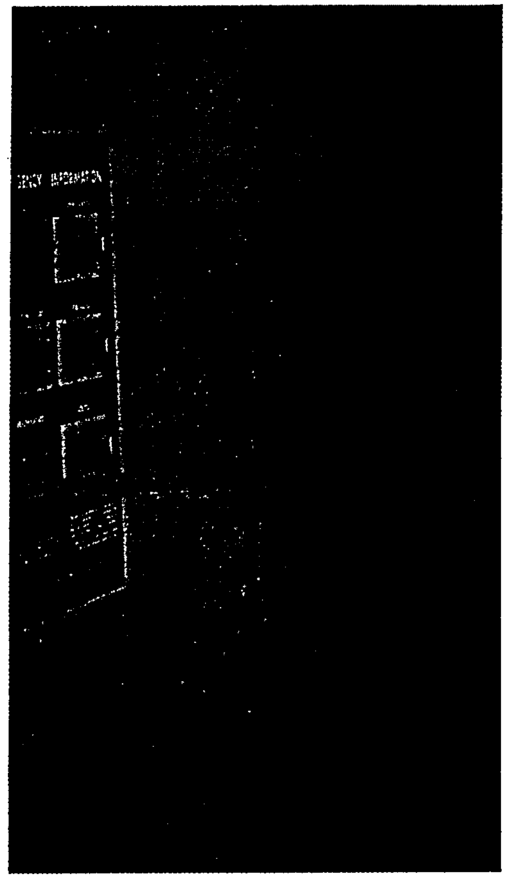

Figure 2. Sigma Pile South Side Sample Trays 3-5 and 3-4 (16 inch apart)